Field Study of a Chlordane Residue Problem in Milk

by J. C. Boyd
Animal Science Department, Montana State University
Bozeman, Montana

In January, 1968, State regulatory officials found 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 Quest For Information

Immediately following the issuance of the first restraining order, there were innumerable requests from dairymen for information. Questions asked most frequently were: (1) how can the residue most quickly be eliminated from the milk, (2) where can hay and milk samples be analysed, (3) how reliable are the test results, (4) what level of residue can be fed, (5) what about the soil and the next hay crop, (6) what is the relationship of residue in milk fat to the level in body fat, and (7) what methods are available for determining when quarantined animals are acceptable for slaughter?

In addition, research personnel were puzzled about the level of heptachlor epoxide being reported in the milk when chlordane was the chemical used on the alfalfa fields.

The Recommendation To Use Chlordane

Chlordane had been recommended for use in the control of adult alfalfa weevil by the state entomologist. It was approved for such use by the U. S. Department of Agriculture from 1950 to January 8, 1968 (2), under the

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label which read in part, "make application in spring when the alfalfa is coming out of the ground and is about 1 to 1½ inches tall. Do not apply during blooming period. Do not feed treated forage to dairy animals or animals being finished for slaughter".

This had been interpreted by the state entomologist to mean application prior to the plant being 1½ inches tall did not constitute forage treatment (1). The Pesticide Regulation Division of the U. S. Department of Agriculture subsequently claimed this interpretation was incorrect (3).

Persistence Of The Residue In The Milk

Claborn had shown in 1953 (4) that chlordane (a) accumulated slower, (b) never reached as high a concentration and (c) was depleted faster from the body fat of animals than aldrin or dieldrin.

Table I shows the depletion rate of "apparent" heptachlor epoxide from the milk of some commercial herds in Montana, following the placing of these herds on feed that had been tested and found to be free of chlordane residue.

TABLE I

Depletion Rate of (apparent) Heptachlor Epoxide from Milk (fat) Following Change to Uncontaminated Hay

Producer No.	1st Sample Date	1st Sample PPM	2nd Sample Date	2nd Sample PPM	3rd Sample Date	3rd Sample PPM	Reduction In PPM	Days
1	3-6	0.42	3-26	0.16			0.26	20
2	3-8	0.49	3-26	0.42	4-3	0.28	0.21	26
3	2-20	1.43	3-19	0.59	4-3	0.43	1.00	43
4	2-26	0.60	3-26	0.29			0.31	29
5	3-7	0.32	3-27	0.28			0.04	20
6	2-26	0.33	3-20	0.25			0.08	23
7	2-20	0.57	3-18	0.25			0.32	26

These data were taken from the report of the Montana regulatory agency (5) and show the results on the date samples were taken. More frequent sampling might have shortened the time required to reduce the residue to below the action level of 0.3 ppm in the milk fat.

However, most dairymen who followed the recommendation of feeding only those feeds known to be free of chlordane residue reduced the apparent heptachlor epoxide residue level of their milk (fat) to below the action level in 20 to 30 days. Where the initial level was relatively high (producer No. 3) a somewhat longer period was required.

Reliability Of Laboratory Results

Early in this situation the reliability of laboratory results, all of which were made by gas-liquid chromatographs without confirmation, became a matter of concern.

In an attempt to locate reliable analytical facilities available to individual dairymen, five samples were split and submitted to three different laboratories. One sample was submitted to a fourth laboratory. The results using gas-liquid chromatography without confirmation are shown in Table II.

TABLE II

Results Of Analysis Of Split Samples
By Different Laboratories

Sample No.	Product	PPM Chlordane Laboratory 1	2	3	4	PPM (apparent) Heptachlor Epoxide Laboratory 1	2	3	4
A	Milk (fat)	0.0	0.03	0.02		0.036	0.04	0.03	
B	Milk (fat)	0.153	0.12	0.02	2.54	0.129	0.0	0.16	0.59
C	Milk (fat)	0.0	0.02	0.02		0.0	0.0	0.01	
DD	Hay	0.10	0.0	0.01		0.0012	0.0	0.005	
E	Grass	0.14	0.0	0.01		0.004	0.0	0.005	

These results showed apparently good agreement on Sample A. On sample B laboratory No. 3 was low on the chlordane result (underlined) and laboratory No. 2 was low on the heptachlor epoxide result. Laboratory No. 4 was exceptionally high on both values. Agreement was good on sample C but on sample D and E, laboratory No. 1 was high on the chlordane values.

If the residue (ppm) for each of the first three laboratories is totaled and the variation between laboratories determined by the method of Snedecor (6), we find the mean result for chlordane to be 0.2143 ppm with a standard deviation of ± 0.4001. This is a coefficient of variation of 186%. The mean value for heptachlor epoxide was 0.1400 ± 0.6828, which is a coefficient of variation of 485%.

Stull et al. (7) found similarly high variability in working with DDT residues in milk from individual cows but suggested that pooled herd milk should be less variable. However, in a later report (8), where DDT intake was compared to levels in the milk of a herd of 500 cows, they found wide fluctuations and unexplainably high levels of residue in the milk.

Level of Chlordane Residue Advisable To Be Fed

No information was available relative to what level of chlordane residue might be fed and still keep the residue level in the milk below the action level. This was critically needed since a large amount of hay was being held in hopes it could be fed. We had been advised that possibly 0.02 ppm of residue in the hay might be maximum (9).

Thus, in September, 1968, twelve cows were placed on hay that had been analysed in March and reported to contain 0.25 ppm of chlordane. Feeding continued until December 1968. The hay was sampled by a core sampler, drilled approximately 18" to 20" into the end of each bale as it was fed. Samples from each bale were composited and analysed along with a milk sample, which was composited from the morning and evening milking of the twelve cows -- approximately every seven days for 16 weeks. A normal grain ration was fed, which on periodic sampling consistantly gave negative results for chlordane residue. The results are shown in Table III.

These results show hay sprayed in April or May of 1967, harvested the early part of July and stored in an open stack, contained 0.25 ppm of chlordane residue in March of 1968, based on one sample. Starting in September of that same year and continuing for 16 weeks no sample ever reached 0.25 ppm, the highest level being 0.182. Six weekly samples were negative and the average residue (using 0.01 ppm for the negative values) over the 16 week period was 0.065 ppm. Milk produced from cows fed this hay resulted in no significant change in heptachlor epoxide residue, the average residue being 0.099 ppm on a fat basis. Analysis showed the coeffici-

ent of variation for the hay samples to be 53.7% and for the milk samples 23.16%.

In-as-much as this level of residue in the hay did not produce any appreciable change in the residue in the milk a second group of six cows was started in September 1968 on hay that had tested 18 ppm of chlordane residue in February 1968. This group was handled in the same manner as the one above and continued on this feed for 12 weeks. The results are shown in Table IV.

TABLE III

Level Of Chlordane In Hay And (apparent) Heptachlor Epoxide In Resulting Milk (fat)

Date	Hay Chlordane PPM (as fed)	Milk Heptachlor Epoxide PPM (fat)	
		Cattle on Treatment	Balance of Herd
March 1968	0.25*	-----	-----
8-8-68**	0.092*	-----	.054
8-15-68	Neg.(0.010)***	0.095	
8-22-68	0.122	0.090	
8-29-68	Neg.(0.010)	0.076	
9-5-68	0.177	0.067	
9-12-68	Neg.(0.010)	0.095	
9-20-68	Neg.(0.010)	0.079	.062
9-27-68	Neg.(0.010)	0.085	
10-3-68	Neg.(0.010)	P.Tr.0.010	
10-15-68	0.085	0.056	
10-23-68	0.182	0.141	
10-29-68	0.011	0.149	
11-5-68	0.055	0.155	
11-13-68	0.105	0.097	.087
11-20-68	0.025	0.147	
11-26-68	0.022	0.131	
12-6-68	0.171	0.171	.056
AVERAGE	0.065	0.099	

* Stack samples

** Feeding contaminated hay began

***Residue in hay fed previous period, i.e. 8-8-68 to 8-15-68

It will be noted that the level of chlordane in the hay varied over a wide range but never exceeded 11.9 ppm.

Yet, a portion of the original sample, stored in a closed metal container, still gave essentially that same (18 ppm) value in January 1969. Thus, as previously indicated, there appears to be a significant decrease in the chlordane level when hay is stored in an open stack.

TABLE IV

Level Of Chlordane In Hay And (apparent) Heptachlor Epoxide In The Resulting Milk (fat)

Date	Hay Chlordane PPM (as fed)	Milk Heptachlor Epoxide PPM (fat)	
		Cattle on Treatment	Balance of Herd
9-68	18.0*		
10-9-68**	----	.095	.084
10-15-68	3.729***	1.338	
10-23-68	.476	.271	
10-29-68	1.806	.261	
11-5-68	2.973	.478	
11-13-68	1.405	.614	
11-20-68	.672	1.140	.064
11-26-68	.078	.208	
12-6-68	1.043	.154	
12-12-68	1.080	.340	
12-19-68	8.074	.180	
12-26-68	11.902	.763	.032
1-3-69	8.281	.876	
AVERAGE	3.549	.551	

* Stack sample

** Feeding contaminated hay began

***Residue in hay fed the previous period, i.e. from 10-9-68 to 10-15-68

An analysis of these data show the mean value for the twelve samples of hay to be 3.459 ppm of chlordane, with a minimum level of .078 and a maximum of 11.902 ppm. The standard deviation was ± 2.4433 and the coefficient of variation 70.61%.

For the milk, five of the twelve samples were below the action level and seven above. The mean value was 0.551 with a minimum of 0.180 and a maximum of 1.338. The standard deviation was ± 0.2555 and the coefficient of variation 46.29%.

These two feeding trials would indicate chlordane residue in the hay in excess of 0.065 ppm may be tolerated without the apparent heptachlor epoxide exceeding the action level of 0.3 ppm but that chlordane residues in the hay averaging 3.45 ppm would result in milk exceeding the action level and thus be subject to removal from the market.

Summary and Discussion

This experience seems to point out that the continued registration and approval of the ambiguous language on the label for chlordane by the U. S. Department of Agriculture, Pesticide Regulation Division, particularly after terminating registrations for other organochlorine pesticides on alfalfa, was misfortunate. Our experience supports the work of Claborn of 1953 (4) in that chlordane did not build up to very high levels in the animals and was dissipated relatively quickly once the animals were taken off the contaminated feed. Also, we found a wide range in values reported by different laboratories analysing the same sample. Also, wide ranges in laboratory results when one laboratory follows the feed and milk residues on control feeding trials, as did Stull *et al.* (8).

Based on our results, as well as others (7,8), it is doubtful if the one sample procedure used by the regulatory agency as a basis for legal action on highly perishable products such as milk, where the product cannot be held until analytical results are available, is justifiable. A more realistic approach would seem to be the three out of five procedure used by the U. S. Public Health Service in evaluating other violations of milk production procedures, *i.e.* temperature, standard plate count and coliform count of fluid milk. Applying this procedure to the data in Table IV such milk would not have been removed from the market until November 13, 1968, when three of the last five samples were above the action level of 0.3 ppm in the milk fat. This would be more acceptable to the milk producer than a "single sample" procedure of removing the milk from the market on October 15, 1968, permitting it to again be sold from October 23 to November 5 and then taking it off again on November 5. Following November 5 this milk source, using the three out of five approach, would not again be permitted to be sold for the duration of the period, even though on November 26, December 6 and December 19 the analysis are below the action tolerance. This would provide considerably more assurance that the consumer is getting a safe milk supply.

References

1. PEPPER, J. H., State Entomologist, Montana State University. Information release March 11. (1968).

2. Federal Register. January 8. (1968).

3. HAYS, H. W. USDA, ARS, Pesticide Regulation Division, Personal correspondence. April 29. (1968).

4. CLABORN, H. V., Agric. Chemicals, 8;38. (1953).

5. BALLOU, H., Milk Control Officer, Montana Livestock Sanitary Board. Personal Conference. (1968).

6. SNEDECOR, G. W. and COCHRON, W. G. Statistical Methods. Iowa State University Press. 6th ed. (1967).

7. STULL, J. W., BROWN, W. H., WHITING, F. M. and WITT, J. M. J. Dairy Sci. 49;945. (1966).

8. STULL, J. W., BROWN, W. H., WHITING, F. M. and WHITT, J. M. Environ Contam and Toxicol 3;135. (1968).

9. STREET, J. C. Animal Science Deparrment, Utah State University. Personal Conference, February. (1968).