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DCRB 71-4

UNITED STATES DEPARTMENT OF AGRICULTURE
Agricultural Research Service

P-130

SIMILARITY IN BEHAVIOR OF DDE AND POLYCHLORINATED BIPHENYL (Aroclor 1254) RESIDUES IN AN ENVIRONMENTALLY CONTAMINATED HERD OF DAIRY COWS

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Polychlorinated biphenyls (PCB) are industrial compounds that have been manufactured since about 1930. Recently the PCB residues have been detected in the environment. They were first found as interferences in pesticide analysis. The PCB's have many chemical and biological characteristics similar to those of organochlorine pesticides (7). The U. S. Food and Drug Administration has established an actionable level equivalent to 5 ppm PCB in milk fat. One of the PCB's Aroclor 1254[1/], has been used as an ingredient in paints used to coat the interior walls of silos. Feeding of silage from these silos has resulted in residues of Aroclor 1254 in milk.

We have been working with a herd of cows that had been taken off the market because of excessive residues of DDT and related compounds. In the course of this study, PCB contamination was also detected. The purpose of this paper is to compare the behavior of these two contaminants. The primary emphasis is on the reduction in milk concentration of these compounds when the herd is returned to clean feed.

Materials and Methods

The data in this study were obtained from a herd of cows that had been inadvertently contaminated with DDT and DDE. The initial total concentration of residue was about 15 µg/g milk fat. The ratio of DDE to DDT was about 5 to 1 when we first observed the herd.

The herd was removed from the apparent source of DDT contamination about June 15, 1970. When a clean cow was introduced to the herd about July 1, we found other gas chromatographic peaks which were subsequently identified as Aroclor 1254. We also determined that milk from the main herd contained Aroclor 1254. These peaks had originally been assumed to be DDD and DDT.

At this time the cows were being fed corn silage from an upright silo. This silo was later established as a source of PCB contamination. In late July the source of corn silage was switched to a trench silo. It is not possible in retrospect to determine the precise date that this change occurred.

1/ Trademark, Monsanto Chemical Co., St. Louis, Missouri.

Paper 130 presented at the Annual Meeting of the American Dairy Science Association, East Lansing, Michigan, June 1971.

Milk samples from individual cows were obtained at 3-week intervals beginning July 27 and continuing through December 1. Concentrations of DDE in the milk were determined by routine methods in our laboratory (3).

Concentration of PCB was determined by somewhat arbitrary methods. PCB's are a complex mixture of compounds and authentic standards for the individual components of the mixture are not available. This would pose no problem in silage analysis since material recovered was identical to Aroclor 1254 in gas and thin layer chromatographic behavior.

However, many of the peaks of Aroclor 1254 with retention times equal to DDE or less did not appear in significant amounts in milk. Since PCB's are resistent to alkaline dehydrochlorination we treated the samples with alcoholic KOH to convert any DDT or DDD to the corresponding dehydrochlorination products with retention times equal to DDE or less. This eliminated possible interfernces of DDT or DDD with the analysis for PCB. Concentrations of PCB's were calculated as Aroclor 1254 based on the three major peaks with gas chromatographic retention times greater than the retention time of DDE. The column used was 10% DC-200 on 80/100 mesh Gas-Chrom Q and detection was by electron capture.

Two experiments were conducted with groups of these cows at various times in an attempt to accelerate the decline in milk concentration of DDE. In the first experiment sodium phenobarbital and a combination of sodium phenobarbital and activated carbon were tested. In the second, cows were fed additional concentrate for six weeks to test the effect of increased milk production.

Results and Discussion

The uncontaminated cow was introduced to the herd on about July 1. Concentrations of PCB in the milk of this cow are shown for the period before she was introduced to the herd and at weekly intervals until September 7 (Table 1). The cow was sampled less frequently until November 10.

The concentration of PCB rose rapidly after the cow was introduced to the herd. The peak level was reached on July 20th with a small decrease by July 28. We interpret this to indicate that the herd was removed from the contaminated silage sometime during this period. However, the main herd had not been sampled between July 1 and July 27 so that we do not have a reliable estimate of the date.

The concentration of PCB in the milk of this cow declined very rapidly after she was removed from the contaminated silage. At about two weeks after the removal, the rate of decline is much less. When plotted on semilog paper, the concentration curve is remarkably similar to the two-component first-order curves that we have previously published for the reduction of DDE levels in milk after feeding of DDE stopped (3).

The average concentration of PCB in milk fat of the remaining cows declined from 19.3 to 10.3 μg/g from July 27 to August 17. Without intervening points and exact knowledge of when feeding stopped, it is not possible to make estimates of rates for the first component of the decline. The slower second component was the only component of importance after August 17.

Only the rate for the second component was estimated. The rate constants were calculated for individual cows by least squares using the linear form of the first-order equation:

$$\ln C = \ln C_o - kt$$

where C is the concentration at any time, C_o is the initial concentration, k is the rate constant, and t is time in days.

The first order rate constants calculated for the individual cows of the herd are summarized in Table 2. The cows have been separated into three broad groups. The mid-lactation cows are the cows that were milking at the start of the study on August 17 and were still milking on December 1. Late lactation cows are cows that were milking at the start but went dry before December 1, while early lactation cows are those cows that were dry on August 17 but freshened before December 1. With mid-lactation cows the rate constants were calculated using 6 points whereas with the early or late lactation cows a minimum of 3 points were used and more if they were available.

The overall average rate constant for DDE is very similar to the previously studied published rate constant for DDE (3). The average rate constants for PCB, both overall and within groups, were practically identical to the averages for DDE. The cows in late lactation were similar to the cows in mid-lactation, but the cows in early lactation had lower rate constants than the other two groups. The lower rate for early lactation is consistent with some earlier published work on DDT (2, 6).

There was some variation between compounds within a given cow. However, we did not find a significant difference when the constants were tested statistically (8). The overall correlation between the rate constants for the two compounds was 0.82.

The usefulness of phenobarbital alone or in combination with activated carbon [2/] in accelerating the reduction in milk concentration of DDE and PCB was tested using three groups of cows. This study was carried out for 6 weeks from July 27 to September 7. The cows may have been receiving contaminated silage when the study was started. If they were, it should have increased the opportunity of observing a possible effect from activated carbon.

Neither phenobarbital nor phenobarbital in combination with activated carbon had an effect on the relative reduction in concentration of either compound (Table 3). We have previously reported a small effect of phenobarbital on DDE but we did not consider it of practical significance (5). The absence of an effect here is consistent with that interpretation.

The failure of activated carbon, even in combination with phenobarbital, to affect the reduction of DDE and PCB concentrations is consistent with our previous interpretation of laboratory experiments (4). Activated carbon has been recommended for accelerating the return of pesticide contaminated cows to the market (1). The studies to support the recommendation have involved

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2/ Darco S-51, Atlas Chemical Industries, Wilmington, Delaware.

dieldrin contaminated cows and the activated carbon was used in combination with phenobarbital. Phenobarbital alone is effective with dieldrin (5). Thus, there has been no evidence to demonstrate that activated carbon speeds up the elimination of stored pesticides from the cow, and its use for that purpose is unwarrented.

The cows had been fed little concentrate after they were removed from the market and milk production was low. An increased level of concentrate feeding was imposed in an attempt to speed up pesticide elimination by increasing production. A group of cows was fed an additional 3 kg of concentrate per day for six weeks. A comparable group of cows was used as a control.

The results are presented in Table 4. The extra concentrate increased milk production at 3 weeks and reduced the decline in production over the six-week period. However, the milk concentration of DDE and PCB was not affected.

Milk is a major route of organochlorine pesticide elimination in cows. One would expect the total amount of compound excreted to be increased by increased milk production and a more rapid decline in milk contamination. However, we were not able to demonstrate it in this relatively short-term study. The study had to be terminated at the end of six weeks because many of the cows were producing marketable milk and it was no longer practical to have a control group with low concentrate feeding.

Conclusions

In this field study of contamination by DDE and Aroclor 1254, we observed nearly identical behavior of these compounds in cows. This has important implications in view of the small amount of inforamtion on the behavior of PCB cows. It is probably a first good approximation to assume that the behavior of PCB is quite similar to DDE. If a situation occurs in which a dairyman has PCB contamination, one can use information on the more thoroughly studied DDE to make recommendations until more information on PCB is collected.

The similarity of the two compounds also suggests that one can use previous knowledge of the behavior of DDE in order to properly design the most efficient studies for establishing the parameters of PCB deposition in body fat and excretion in milk.

Acknowledgement

We thank Mr. Ray Brannon, County Extension Agent, Martinsburg, West Virginia, for assistance in carrying out the field portion of this study.

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Table 1. PCB concentration in milk fat of a cow introduced to and removed from contaminated silage [1/]

Date	PCB
	(µg/g)
6/22	<1.0
7/2	1.2
7/8	12.1
7/20	12.6
7/27	9.0
8/5	5.8
8/12	4.2
8/18	4.4
8/26	3.5
9/2	3.7
9/7	3.2
9/29	3.1
10/21	2.4
11/10	2.1

1/ Introduction was about 7/1 and removal was about 7/25.

Table 2. Average rates of decline of DDE and PCB concentration in milk fat of cows in various stages of lactation [1/]

Group	Cows	DDE	PCB
	(No.)	(day^{-1})	(day^{-1})
Mid lactation [2/]	16	0.0145	0.0147
Early lactation [3/]	9	0.0107	0.0106
Late lactation [4/]	6	0.0147	0.0140
All	31	0.0134	0.0134

1/ The values are for the constant, k, in the linear form of the first-order equation: $\ln C = \ln C_o - kt$

where C is the milk fat concentration, C_o is the initial concentration, and t is time in days. Fitted by least squares.

2/ Calculated for the period 8/17 to 12/1 from six measurements.

3/ Calculated from calving to 12/1. Includes at least three measurements.

4/ Calculated from 8/17 until dry. Includes at least three measurements.

Table 3. Effect of phenobarbital and phenobarbital with activated carbon on the concentration of DDE and PCB in milk fat [1/]

Treatment	Cows	Concentration		Final / Initial	S.D.
		Initial	Final		
	(No.)	(µg/g)	(µg/g)		
			DDE		
Control	5	11.6	5.2	0.45	±0.05
Phenobarbital	10	9.5	4.3	0.45	±0.15
Phenobarbital + Carbon	6	8.0	3.7	0.46	±0.20
			PCB		
Control	5	19.3	7.4	0.38	±0.08
Phenobarbital	10	17.7	7.0	0.40	±0.11
Phenobarbital + Carbon	6	22.3	8.5	0.37	±0.14

1/ Observation period was 6 weeks. Phenobarbital was fed at 5 grams per day for the first three weeks and activated carbon at one kilogram per day for the 6 weeks.

Table 4. Effect of feeding extra concentrate on milk fat concentrations of DDE and PCB [1/]

Item	Weeks			Final / Initial	S.D.
	0	3	6		
				Control	
DDE (µg/g)	2.38	1.87	1.54	0.64	±0.08
PCB (µg/g)	4.13	3.38	2.79	0.66	±0.09
Milk (kg/day)	9.93	9.94	8.39	0.86	±0.09
				Concentrate	
DDE (µg/g)	3.61	2.71	2.37	0.65	±0.13
PCB (µg/g)	7.17	5.28	4.50	0.64	±0.18
Milk (kg/day)	12.81	14.98	12.22	0.97	±0.15

1/ Five cows per group. Control cows received roughage ad lib and 3 kg concentrate. Treated cows received an additional 3 kg concentrate.