

Field Study of a Chlordane Residue Problem: Soil and Plant Relationships

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In January, 1968, state regulatory officials found evidence of unacceptable levels of what they described as heptachlor epoxide in the milk supply of the Gallatin Valley of Montana. In the following months almost one-third of the dairymen in this milk shed were restrained from selling milk; approximately 1,500 head of cattle were quarantined and some cheese was seized and destroyed.

The situation developed as a result of using chlordane as recommended by the State Entomologist (1) for the control of the adult alfalfa weevil. In a previous article (2) data on the reliability of laboratory analysis and feed-milk residue relationships were presented. This paper will deal with soil-plant relationships.

Residue Location In The Soil

Chlorinated hydrocarbons are not very water soluble and attach themselves readily to organic matter. King (3) reported that when applied as surface spray, heptachlor will be confined largely to the top 1 inch of soil.

As no information relative to the location of chlordane in the soil was available, a number of soil samples were gathered in April 1968. This was about one year after the last chlordane application. Samples were gathered from various depths from fields where the rate of application had been according to the manufacturer's directions, i.e. 1½ to 2 pounds per acre applied prior

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to the plant being 1½ inches tall. Table I shows typical results from some 5 samplings of both new and old alfalfa stands. Hay samples were taken on the same date.

TABLE I

Chlordane Residue In Soil Samples Taken
At Various Depths And Hay Samples
Harvested From Same Fields

(1) New Stand - Sprayed Once - Spring of 1967

Sampled April 1968	Surface Trash	3.75 ppm
	Surface Soil (¼")	0.38 ppm
	Soil ¼" to 2" deep	0.11 ppm
	1st Cutting Hay*	0.46 ppm
	2nd Cutting Hay**	0.17 ppm

(2) Old Stand - Sprayed 4 or 5 Times - Last
Application Spring of 1967

Sampled April 1968	Surface Trash	2.08 ppm
	Surface Soil (¼")	1.32 ppm
	Soil ¼" to 2" deep	0.11 ppm
	1st Cutting Hay*	0.52 ppm
	2nd Cutting Hay**	0.20 ppm

* Harvested in June or July 1967 and stored in an open stack

** Harvested in August or September 1967 and stored in an open stack

These data show the highest concentration in the surface trash, i.e. leaves, stems, etc., of the previous crop. The next highest concentration was in the top ¼ inch of soil and a sharp decline in residue was found in the soil taken at the ¼ to 2 inch level.

Translocation vs Non-translocation

At the time of this incident, translocation of pesticides from the soil to the foliage of a plant was considered unlikely (4,5). Foliage contamination, when not directly sprayed, was considered to come from dust

and dirt accumulation on the plant surface from rain splash, harvesting, etc.

Engle et al. (5) in 1963 reported no translocation of heptachlor to corn, sudan or millet, although translocation to root crops had been demonstrated (6). Further, they (5) showed excessive ground contact to be a factor in increasing the heptachlor residue in alfalfa. King (3) demonstrated translocation of heptachlor to the alfalfa plant in 1966, but Lichtenstein and Schuly (7) showed that certain crops did not absorb measurable amounts of insecticides and the absorption rates for aldrin and heptachlor were different, i.e. $0.88\% \pm .23$ for aldrin and $3.01\% \pm .85$ for heptachlor.

Based on this information relative to translocation and the information on residue location in the soil of the contaminated fields, it did not seem desirable to recommend working the fields to distribute the residue down into the soil. This practice would, presumably, reduce the amount of contamination to the hay crop from dust, dirt and soil contact, but in so doing place more residue in the root absorption area. If translocation were a major factor, then working the soil could result in a greater residue in the plant.

Relationship Of Concentration In The Crown
To Concentration In The Plant

King (3) presented data on the concentration of heptachlor in the crown of the plant and also in the top, middle and bottom of the same plant. For example, where the crown contained 1.78 ppm the top contained 0.20, the middle 0.32 and the bottom 0.38 ppm.

In an attempt to find a method of estimating what the situation might be the season following discontinuance of spraying, we collected and analysed some alfalfa crowns. The results are shown in Table II.

TABLE II
Residual Chlordane In Alfalfa Crowns
(April 1968)

<u>Sample No.</u>	<u>Description</u>	<u>Chlordane PPM</u>
71	Sprayed - 1967 only*	1.01
72	Sprayed - 1966-67*	0.66
73	Sprayed - last 1966*	0.77
75	Sprayed 1966-67*	0.29

* March or April

If the hay contains about 11% of the residue in the crown, as shown by King (3), then we could expect the following concentrations in the hay:

Sample 71 - 0.11
 72 - 0.07
 73 - 0.08
 74 - 0.1089
 75 - 0.0319

These values are comparable to what we found in 1967 hay from fields sprayed in 1966 but not in 1967, as shown in Table III.

TABLE III

Residue In Alfalfa Hay Samples From Fields
 Sprayed In 1966 But Not In 1967

<u>Sample No.</u>	<u>Description</u>	<u>Chlordane Residue-PPM</u>
55	1st Cutting (1967)	0.10
56	2nd Cutting (1967)	0.04
59	1st Cutting (1967)	0.06
F-1	Cutting Unknown	0.08
F-4	1st Cutting (1967)	0.06
F-5	2nd Cutting (1967)	0.06
77	2nd Cutting (1967)	0.101

Disposal of Milk and Manure From Contaminated Cattle

Once cattle become contaminated with insecticide residues the question of how to dispose of contaminated milk and animal wastes arises. Most dairy farms do not have adequate septic tank capacity to dispose of the milk as sewage and manure must be disposed of in some manner.

One producer who was restrained from selling milk in January 1968 put his milk, which was reported to contain 0.9 ppm of residue in the milk fat, into a manure spreader and spread it onto an alfalfa field along with the manure from the same herd. This field, located close to the barns, had not previously been sprayed. The chlordane residues found in soil samples taken from one portion of the field where no manure or milk had been applied and from another portion where manure and milk had been applied is shown in Table IV.

TABLE IV

Influence Of Manure And Milk From A Contaminated
Herd When Disposed Of By Spreading As
Fertilizer On The Pesticide
Residue Of The Soil

<u>Untreated</u>	<u>Chlordane Residue-PPM</u>
a) Surface Trash	0.09
b) Top $\frac{1}{4}$ " Soil	0.03
c) $\frac{1}{4}$ " to $\frac{1}{2}$ " Depth	0.03
<u>Treated (Manure and Milk)</u>	
a) Surface Trash	0.44
b) Top $\frac{1}{4}$ " Soil	0.042
c) $\frac{1}{4}$ " to $\frac{1}{2}$ " Depth	0.02

These data seem to show the contaminated milk and manure did increase the residue level of the surface trash of the field.

Heptachlor Epoxide vs Apparent Heptachlor Epoxide

As indicated (2), one of the first questions asked by scientists was, "how could the level of heptachlor epoxide being reported in the milk be so high if chlordane was the chemical reported to be used and the only significant residue present in the hay?"

In December 1969 the Bureau of Science of FDA (8), using milk samples produced from cattle fed hay sprayed with chlordane, harvested and stored in an open stack and which, upon analysis, shows consistantly the presence of chlordane (2) (see Table V), determined that the milk contaminant which had been reported as heptachlor epoxide was in fact only about 10% heptachlor epoxide and 90% an unidentified compound. Presumably this unidentified portion was some degradation product of technical chlordane which was indistinguishable from heptachlor epoxide by the gas-liquid chromatography technique. (This compound has now been identified as orychlordan, a previously undescribed metabolite of chlordane) (10).

Thus, as pointed out by Bevenue and Beckman (9) in work with toxephene, "The use of gas chromatography data alone as the basis for legal actions is an invitation to criticism and rebuttal".

Summary

We found the highest residue in the organic trash lying on top of the ground of the alfalfa fields and the next highest concentration in the top $\frac{1}{4}$ " of soil. Concentrations dropped sharply at lower depths. Analysis of alfalfa crowns from fields previously sprayed with chlordane as a means of predicting residue levels in the next crop gave results comparable to those reported by King (3) and seem to hold for alfalfa harvested the first season following discontinuance of spraying.

Disposal of milk and manure from a contaminated herd by spreading on a field apparently raises the residue level in the surface trash.

Finally, the reliance on the gas chromatograph alone, even though the analysis of any single sample is done by two or more laboratories, is questionable and an invitation to criticism, as suggested by Bevenue and Beckman (9).

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