

entry of the toxicant into the body, as it was found following oral ingestion and subcutaneous injections.

Dieldrin is stored unchanged in the body and is recovered as such from animal products and body tissues.

#### Acknowledgments

The authors gratefully acknowledge the help of D. A. Greenwood, Utah Agricultural Experiment Station, Logan, Utah, R. H. Carter, USDA, Beltsville, Md., R. C. Bushland, USDA, Kerrville, Tex., and T. B. Davich, Tidewater Research Station, Holland, Va., for furnishing the samples used in this study. The authors are especially thankful to R. H. Carter for performing all of the total chlorine analyses reported herein and for his release of the analytical data.

Further acknowledgment is made for the help of F. I. Burleigh, E. R. Johnson, G. E. Pollard, and Pauline A. Baumgartner of the Shell organization with the analyses required to provide the data presented in this paper.

#### Literature Cited

(1) Biddulph, Clyde, Bateman, O. Q., Bryson, J. J., Harris, J. R., Greenwood, D. A., Binns, W., Miner, M. L., Harris, L. E., Madsen, L. L., *Advances in Chem. Ser.*, No. 1, 237 (1950).

- (2) Bushland, R. C., Claborn, H. V., Beckman, H. F., Radeleff, R. D., Wells, R. W., *J. Econ. Entomol.* **43**, 649 (1950).
- (3) Carter, R. H., *Anal. Chem.* **19**, 54 (1947).
- (4) Claborn, H. V., Wells, R. W., *Agr. Chemicals* **7** (1952).
- (5) Condron, C. H., Wells, R. W., Schwardt, H. H., Baker, D. W., Norton, L. B., Hausens, E. J. Bur. Entomol. & Plant Quarantine, Bull. **E-800** (1950).
- (6) Danish, A. A., Lidov, R. E., *Anal. Chem.* **22**, 702 (1950).
- (7) Davidow, B., Radomski, J. L., *J. Pharmacol. Exptl. Therap.* **107**, 259 (1953).
- (8) Davidow, B., Radomski, J. L., Ely, R., *Science* **118**, 383 (1953).
- (9) Diephuis, F., Dunn, C. L., *Montana Agr. Expt. Sta. Bull.* **461** (June 1949).
- (10) Finnegan, J. K., Haag, H. B., Larson, P. S., *Proc. Soc. Exptl. Biol. Med.* **72**, 357 (1949).
- (11) Garhart, M. D., Witmer, E. J., Tajima, Y. A., *Anal. Chem.* **24**, 851 (1952).
- (12) Gowdey, C. W., Stavrazy, G. W., *Can. J. Biochem. Physiol.* **33**, 272 (1955).
- (13) Harris, J. R., Biddulph, C., Greenwood, D. A., Harris, L. E., Bryson, M. J., Binns, W., Miner, M. L., Madsen, L. L., *Arch. Biochem.* **21**, 370 (1949).
- (14) Laug, E. P., Fitzhugh, O. G., *J. Pharmacol. Exptl. Therap.* **87**, 18 (1946).
- (15) Laug, E. P., Nelson, A. A., Fitzhugh, O. G., Kunze, F. M., *Ibid.*, **98**, 268 (1950).
- (16) Lehman, A. J., *Assoc. Food & Drug Officials U. S., Quart. Bull.* **12**, 82 (1948).
- (17) Ludewig, S., Chanutin, A., *Proc. Soc. Exptl. Biol. Med.* **62**, 20 (1946).
- (18) Mattson, A. M., Spillane, J. T., Baker, C., Pearce, G. W., *Anal. Chem.* **25**, 1065 (1953).
- (19) O'Donnell, A. E., Johnson, H. W., Jr., Weiss, F. T., *J. Agr. Food Chem.* **3**, 757 (1955).
- (20) O'Donnell, A. E., Neal, M. M., Weiss, F. T., Bann, J. M., Decino, T. J., Lau, S. C., *J. Agr. Food Chem.* **2**, 573 (1954).
- (21) Princi, F., Spurbeck, G. H., *Arch. Ind. Hyg. Occupational Med.* **3**, 64 (1951).
- (22) Radomski, J. L., Davidow, B., *J. Pharmacol. Exptl. Therap.* **107**, 266 (1953).
- (23) Sun, Y. P., Sun, J. T., *J. Econ. Entomol.* **45**, 26 (1952).
- (24) *Ibid.*, **46**, 927 (1953).
- (25) Treon, J. F., Cleveland, Frank, J., *AGR. FOOD CHEM.* **3**, 402 (1955).
- (26) van Asperen, K., Oppenoorth, F. J., *Nature* **173**, 1000 (1954).
- (27) Von Oettingen, W. F., U. S. Public Health Service, Bull. **414**, 314, 317 (1955).
- (28) Woodward, G., Ofner, R. R., *Federation Proc.* **5**, 215 (1946).

Received for review October 7, 1955. Accepted July 5, 1956.

## PESTICIDE RESIDUES

### Malathion in Milk and Fat from Sprayed Cattle

H. V. CLABORN, R. D. RADELEFF,  
H. F. BECKMAN, and  
G. T. WOODARD

U. S. Department of Agriculture,  
Kerrville, Tex.

Entomological studies have indicated that malathion, S-(1,2-dicarbethoxyethyl)O,O-dimethyl dithiophosphate, is an effective insecticide against livestock pests, and toxicology studies have shown that it may be safely applied to livestock. However, before it could be recommended for use on beef and dairy cows it was necessary to determine whether its use as a spray would cause contamination of meat or milk. Hereford cattle were sprayed 16 times with 0.5% malathion. Fat samples taken 1 week after the last spraying contained no detectable amounts of the insecticide. When dairy cows were sprayed with 0.5 and 1.0% malathion the insecticide was present in all milk samples taken 5 hours after spraying, ranging from 0.08 to 0.36 p.p.m. Only traces were present 24 hours after spraying and samples taken 3 and 7 days after spraying were free of contamination.

**S**MALL-SCALE EXPERIMENTS WITH MALATHION against insects affecting livestock have been made during the past 2 years by entomologists of the Kerrville, Tex., laboratory. Their results have indicated that 0.5% concentration of malathion kills flies, ticks, and lice, but that its residual effect does

not persist as long as that of the chlorinated hydrocarbon insecticides. Veterinarians at Kerrville found that even such susceptible animals as baby calves appeared unharmed by single sprayings of 0.5% malathion (3).

As malathion was indicated to be toxic to a number of livestock insects but not

acutely poisonous to cattle, experiments were conducted in cooperation with the American Cyanamid Co. to investigate some toxicological aspects of its use. One phase of that study was to establish whether repeated sprayings would result in a deposition of the insecticide in the fat of cattle. Another phase was

to determine whether there was any milk contamination from spraying malathion on dairy cows.

### Experimental

Malathion was determined by the colorimetric method described by Norris, Vail, and Averell (2), and modified as follows for the analysis of milk and beef fat by Norris and Fuller (7).

The milk samples were freeze-dried and extracted with carbon tetrachloride. The filtered extract was concentrated to a volume of 200 ml. and washed with a sodium chloride solution to remove interfering substances. The malathion was hydrolyzed with alcohol and sodium hydroxide and the sodium dimethyl dithiophosphate extracted with a cold salt solution. The aqueous solution was acidified and then washed twice with carbon tetrachloride. After the addition of cupric ions, a third extraction with carbon tetrachloride removed the colored copper salt of *O,O*-dimethyl dithiophosphoric acid and the absorption was measured in a 5-cm. cell.

For the experiment on meat contamination eight Hereford cattle were sprayed 16 times at weekly intervals with 0.5% malathion, four of the animals with an emulsion and the other four with a suspension. Samples of fat were taken by biopsy before the first spraying and 1 week after the last spraying. No malathion was found in any of the samples by the above method, which is sensitive to 0.5 p.p.m. on a 20-gram sample.

For the milk-contamination study four Jersey cows were sprayed twice 1 week apart. One cow was sprayed with 0.5% emulsion, another with 1.0% emulsion, a third with 0.5% suspension, and a fourth with 1.0% suspension. Two quarts of spray were used for each cow. The cows were milked by milking machine, with all possible precautions to prevent outside contamination. Before spraying, and 5 hours and 1, 3, and 7 days after each spraying, samples were taken for butterfat and malathion determinations. The milk samples were freeze-dried, most of them immediately after they were taken, and none of them were allowed to stand in the refrigerator more than 24 hours. After drying they were analyzed by the method described above, which is sensitive to 0.02 p.p.m. on a 1-liter sample of milk.

### Results

The results of these determinations are shown in Table I. As there was a wide variation in the butterfat content of the samples and malathion is known to be fat-soluble, better comparisons can be made when the results are adjusted to a uniform butterfat content. Data calculated for milk containing 4% of butterfat are therefore included.

The higher concentration caused

**Table I. Butterfat and Malathion Determinations in Milk from Dairy Cows Sprayed with Malathion**

Formulation, %	Time of Sampling	Butterfat, %	Malathion, P.P.M.		
			Found	Calculated to 4% butterfat	
Emulsion 0.5	Before spraying	5.47	0	0	
	After first spraying				
	5 hours	5.65	0.11	0.08	
	1 day	5.75	0.01	0.01	
	3 days	5.30	0	0	
	7 days	4.77	0	0	
	After second spraying				
	5 hours	8.40	0.17	0.08	
	1 day	4.30	0	0	
	3 days	5.67	0	0	
	7 days	5.60	0	0	
	1.0	Before spraying	4.90	0	0
		After first spraying			
		5 hours	8.00	0.39	0.20
1 day		4.30	0.03	0.03	
3 days		3.70	0	0	
7 days		1.50	0	0	
After second spraying					
5 hours		6.50	0.29	0.18	
1 day		6.12	0.01	0.01	
3 days		4.60	0	0	
7 days		4.80	0	0	
Suspension 0.5		Before spraying	5.00	0	0
		After first spraying			
		5 hours	6.20	0.39	0.27
	1 day	5.80	0.06	0.04	
	3 days	4.80	0	0	
	7 days	3.10	0	0	
	After second spraying				
	5 hours	5.60	0.42	0.30	
	1 day	4.30	0.03	0.03	
	3 days	3.60	0	0	
	7 days	3.60	0	0	
	1.0	Before spraying	4.75	0	0
		After first spraying			
		5 hours	7.30	0.64	0.36
1 day		4.60	0.06	0.06	
3 days		4.60	0	0	
7 days		4.22	0	0	
After second spraying					
5 hours		5.70	0.47	0.33	
1 day		4.50	0.11	0.10	
3 days		4.30	0	0	
7 days		4.30	0	0	

greater residues than the lower concentrations, and the residues from the suspensions were greater than those from the emulsions. The 0.5% suspension caused a greater residue than the 1.0% emulsion. Therefore, emulsions would be preferable for use on dairy cattle.

### Literature Cited

(1) Norris, M. V., Fuller, L. T., Stam-

- ford Research Laboratories, Stamford, Conn., private communication.  
 (2) Norris, M. V., Vail, W. A., Averell, P. R., J. AGR. FOOD CHEM. 2, 570-3 (1954).  
 (3) Radeleff, R. D., Woodard, G. T., Nickerson, W. J., Bushland, R. C., U. S. Dept. Agr., Tech. Bull. 1122 (1955).

Received January 30, 1956. Accepted June 21 1956.