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Aldrin, heptachlor and β-hexachlorocyclohexane to dairy cows at three oral dosages. 1. Residues in milk and body fat of cows early and late in lactation

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Summary

This study was done to investigate the transfer of aldrin, heptachlor and β-hexachlorocyclohexane (β-HCH) from feed to milk and body fat of dairy cows. Nine cows early and nine cows late in lactation were assigned to groups of three cows each and given different daily dosages of a mixture of pesticides: 1 mg, 2 mg or 4 mg each of aldrin, heptachlor and β-HCH per cow daily for a period of 4 weeks. Milk fat and body fat samples were taken and analysed for pesticide content.

Aldrin and heptachlor could not be detected in milk fat nor in body fat. Average contents of β-heptachlorepoxide were below 0.1 mg/kg even for the highest oral dosage.

However, β-HCH and dieldrin contents in milk fat increased rapidly in the dosing period. The ratio of contents between the different groups roughly corresponded to that between the dosages.

The cows early in lactation did not have less β-HCH and dieldrin than the cows late in lactation. During the first 14 days of the final period the contents of β-HCH and dieldrin usually decreased by more than half. The average transfer coefficients for cows late in lactation were all below 10 % and for cows early in lactation about three times as high because yield of milk fat was nearly three times as high for cows early in lactation.

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Table 1. Transfer coefficients (%) for organochlor pesticides from feed to milk reported in the literature.

Pesticide	Ely et al. (1952)[1]	Ely et al. (1955)[1]	Williams et al. (1964)[1]	Demott et al. (1966)[1]	Fries et al. (1969)	M. Hascoet (pers. commun., 1976)	van den Hoek et al. (1975)
HCB	—	—	—	—	—	—	25.8
α-HCH	—	—	—	—	—	17.6	12.2
β-HCH	—	—	—	—	—	—	31.2
γ-HCH	—	—	2.4	—	—	1.98	2.7
α-Hepo	—	—		—	—	—	13.8
β-Hepo	—	—	32.6	22.0	—	39.6	—
Dieldrin	—	—	21.3	—	—	29.3	26.6
Aldrin	—	—	—	—	—	—	—
Heptachlor	—	2.8	—	—	—	—	—
DDE	—	—	—	—	25.8	—	—
DDD	—	—	—	—	7.6	—	—
DDT	7.0	—	2.4	—	5.1	—	—
Endrin	—	—	4.8	—	—	—	—

[1] Investigated at several dosage levels.

The pesticide contents in body fat too increased but did not reach the same level as in milk fat. Cows early in lactation usually had higher contents in body fat than cows late in lactation. Finally, an estimate of the average relative mass of β-HCH and dieldrin in milk fat and body fat is given.

Introduction

Various workers have investigated transfer of organochlorine pesticides from feed into milk and body fat of dairy cows. Table 1 lists the highest values for the transfer coefficients of different pesticides, whereby the transfer coefficient to milk is defined as the proportion of oral intake recovered in milk. The results of these transfer studies can be used to establish tolerable content of pesticides in feedstuffs. The maximum tolerable content of pesticide in the feed on dry basis can be calculated from tolerable content of pesticide in milk fat, provided that there is a constant relationship between the amount of a pesticide in the daily ration and the amount of this pesticide or metabolite excreted into the daily milk. The tolerable content of pesticide in the daily feed is given by:

$$\mathrm{w\ (tol., feed)} = \frac{\mathrm{w\ (tol., milk)} \times \mathrm{m\ (daily\ milk\ fat)}}{\mathrm{a} \times \mathrm{b} \times \mathrm{m\ (daily\ feed)}}$$

where: w (tol.) is tolerable mass fraction or content of pesticide
m is mass or weight produced or consumed
a is transfer coefficient, and
b is a safety factor.

As the transfer coefficient may be influenced by various factors (van den Hoek et al., 1975), it is advisable to introduce a safety factor. The present study was done to investigate the influence of dosage, stage of lactation and daily yield of milk fat on transfer of aldrin, heptachlor and β-hexachlorocyclohexane (β-HCH) from feed to milk and body fat.

Material and methods

Nine cows early in lactation and nine cows late in lactation were assigned to three groups of three cows each at Hoorn. The groups were given different daily dosages of a mixture of pesticides: 1 mg, 2 mg or 4 mg each of aldrin, heptachlor and β-HCH per cow daily. The pesticides were dissolved in acetone and pipetted into hollowed-out pieces of cattle cake. The acetone was allowed to evaporate. Details of the experimental cows, of the rations fed and of the pesticides given are summarized in Table 2. The total experimental period comprised a preliminary period of two weeks without pesticide to estimate background levels, a dosing period of 4 weeks and a final period without pesticide feeding of 4 weeks. This final period was only 3 weeks for the cows late in lactation.

Every week 3-day composite milk samples were collected for estimation of fat content (Gerber method). Milk yield was recorded daily. One-day milk samples

Table 2. Summary of experimental conditions for each cow early or late in lactation.

	Early	Late
Age (mean and range, years)	7 (5–8)	7 (4–10)
Body weight (mean and range, kg)	554 (512–597)	585 (502–667)
Month of lactation	2nd	10th
Daily milk production (kg)	23.1	6.3
Daily milk fat production (g)	938	314
Daily intake hay (kg)	7	8
Daily intake concentrate (kg)	9	5
Daily dosage (mg of each pesticide)		
Group 1	1	1
Group 2	2	2
Group 3	4	4
Dosing period (weeks)	4	4

were collected twice a week, stored in plastic bottles and frozen at —25 °C until pesticide analysis.

Samples of body fat from the flank area were taken at the end of the preliminary, dosing and final period and were also stored at —25 °C until analysis.

Composite feed samples were also collected and analysed. Contents of pesticides in feed, milk and biopsy samples were estimated by electron-capture gas-chromatography after extraction and cleaning up at the Government Dairy Station, Leyden. Methods of estimation were largely as previously described (van den Hoek et al., 1975). Recoveries for pesticides from milk and body fat are about 95 % or more, so no correction was made for losses.

The detection limits for aldrin, heptachlor, β-HCH, dieldrin and β-heptachlorepoxide in the feed were less than 0.01 mg/kg. In milk fat and body fat, the detection limits were 0.01 mg/kg for aldrin and heptachlor and 0.02 mg/kg for dieldrin, β-heptachlorepoxide and β-HCH.

Results and discussion

During the whole experimental period no organochlor pesticides were detected in the hay and concentrates so that the rations can be taken as pesticide-free. Therefore the presence of pesticides in milk and body fat can only be due to oral administration.

In the whole experimental period, aldrin and heptachlor could not be detected in milk fat nor in body fat.

Average concentrations of β-HCH, dieldrin and β-heptachlorepoxide per group of 3 cows are presented in Fig. 1-3. Table 3 (A and B) gives the yield of milk fat and content of pesticides at the end of the dosing period for each cow and each dosage.

Pesticide levels at the end of the dosing period cannot be assumed to refer to equilibrium conditions. For this evaluation, the dosing period of 4 weeks is probably

Table 3. Average daily yield of milk fat; content of pesticides or their metabolites in milk fat at the end of the dosing period; transfer coefficients, based on the highest content of pesticides found in milk fat in the last week of the dosing period. A. Cows early in lactation.

Group	Cow no	Av. daily fat	Content in milk fat (mg/kg)[1]			Transfer coefficients (%)			
		yield (g)	β-HCH	β-hepo	dieldrin	β-HCH	heptachlor	aldrin	average
1	5	938	0.36	0.028	0.33	33.8	2.6	31.0	22.4
	7	1170	0.28	0.036	0.25	32.8	4.2	29.3	22.1
	77	620	0.37	<0.020	0.25	22.9	<1.2	19.2	14.4
Average		909	0.34	0.028	0.28	29.8	2.7	26.5	19.7
2	14	873	0.64	0.072	0.48	29.2	3.6	23.6	18.8
	18	713	0.68	0.057	0.49	24.2	2.1	17.2	14.5
	58	1054	0.68	0.081	0.48	35.8	4.3	25.3	21.8
Average		880	0.67	0.070	0.48	29.7	3.3	22.0	18.3
3	34	1010	1.19	0.094	0.83	30.0	2.4	21.0	17.8
	41	899	1.19	0.064	0.78	26.7	1.5	24.7	17.6
	73	825	1.17	0.130	0.80	24.1	2.7	16.5	14.4
Average		911	1.18	0.096	0.80	26.9	2.2	20.7	16.6
Standard deviations between cows for all three groups			0.031	0.012	0.033	5.13	1.14	5.03	3.54

[1] Heptachlor and aldrin were not detectable.

Table 3. Average daily yield of milk fat; content of pesticides or their metabolites in milk fat at the end of the dosing period; transfer coefficients, based on the highest content of pesticides found in milk fat in the last week of the dosing period. B. Cows late in lactation.

Group	Cow No	Av. daily fat yield (g)	Content in milk fat (mg/kg)[1]			Transfer coefficients (%)			
			β-HCH	β-hepo	dieldrin	β-HCH	heptachlor	aldrin	average
1	1	444	0.33	<0.020	0.25	14.7	<0.9	11.1	8.9
	46	309	0.28	<0.020	0.21	9.0	<0.6	6.8	5.5
	67	193	0.24	<0.020	0.20	5.6	<0.4	4.4	3.5
Average		315	0.28	<0.020	0.22	9.8	<0.6	7.4	5.9
2	25	350	0.58	0.020	0.50	10.2	0.4	8.8	6.5
	26	190	0.63	0.045	0.55	6.0	0.4	5.3	3.9
	57	274	0.52	0.033	0.46	7.1	0.5	7.9	5.2
Average		272	0.58	0.033	0.50	7.8	0.4	7.3	5.2
3	16	350	1.04	<0.020	0.89	9.3	<0.2	7.8	5.8
	84	308	1.13	0.084	1.00	8.7	0.6	7.7	5.7
	96	178	0.99	0.073	1.08	5.1	0.3	4.8	3.4
Average		279	1.05	<0.059	0.99	7.7	<0.4	6.8	5.0
Standard deviation between cows for all three groups			0.058	0.011	0.063	3.22	—	2.43	1.91

[1] Heptachlor and aldrin were undetectable.

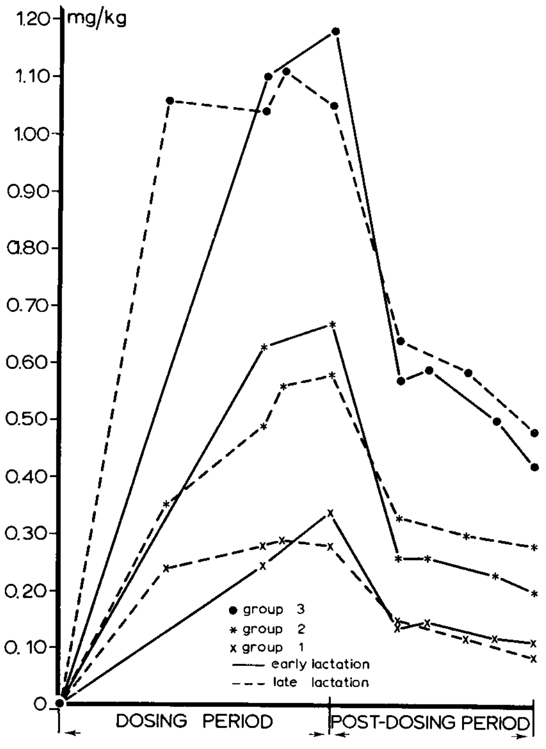

Fig. 1. Average content of β-HCH in milk fat of cows given three levels.

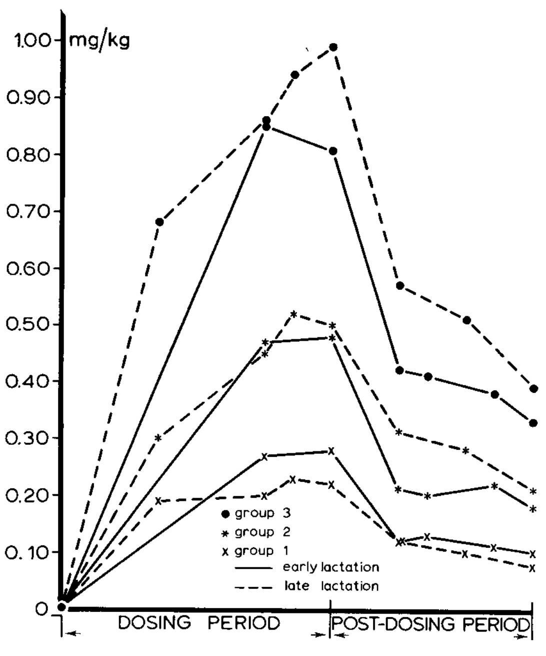

Fig. 2. Average content of dieldrin in milk fat of cows given three levels.

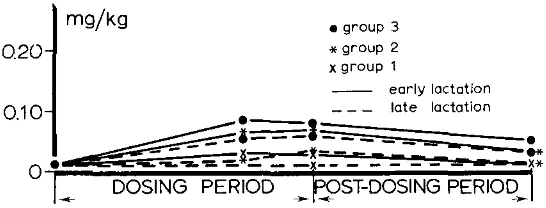

Fig. 3. Average content of β-heptachlorepoxide in milk fat of dairy cows given three levels.

too short. However, van den Hoek et al. (1975) stated that equilibrium for β-HCH, dieldrin and α-heptachlorepoxide is approached about 20-25 days after dosing began. For similar and other organochlor pesticides steady-state conditions are approached after 40-60 days (Fries et al., 1969, 1973; Fries & Marrow, 1976). In the dosing period of 4 weeks the β-HCH content in milk fat rapidly increased initially and thereafter sometimes levelled off near plateau level at the end of the dosing period (Fig. 1, late in lactation groups 1 and 3). The results for dieldrin are essentially identical to those for β-HCH (Fig. 2). However, the dieldrin content at the end of the dosing period is somewhat lower than for β-HCH. For β-heptachlorepoxide, contents for both types of cows were below 0.1 mg/kg even for the highest dosage at the end of the dosing period (Fig. 3). The transfer of heptachlor from feed into milk is not marked. The ratio of the average β-HCH and dieldrin contents in milk fat at the end of the dosing period between the three different groups roughly corresponds to that between the dosages.

The cows early in lactation had somewhat more β-HCH than cows late in lactation, but not significantly at $\alpha = 0.01$. Neither was the difference significant ($\alpha = 0.01$) between the average dieldrin contents for group 3 of cows early and late in lactation (Fig. 2). The decline for β-HCH in the first week of the final period seems a little faster for cows early in lactation, especially at higher dosages.

The calculated transfer coefficients are based on the highest pesticide contents in milk fat and the average milk fat yields, both found in the last week of the dosing period (Table 3). The average values per group for cows late in lactation were all below 10 %. The differences between groups were small, but the differences between cows within a group show the opposite, e.g. the transfer coefficients for β-HCH for cows of Group 1 range from 5.6 to 14.7, mainly because of variation in yield of milk fat rather than of variation in β-HCH content of milk fat.

Transfer coefficients differed markedly also between cows early and late in lactation. The average values for cows early in lactation were about three times those for cows late in lactation because yield of milk fat was nearly three times as high for cows early in lactation. When we define the accumulation coefficient as the content of a pesticide or metabolite in milk fat divided by its content in the feed based on 87 % dry matter, this accumulation coefficient for β-HCH would range 3.1-5.9. The dairy farmer must therefore keep total content in diet less than a sixth of the β-HCH tolerance level in milk fat. To provide a margin of safety, one should

Table 4. Accumulation coefficients for cows early and late (in parenthesis) in lactation.

Group	Cow No	Accumulation coefficients		
		β-HCH	heptachlor	aldrin
1	5 (1)	5.76 (4.29)	0.45 (<0.26)	5.28 (3.25)
	7 (46)	4.48 (3.64)	0.58(<0.26)	4.0 (2.73)
	77 (67)	5.92 (3.12)	<0.32 (< 0.26)	4.0 (2.60)
Average		5.44 (3.64)	0.45 (<0.26)	4.48 (2.86)
2	14 (25)	5.12 (3.77)	0.58 (0.13)	3.84 (3.25)
	18 (26)	5.44 (4.10)	0.46 (0.29)	3.92 (3.58)
	58 (57)	5.44 (3.38)	0.65 (0.21)	3.84 (2.99)
Average		5.36 (3.77)	0.56 (0.21)	3.84 (3.25)
3	34 (16)	4.76 (3.38)	0.38 (<0.06)	3.32 (2.89)
	41 (84)	4.76 (3.67)	0.26 (0.27)	3.12 (3.25)
	73 (96)	4.68 (3.22)	0.52 (0.24)	3.20 (3.51)
Average		4.72 (3.41)	0.38 (0.19)	3.20 (3.22)

aim at dietary content of less than a tenth of the tolerable content of β-HCH in milk fat. Despite a slightly lower accumulation coefficient for dieldrin, the same holds for this pesticide.

The individual accumulation coefficients (Table 4) were calculated from the contents of pesticides in milk fat in Table 3 and from the content in feed in Table 2. The accumulation coefficients varied considerable less than the transfer coefficients, which were clearly influenced by the stage of lactation, which in turn governed the milk fat production. A high milk fat production was not associated with a proportionally lower content of pesticide.

To calculate transfer coefficients, necessary for the calculation of pesticide tolerance in the daily feed, we must use cows early in lactation, which produce large amounts of milk fat.

The highest transfer coefficients observed in our trials (Table 3) did not differ much from those in other studies (Table 1).

When dosage was discontinued, the content of β-HCH and dieldrin declined rapidly in the first 7 days and thereafter much slower. The decline in pesticide content can be described by a two-component first-order system (Fries et al., 1969, 1973). In our study, the estimate of rate constants was practically impossible because of the small number of points in the first weeks of the final period and because of the deviation from linearity when pesticide contents were transformed to logarithmic scale. The contents of β-HCH and dieldrin usually decreased by more than half during the first 14 days of the final period (Fig. 1 and 2). The average contents in body fat of β-HCH and dieldrin at the end of the preliminary period were identical with those in milk fat at the same time. At the end of the dosing period, the pesticide content in body fat increased but did not reach the levels in milk fat (Table 5). The

Table 5. Average contents of β-HCH and dieldrin in body fat, at the end of the dosing period and final period expressed relative to average contents in milk fat at the end of the dosing period.

Stage of trial and of lactation	β-HCH			Dieldrin		
	1	2	3	1	2	3
Dosing period						
early in lactation	0.50	0.61	0.52	0.57	0.73	0.77
late in lactation	0.28	0.48	0.52	0.38	0.48	0.44
Final period						
early in lactation	0.35	0.28	0.34	0.50	0.50	0.59

contents of β-heptachlorepoxide were low and about equal to those in milk fat. They are not tabulated.

Cows early in lactation usually had higher contents of pesticides in body fat than cows late in lactation, although they produced more milk fat and secreted more of the pesticides. Presumably, the higher contents in body fat were associated with a smaller amount of body fat which could take up the pesticide. These cows were fed below standard for milk production. At the end of the final period, pesticide contents decreased but not so far as in milk fat. To estimate the total retention of pesticides in body fat, we need to know both the concentration in body fat and the total amount of body fat. The total amount of body fat cannot be measured exactly. Although the β-HCH and dieldrin contents at the end of the dosing period were less in body fat than in milk fat, one can expect larger amounts of pesticides in body fat (Table 6).

Table 6. Average relative mass of β-HCH and dieldrin secreted in milk fat or accumulated in body fat[1] in the whole dosing period, to that administered.

Group	β-HCH		Dieldrin	
	milk fat	body fat	milk fat	body fat
Early in lactation				
1	0.16	0.21	0.16	0.20
2	0.19	0.26	0.14	0.22
3	0.17	0.19	0.13	0.20
Late in lactation				
1	0.08	0.23	0.06	0.23
2	0.05	0.40	0.05	0.35
3	0.07	0.39	0.05	0.32

[1] Calculation based on an estimate of 80 kg body fat for cows late in lactation and 40 kg body fat for cows early in lactation (P.L. Bergström, pers. commun., 1976) and on the assumption that pesticide content in fat of biopsy sample is representative for total body fat.

A considerable part of the oral intake, more than half, was not recovered. It was probably lost as unabsorbed pesticides or metabolites in faeces or as metabolized pesticides in urine.

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