INSECTICIDE RESIDUES IN MILK AND MILK PRODUCTS[1]

I. Insecticide Residues in Milk from Treatment of Dairy Cows and Barns

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INTRODUCTION

During the last decade numerous organic insecticides have been synthesized. The impetus for this activity came during the war when supplies of botanical insecticides such as pyrethrum and derris from Africa and the East were cut off and when the demand was great for high levels of agricultural production (*29*). The first of these organic insecticides to be synthesized were DDT and benzene hexachloride. Since then, a considerable number of insecticides of the chlorinated hydrocarbon type and more recently of the organic phosphate type have been synthesized. Many of these insecticides have been used to control dairy cattle insects and also insects which infest forage crops that are consumed by dairy cattle. Such use has led to contamination of milk and milk products with these chemicals.

Dairy cattle are most commonly treated with insecticides to control flies, lice and grubs. The largest quantity of insecticides probably is used in attempts to control various kinds of flies. The importance of fly control to the producer can be appreciated when it is realized that twenty biting hornflies can consume one pint of a cow's blood in a week and can cause milk production to be reduced by three pints per day (*2*).

This paper will attempt to summarize information on residues of insecticides present in milk as a result of the treatment of dairy cows and barns. Other papers in this series will summarize information on insecticide residues in milk as a result of feeding treated crops to dairy cows (*35*) and on insecticide residues in milk products together with associated problems (*34*).

CHLORINATED HYDROCARBON INSECTICIDES

DDT

This insecticide has been suggested for use in the control of various types of flies (*3, 4, 5, 8, 10, 25, 28, 30, 40*) and lice (*43, 45*). It was found ineffective in the control of cattle grubs (*36*).

Recommendations in regard to the concentration of DDT in sprays applied to cattle vary somewhat. Laake (*30*) found that treatment of all cattle in a herd with a 0.2 per cent suspension of DDT gave protection against horn flies. Cory and Langford (*10*) applied a one per cent solution of wettable DDT powder to cattle and buildings at a pressure of 400 to 500 p.s.i. Cattle were protected against flies for three to six weeks and buildings for ten weeks. A two per cent solution of DDT in water applied to heifers at a pressure of 250 p.s.i. protected against horn flies from July to October (*40*). Bret and Fenton (*3*) reported that DDT was effective in the control of house flies and horn flies but not horse flies and heel flies when the barn was sprayed with a five per cent solution of watermiscible DDT powder at the rate of one gallon per 1000 square feet of surface area.

Experiments were reported (*28*) in which Holstein and Jersey cows were sprayed either daily with a five per cent water emulsion or suspension of DDT or semi-monthly with a 0.25 per cent suspension or emulsion. They found no DDT residues in the milk until the seventeenth day after the start of the spraying procedure. Maximum levels of DDT were attained more rapidly in the milk of cows sprayed with emulsions instead of suspensions, however, the levels ultimately obtained with both types of spray were about the same (*e.g.*, 19 p.p.m.). The milk from one animal which was sprayed daily with a five per cent suspension attained a maximum of 33.6 p.p.m. of residual DDT. Appreciable quantities of DDT were present in the milk of two cows at the close of their lactation periods 119 and 126 days after the final spraying. When these cows freshened 91 days later, no DDT was found in the milk.

Four dairy herds and barns were sprayed with a 0.5 per cent suspension of DDT as frequently as necessary for the control of horn flies during the 1948 season (*7*). Milk contamination during this period was in the range of zero to 1.3 p.p.m. with an average of 0.21 p.p.m. of DDT.

Carter, *et al.* (*5*) reported that the average DDT concentration in milk for a seven month period was 0.5 p.p.m. with a maximum of 2.0 p.p.m. when cows were sprayed with a 0.5 per cent suspension. When a 0.25 per cent suspension was used, the average DDT concentration in milk for a seven month period was 0.3

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

p.p.m. with a maximum of 1.5 p.p.m. Further work (*4*) showed that when grade Jerseys in full lactation were sprayed with a 0.5 per cent concentration of DDT, 21.8 p.p.m. appeared in milk two days later. This amount gradually dropped to 0.6 p.p.m. in 21 days.

The most important factor contributing to the loss of DDT from the hair of cattle is the licking of themselves or each other (*21*). Sunlight, rain, loss of hair, sloughing off of sebaceous secretions and absorption by the hair were found to be of little significance (*21*). A study (*7*) was made to determine the source of milk contamination when barns were sprayed with a 2.5 per cent concentration of DDT. Three experiments showed that the insecticide was actually secreted in the milk and did not get into the milk after milking by mishandling of it or milking equipment. Furthermore, contamination of milk did not result from inhalation of the insecticide by the cows but instead it came from ingestion of spray residues left on feed troughs. Another view was reported by Harris, *et al.* (*25*) when they found that milk contamination in barns sprayed with DDT resulted only from the careless handling of milk rather than from the cow after inhalation, or ingestion of the insecticide.

Strains of DDT-resistant flies have developed since the widespread use of the insecticide was instituted. This tendency toward development of resistant strains was already noted in 1948 (*33*). The presence of high levels of DDT-dehydrochlorinase in some tissues of DDT-resistant house flies was observed by Miyake, *et al.* (*37*). It was suggested that this enzyme catalyzes the dehydrochlorination of DDT to a nontoxic compound and hence may be responsible together with lipoproteins for protecting resistant flies. A recent report (*1*) suggested that cattle lice have also developed resistance to DDT.

Methoxychlor

This chlorinated insecticide is related to DDT but is less likely to accumulate in milk and the fatty tissues of animals. Some flies have developed resistance to methoxychlor (*32*).

The use of methoxychlor as a spray for the control of flies on dairy cattle has been suggested by several investigators (*4, 5, 6*). Other suggested uses include application to dairy barns as a residual spray for fly control (*12, 24*) and application to dairy cattle by means of back rubbing devices for control of flies (*42*) and lice(*20*). When applied to dairy barns as a residual spray, methoxychlor controlled flies for periods of three to four weeks (*12, 24*).

A mixture of methoxychlor and butoxypolypropylene glycol has also been suggested for use as a residual spray in dairy barns (*24*) and for direct application to dairy cattle by means of a back rubbing device for control of horn flies (*42*).

Carter, *et al.* (*5*) found that organic chlorine residues in milk from cows sprayed with methoxychlor were too small to positively indicate the presence of the insecticide. More recently, they reported (*4*) that when cows were sprayed with a 0.5 per cent solution of methoxychlor, 0.4 p.p.m. appeared in milk drawn two days later and 0.08 p.p.m. was found in milk drawn after 21 days. Clayborn and Wells (*6*) found that only relatively small amounts of methoxychlor were secreted for short periods of time by cows sprayed with the insecticide. Cows were sprayed with either a 0.5 per cent methoxychlor solution or a solution which contained five per cent methoxychlor and 50% butoxypropylene glycol in experiments reported by Helrich, *et al.* (*26*). They found methoxychlor residues of 0.13 to 0.32 p.p.m. in milk 12 hours after application. The residues gradually disappeared from subsequently drawn milk.

Benzene Hexachloride and Lindane

Benzene hexachloride is a chlorinated insecticide referred to as BHC. It has a persistent, disagreeable odor which may impart off-flavors to foods (*32*). Lindane is a purified form of the gamma isomer of BHC and has little or no objectionable odor (*32*). House flies have developed resistance to lindane (*24, 32*).

Benzene hexachloride has been suggested for use on dairy cattle to control flies (*17*) and lice (*16*). Spraying cattle with a 0.5 per cent solution was suggested for the control of flies while a 0.15 to 0.30 per cent solution used as a dip was suggested for the control of lice (*16, 17*).

It was indicated in 1947 that BHC might appear in milk from treated cattle (*16*). This was confirmed in 1948 (*17*) when BHC was detected in cream produced by previously sprayed cows. A maximum of 5.5 p.p.m. of the gamma isomer of BHC was detected on the day after treatment and smaller amounts were found for eight additional days. Cows were unable to lick themselves or each other and hence it was believed that BHC was absorbed through the skin.

Lindane has also been suggested for use on dairy cattle to control flies (*41*) and lice (*20*). Several reports (*1, 13*) have indicated that lindane was inferior to other insecticides in the control of cattle lice, perhaps because resistant strains have developed.

Ely *et al.* (*15*) found that lindane, in concentrations up to 3.5 p.p.m. was present in milk from sprayed cattle on the day after treatment.

Dieldrin

Dieldrin is a chlorinated insecticide primarily used for the control of fruit and vegetable insects (*32*).

It has been suggested for use in the control of flies and lice on dairy cattle (*4, 6, 13*). Roth and Johnston (*44*) have found that grubs can be controlled by injecting cattle subcutaneously with 25 mg. of dieldrin.

Claborn and Wells (*6*) found dieldrin in milk from sprayed dairy cows for 14 to 20 days after treatment, although highest concentrations were present after two to three days. Carter, *et al.* (*4*) detected 5.6 p.p.m. of dieldrin in milk from cows which had been sprayed two days previously with a 0.5 per cent solution of the insecticide. Nineteen days later the level of dieldrin in milk had dropped to 0.12 p.p.m.

Chlordane

Chlordane is a chlorinated insecticide which is widely used to control household pests, termites, soil insects and plant-feeding pests. Some animal parasites have developed resistance to this insecticide (*32*). A 0.5 per cent concentration was suggested (*5*) for use to control horn flies on dairy cattle. An examination of milk from sprayed cows failed to show detectable amounts of the insecticide.

Cattle lice were controlled with a five per cent concentration of chlordane in oil applied by means of a back rubbing device (*39*). De Foliart (*13*) found that lice on cattle could be satisfactorily controlled for four to five months with a single spray application of a 0.5 per cent chlordane solution. It was further noted that combinations of chlordane with BHC, lindane or malathion did not prolong control beyond that obtained with chlordane alone.

Other Chlorinated Insecticides

Dairy cows were sprayed with a 0.5 per cent concentration of Dilan for fly control according to Carter, *et al.* (*4*). They found 0.7 p.p.m. of Dilan in milk from treated cows after two days and 0.15 p.p.m. after 21 days. Similar experiments were carried out with a 0.5 per cent solution of perthane and 0.4 p.p.m. appeared in milk after two days while 0.08 p.p.m. was present after 21 days. Perthane has also been suggested for use in the control of lice (*13*).

Toxaphene has been suggested for use on dairy cattle to control horn flies (*5*) and lice (*38*). No toxaphene was found in milk from cows which were sprayed with a 0.5 per cent concentration (*5*).

When cows were sprayed with a 0.5 per cent concentration of TDE, 1.2 p.p.m. appeared in milk from treated cows.

Heptachlor, an insecticide similar to chlordane (*32*), has been found effective in the control of cattle lice (*13*). Its low cost may be an important point in its favor (*13*).

ORGANIC PHOSPHATE INSECTICIDES

Diazinon

This compound is a phosphate insecticide and miticide used primarily in the control of DDT-resistant flies (*32*). Diazinon has been suggested for use in the control of flies in dairy barns and thus on dairy cattle (*18, 19, 22, 23*).Hansens (*22*) found that diazinon, when applied to barns in a one per cent concentration, controlled flies for 12 or more weeks. An application of a 0.5 per cent concentration resulted in seven to twelve weeks of control. Milk from cows housed in barns sprayed with diazinon contained none of the insecticide and showed no flavor changes (*22*).

Malathion

This insecticide is widely employed for the control of insects on trees, shrubs, flowers and field crops. It is also used for the control of insects in and around buildings (*32*).

Malathion has been suggested for the control of flies in dairy barns (*12, 22, 23*) and flies (*4, 9*) and lice (*13, 20*) on dairy cattle. It has been reported that malathion-resistant house flies have developed in an area where the insecticide was used for 2.5 years (*31*).

When used in dairy barns, malathion mixed with sugar controlled flies for two to four weeks (*22, 24*). De Foliart (*12*) reported that flies in dairy barns were controlled for one to two weeks with a 1.25 per cent concentration of malathion with or without added sugar.

A two or five per cent concentration of malathion applied by means of a back rubbing device has been suggested for the control of cattle lice (*20*). De Foliart (*13*) found that a 0.5 per cent malathion spray controlled lice on cattle for four to five months and destroyed 52 per cent of the louse eggs on the animal.

The use of malathion for control of flies on dairy cattle was suggested by Claborn, *et al.* (*9*) who found residues of 0.08 to 0.36 p.p.m. in milk five hours after cows were sprayed with a 0.5 or one per cent solution of the insecticide. Traces of the insecticide were found after 24 hours and no residue was detected in the milk three and seven days after the cattle were sprayed.

Carter, *et al.* (*4*) reported that less than 0.1 p.p.m. of malathion was found in the milk from dairy cows five hours after they were sprayed with a 0.5 per cent concentration of malathion. Traces of the insecticide were present 24 hours after spraying and it was completely absent from subsequent samples.

Chlorthion

This compound is commonly used for the control of insects on non-food plants (*32*). It has been suggested for the control of flies in dairy barns (*22, 24*). A 0.5 per cent concentration provided control of three to five weeks while a one per cent concentration con-

trolled flies for seven weeks. Chlorthion was not detected in milk from cows housed in a previously sprayed barn according to the results of Dahm and Raun (*11*).

Other Organic Phosphate Insecticides

Several other insecticides of this type have been suggested for use in dairy barn fly control. They include: isochlorthion (*11*), malrin (a mixture of malathion and perthane), American Cyanamid 4124, Bayer 21/199 (*19*) and pirazonin (*23*). The work of Dahm and Raun (*11*) showed that no residues of isochlorthion appeared in the milk from cows which were housed in previously sprayed barns. No information appears to be available on residues in milk of the other compounds mentioned above when they were used for the control of insects on dairy cattle or in dairy barns.

OTHER INSECTICIDES AND INSECT REPELLENTS

Emulsions and suspensions of piperonyl butoxide and pyrethrins have been suggested for the control of flies (*14, 27*) and lice (*45*) on dairy and beef cattle.

More recently, the use of repellents either singly or in combination with an insecticide has been suggested (*2, 46*). It has been claimed (*2*) that the use of repellents would serve to: (a) prevent the harming of desirable forms of wildlife, (b) prevent extermination of the bee, and (c) permit man to employ certain pests to destroy other pests. At present, the U.S.D.A. is taking a "dim view" of repellents since they do not persist on animals for long periods of time (*2*).

Repellents used include: butoxypolypropylene glycol; N, N-diethyl-m-toluamide; 2, 3, 4, 5-bis (Δ2-butylene) tetrahydrofurfural; n-octyl bicycloheptene; di-n-butyl succinate (approved in 1957 by F. & D. A. for use on dairy cows), and di-n-propyl isocinchomeronate (registered with U.S.D.A. for use in spraying barns, dairy and beef cattle).

Toren, *et al.* (*46*) investigated the presence of 2, 3, 4, 5-bis (Δ2-butylene) - tetrahydrofurfural in milk from cows sprayed daily for five weeks with this repellent. Their method of analysis was sensitive to the presence of 0.1 p.p.m. of the repellent. No milk samples tested were found to contain this minimum amount.

SUMMARY

When clorinated hydrocarbon insecticides were used in dairy barns or on dairy cows, residues of benzene hexachloride, DDT, dieldrin, dilan, lindane, methoxychlor, perthane and TDE appeared in the milk. Such residues varied in magnitude from 0.4 p.p.m. for methoxychlor and perthane to 33.6 p.p.m. for DDT. Levels of insecticide residues in milk generally decreased after exposure of the cow but, in some instances, low levels persisted for long periods of time. Some insects have developed resistance to certain chlorinated hydrocarbon insecticides.

Organic phosphate insecticides suggested for use in the management of dairy cattle include: American Cyanamid 4124, Bayer 21/199, chlorthion, diazinon, isochlorthion, malathion and pirazonin. Residues of organic phosphate insecticides tested generally did not appear in milk after use in dairy barns. Resistance of files to malathion has been reported.

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