INSECTICIDE RESIDUES IN MILK AND MILK PRODUCTS[1]

II. Insecticide Residues in Milk from Dairy Cattle Fed Treated Crops

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INTRODUCTION

Chlorinated hydrocarbon and organic phosphate insecticides are commonly used by farmers to control insect infestations of various feed crops during the growing season. Residues of insecticides may be present on such feeds (hay, corn, pea vines, etc.) and hence consumed by dairy cattle. This paper will attempt to summarize information on insecticide residues in milk from dairy cattle which have ingested such treated crops. Information on insecticide residues in milk from the treatment of dairy cattle and barns has been summarized in a previous paper (*26*). Another paper will summarize information on insecticide residues in dairy products and associated problems (*25*).

CHLORINATED HYDROCARBON INSECTICIDES

DDT

The presence of DDT residues in milk from dairy cows who ingested feeds treated with the insecticide was reported in 1947 (*4*, *31*). Carter (*4*) found 1.5 to 25 p.p.m. and Schechter, *et al.* (*31*)reported the presence of three to 26 p.p.m. of DDT in the milk obtained from cows which were on a diet of treated feed.

Shepherd, *et al.* (*32*) aerosol treated two plots of alfalfa with 0.6 and 2.4 pounds of DDT per acre respectively. Hay was made from the treated alfalfa and fed to dairy cows daily at the rate of 1.5 pounds per 100 pounds of body weight if it was sprayed with the low concentration of DDT or one pound per 100 pounds of body weight if sprayed with the high concentration. Milk from cattle receiving the former contained a maximum of 0.9 mg. of DDT per g. while that from the cows receiving the latter contained up to 10.1 mg. per g. These investigators found the insecticide in milk three days after the hay feeding had begun and also 160 to 170 days after feeding was discontinued if hay with the higher concentration was fed. Persistence of the insecticide in the milk dropped to 30 to 40 days when the low concentration was fed. From 5 to 30 per cent of the DDT ingested by the cow was recovered in its milk.

Alfalfa was sprayed with 0.25 pounds DDT per acre ten days before cutting in experiments reported by Smith *et al.* (*33*). The resulting hay contained between seven and eight p.p.m. of residual DDT. When it was fed to cows, DDT appeared in milk at the rate of 2.3 to three p.p.m. Later one half of the hay was replaced with untreated hay and the DDT content of the milk dropped to two thirds of the former value.

Wilson, *et al.* (*37*) studied DDT residues in silage made from dusted pea vines. It was estimated that about one pound of DDT was applied to six tons of vines during the dusting operation, however, less than one p.p.m. was found in the silage at feeding time. No DDT was found in milk produced by cattle fed this silage.When cows were fed pea vine silage to which one pound of DDT per ton was added at the time of ensiling, 15 p.p.m. of the insecticide appeared in the milk.

These experiments were further discussed when an accumulation of DDT in certain body tissues was reported (*2*). It was observed that 3.8 p.p.m. accumulated in muscle tissue, 6.1 p.p.m. in the liver and 221 p.p.m. in depot fat. Calves which received milk from cows fed DDT-containing pea vines had 3.1, 6.2 and 305 p.p.m. in similar tissues, respectively.

Carter *et al.* (*6*) reported the presence of less than 0.5 p.p.m. DDT in milk from cows fed silage made from pea vines sprayed previously with 0.4 to 0.5 pounds of insecticide per acre.

Silage was made from sweet corn which was dusted with 20 to 35 pounds of a five per cent DDT powder per acre either two or four times during the growing season (*21*). Milk produced by cows which received this silage contained 0.1 to 0.5 p.p.m. of residual DDT.

When lush pastures were treated with two pounds of DDT per acre and grazed immediately, a "sweetish" taint appeared in the milk (*27*). This was associated with an increase in DDT concentration.

DDT was fed directly to dairy cattle by Wingo and Crisler (*38*). They found that cows which received either 25 doses of 20 g. each or 21 doses of five g. each, showed signs of stiffness and excitability. Milk from

[1] Second in a series of three review articles on this subject which will appear in this Journal.

the treated animals was toxic to houseflies for eight to nine days after the final dose of insecticide had been administered. Similar experiments were carried out by Ely *et al.* (*13*) and they found: (*a*) higher concentrations of DDT in milk when cattle consumed it as a residue on feed than when the insecticide was fed directly; (*b*) there were no consistent differences in DDT-residues in milk when it was fed mixed with soybean oil or in the crystalline form; and (*c*) feeding DDT in the form of capsules or mixing with grain had no effect on the concentration present in milk.

Alekseeva (*1*) reported the presence of DDT in milk drawn from cows 42 to 45 days after they were treated with doses 35 to 40 times higher than normally would be ingested.

Telford (*36*) noted the presence of DDT in milk from goats which were fed either 0.68 or 1.25 gm. of DDT per pound of body weight.

The accumulation of DDT in soils as a result of spraying practices has been observed by Ginsburg (*17*) and Lichtenstein (*23*). Larger quantities of DDT were found in soils of apple orchards and smaller amounts in soils on which potatoes were grown (*17*). The DDT was present in the soil layer which correspond to plowing and cultivation depths. Lichtenstein (*61*) reported that the upper six inches of soil from turf plots contained 11 to 18 per cent of a single DDT treatment applied ten years earlier. Crop soils showed an accumulation of 15.5 per cent of the DDT applied during a ten year period. Such insecticide accumulations could be additional sources for milk contamination.

Methoxychlor

Four plots of alfalfa were sprayed with 0.48, 0.5, 1.0 and 1.94 pounds per acre of methoxychlor (*15*). When hay from these plots was fed to dairy cows, no methoxychlor could be detected in the milk produced. Cows were fed crystalline methoxychlor in soybean oil and it was noted that 100 times as much methoxychlor as DDT had to be consumed by cows before detectable amounts of the insecticide appeared in milk. Ely, *et al.* (*15*) believed that methoxychlor would not be excreted in the milk by cows which were fed forages sprayed with recommended concentrations of the insecticide.

Benzene Hexachloride and Lindane

Dry, closely grazed pastures were treated with two pounds of lindane or technical benzene hexachloride (mixed isomers) per acre (*27*). Cows were permitted to graze the pastures three weeks after treatment. Milk, cream and butter produced by cows on the lindane treated pasture were free from taints although the butter contained 4 to 6 p.p.m. of BHC. An "earthy" or "musty" taint was noticed in milk, cream and butter produced by cows on the BHC treated pasture. The butter, in this instance, contained 16 to 18 p.p.m. of BHC. When similar treatments were applied to long, wet pastures which were grazed immediately, marked taints and high levels of insecticides were found in the resulting milk, cream and butter.

Lindquist and Donaldson (*24*) reported an instance where benzene hexachloride-treated potatoes were fed to dairy cattle. An off flavor similar to that found in the potatoes was noticed in milk produced by these cows.

Dieldrin

Alfalfa was treated with one and four ounces of Dieldrin per acre (*18, 34*). The alfalfa was cut seven days later and stored as hay for four months after which it was fed to dairy cattle for 112 days. Weekly milk samples were tested and dieldrin was found in the milk of cows who consumed hay previously treated at either of the two indicated levels.

Ely, *et al.* (*12*) found 0.8 and 1.8 p.p.m. of dieldrin in the milk produced by cows which were fed alfalfa hay previously sprayed with either 3.5 or seven ounces of insecticide per acre.

Pastures were treated with granulated dieldrin at the rate of 0.53 or 4.18 pounds per acre in experiments reported by App, *et al.* (*3*). Milk produced during the first 21 days after treatment by cows grazing on the former contained 0.11 to 0.18 p.p.m. of dieldrin while that from cows grazing on the latter contained 0.04 to 0.96 p.p.m.

Chlordane

Two plots of alfalfa were treated with a chlordane emulsion at the rates of one and two pounds per acre (*5*). Hay made from this alfalfa contained an average of either 20.4 or 20.8 p.p.m. of chlordane. Cows receiving this for a period of 150 and 100 days produced milk that contained from none to 0.2 p.p.m. of insecticide. Larger dosages of chlordane in soybean oil, when administered to cows, resulted in an increase in milk residues.

Heptachlor

Forage was sprayed with either 3.8 or eight ounces of heptachlor before it was made into hay in experiments reported by Ely *et al.* (*14*). When the hay was fed to cows, no heptachlor or heptachlor-epoxide (a metabolic product of heptachlor) was detected in the milk. A minimum intake of 1.3 mg. of heptachlor per kg. of body weight was necessary before heptachlor-epoxide appeared in the milk of a dairy cow.

Stoddard, *et al.* (*34*) and Harris, *et al.* (*18*) found heptachlor-epoxide in the milk from cows which were fed alfalfa hay made from forage which was treated

with heptachlor at the rate of four ounces per acre.

No heptachlor was found in milk by Polen, *et al.* (*30*) when the feed ingested by Holstein cows for a period of 20 days contained zero, one, 10, 50, 62.5 or 75 p.p.m. of heptachlor. Heptachlor-epoxide was found in milk only when levels greater than 10 p.p.m. were fed.

Aldrin

This chlorinated insecticide is related to dieldrin and is used primarily for the control of soil insects. Aldrin is more volatile than dieldrin, DDT or toxaphene and hence has a shorter residual action when sprayed on plants (*22*).

Kitselman, *et al.* (*20*) found no trace of aldrin in milk when hay made from previously sprayed forage was fed to a Jersey cow for more than 91 days. The work of Ely, *et al.* (*16*) showed similar results when no aldrin was found in milk of cows fed hay from forage previously sprayed with 3.9 ounces per acre. They further noted that aldrin had to be present in the diet of dairy cows at the rate of 28 p.p.m. before the insecticide appeared in the milk. Where sufficient aldrin was ingested, 11 to 14 per cent of the quantity administered was excreted in the milk.

Endrin

This chlorinated insecticide is related to both aldrine and dieldrin. It is most commonly used to control certain tobacco and vegetable pests together with cutworms in small grains (*22*). Ely, *et al.* (*11*) found less than 0.05, 0.14 and 0.15 p.p.m. of endrin in the milk of cows which received hay with residues of 1.9, 2.8 and 3.7 p.p.m. respectively. It was further noted that at least 20 mg. of endrin had to be ingested daily before the insecticide appeared in the milk of dairy cows.

Experiments were reported by Kiigemagi, *et al.* (*19*) in which dairy cattle were fed endrin in concentrations of 0.10, 0.25, 0.75 and two p.p.m. of their total diet. The insecticide was recovered from the milk after one week regardless of the concentration fed. Levels of endrin in the milk increased with both concentration fed and length of time of feeding. Residues of the insecticide were found in milk six weeks after final administration when the diet contained 0.25 p.p.m. or more. No endrin accumulated in the brain, heart, or kidney tissues while levels of 0.1 to 0.2 p.p.m. accumulated in the liver, 0.2 to 0.8 p.p.m. in renal fat, 0.1 to one p.p.m. in body fat and 0.1 p.p.m. in the roast.

Street, *et al.* (*35*) fed cows hay which was treated before harvesting with 1.5, three or 16.0 ounces of endrin per acre. Milk from these cows contained less than 0.1, 0.1 to 0.3, or 0.3 to 1.7 p.p.m. endrin. Cows which received highest levels of the insecticide showed clinical symptoms, reduced levels of food consumption and milk and butterfat production.

Toxaphene

Alfalfa was treated with a toxaphene emulsion at the rate of 1.5 pounds per acre (*5*). Hay was made and later fed to cows for 150 or 100 days. The average toxaphene content of the milk from the two sets of cows was 0.5 and 0.1 p.p.m. When larger doses of the insecticide in a soybean oil solution were administered, a higher concentration of toxaphene appeared in the milk.

ORGANIC PHOSPHATE INSECTICIDES

Organic phosphate insecticide residues on plants consumed by dairy cattle generally do not appear in the milk of such animals.

Dahm *et al.* (*10*) fed parathion to dairy cows at levels of one, five and 40 p.p.m. based on quantities of feed consumed. No parathion was found in milk from such animals when it was tested by both chemical and biological procedures. Alfalfa hay with parathion residues of 14 p.p.m. was fed to dairy cattle in experiments reported by Pankaskie, *et al.* (*28*). Both milk and jugular blood were analyzed and neither parathion nor free p-nitrophenol (believed to be a hydrolytic product of parathion) were detected.

Demeton, an organic phosphate systemic plant insecticide, was fed to a cow daily for three days in increasing doseages from 0.1 to 2.5 mg. per kg. of body weight in work reported by Dahm and Jacobson (*9*). Some cholinesterase inhibiting substances (believed to be the insecticide) were deteced in milk after the highest level of demeton was fed. They disappeared one day later. No cholinesterase inhibiting substances were present in milk when 0.29 to 1.0 mg. of Systox per kg. of body weight was ingested. Systox is a preparation which contains an emulsifying agent and 21.2 per cent by weight of a mixture of the thiono- and thiol-isomers of demeton.

In recent work with Phosdrin (O, O-Dimethyl 1-carbomethoxy-1-propen-2-yl phosphate), Casida, *et al.* (*7*) found no significant residues in milk or tissue of cows which were fed concentrations of one, five and 20 p.p.m. in feed daily for 12 weeks.

The absence in milk of organic phosphate insecticides after their ingestion by dairy cattle is explained in part by the work of Cook (*8*). He found that all of the organic phosphate insecticides tested were partially or completely inactivated by the cow's rumen fluid. More recently Plapp and Casida (*29*) found that Trolene (O, O-dimethyl O-(2, 4, 5-trichlorophenyl) phosphorothicate) was hydrolyzed at either the methyl-phosphate or phenyl-phosphate bond by bovine rumen fluid.

SUMMARY

When feeds treated with chlorinated hydrocarbon insecticides were fed to dairy cattle, residues of benzene hexachloride, chlordane, DDT, dieldrin, endrin and toxaphene appeared in the milk. Aldrin, heptachlor and methoxychlor were not detected in significant amounts. Maximum levels of insecticide present in milk ranged from 26 p.p.m. for DDT to 0.05 p.p.m. for endrin.

Residues of organic phosphate insecticides also occur on feeds and hence are ingested by dairy cattle, however, they generally do not appear in milk. It is believed that the insecticides are broken down in the rumen.

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