1964, over 1,000,000 of these infants will be fed by commercial formula services in at least 25 states.

The future of this industry will depend, to a great extent, on the continued adherence by formula plant operators to exacting standards of environmental sanitation, and complete public health protection can be attained only through the establishment and enactment of a uniform code by federal, state, and local authorities.

**References**


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**CHLORINATED ORGANIC INSECTICIDE RESIDUES IN MILK**

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Editorial Note: Much concern and confusion exists relative to the pesticide problem in milk, milk products and other foods. Dr. Henderson, a member of the Technical Advisory Committee on Pesticides of the Dairy Industry Committee, presents an authoritative account of the background, significant developments and current situation relative to this problem.

The concern of the dairy industry with respect to pesticides in milk and dairy products began with an announcement of Mr. John Harvey of the Food and Drug Administration at the Dairy Conferences at Miami Beach in October 1959 (3). He stated that it was apparent that education would not solve the pesticide problem alone and that the time for positive action and perhaps product seizures was here. This announcement was followed by seizures of evaporated milk and butter in January 1960.

Two surveys of milk supplies in 1955 and 1959 by the Food and Drug Administration indicated that pesticide residues were fairly common in milk (1, 2). A new technique, paper chromatography, was used in these surveys. This technique was not generally available to the public until Paul Mills’ paper was published in the November 1959 issue of the Journal of Official Agricultural Chemists (7). The dairy industry has been criticized for its delay in being concerned with the problem. I do not think this criticism is justified—it required the availability of a fairly simple screening test before progress could be made in this field. The seizures and the availability of the Mills test did stimulate the dairy industry and the state and other regulatory agencies to tackle the problem and to attempt to determine the sources of contamination and to try and reduce pesticide residues to the lowest possible levels. The Dairy Industry Committee, composed of the eight secretaries of the dairy trade associations, appointed a Technical Advisory Committee to explore and find solutions if possible for various aspects of the pesticide problem.

The DuPont Company filed a petition in 1957 for a tolerance of 0.25 ppm methoxychlor in milk under the Miller Amendment of the Food, Drug and Cosmetic Act (5). A Committee of the National Research Council, National Academy of Science, appointed to advise the Secretary of Health, Education and Welfare on the evaluation of the scientific data contained in the petition, recommended that a finite tolerance be denied. The denial of this petition appears to be the basis of the Food and Drug Policy that no finite tolerances be given for pesticide residues in milk and dairy products, or the so-called “zero” tolerance.

In order to determine the presence of residues in milk it was necessary to use a specific test and preferably, an official test published in AOAC analytical methods. The AOAC colorimetric test was available for this purpose. The test is specific for DDT and its analogues and is reported to be not reliable to less than 2.5 ppm on the basis of the fat. This is, of course, not zero in the absolute sense but was an “administrative zero” based on a specific test and apparently was used to determine compliance with the law which prohibited pesticide residues in milk.

The Mills’ paper chromatographic test was more sensitive than the AOAC or Schechter-Haller modification of the test and it was not limited to the detection of the DDT series of chlorinated hydrocarbons; lindane, methoxychlor, aldrin, dieldrin, heptachlor, endrin and many others could be detected if they were present in sufficient amounts. The Mills paper chromatographic test was extremely-useful in the early stages of the problem, but it is a qualitative and only a semi-quantitative test. It received first action by AOAC as a qualitative test but not second...
action for an official test.

In late 1960 and early 1961 the Food and Drug Administration had considerable interest in the microcoulometric-gas chromatographic procedure for determining pesticide residues. The Dohrmann equipment used for this procedure was developed by Dr. Dale Coulson of the Stanford Research Institute. In this procedure the organic chlorine is titrated as chloride ion and thus it is a quantitative test. The graph obtained from the recorder provides information for determining the amount of residue in the sample and establishes an emergence time that can be compared with the standard compound. The microcoulometric equipment can be equipped with a titration cell for sulphur and is thus valuable for use for many organic phosphorus insecticides.

Early in 1961 and in 1962 interest was shown in a new technique that was announced in 1960—the electron-capture or electron-affinity gas chromatographic procedure (6). This procedure was reported to be sensitive to many pesticides at very low levels—one part per billion or lower. The development of the equipment for use in milk was doubtless delayed due to the impression that it was not necessary to require a good “clean up.” For milk products, the Mills or other, efficient “clean up” is essential. A collaborative study reported in November 1962 indicated that more work was necessary for this procedure to reach its potential (4).

Since mid-1962 great strides have been made in this procedure. New packed columns, better understanding of parameters and adherence to efficient “clean up” procedures have contributed to the progress. At the present time I think it is safe to predict that when an official AOAC test is announced it will probably be an electron-capture gas chromatographic procedure. The microcoulometric equipment is valuable in the hands of a skilled operator and results comparable to electron-capture are possible. The following data indicate results that may be obtained (data in ppm in the fat):

<table>
<thead>
<tr>
<th>DDT</th>
<th>DDE</th>
<th>DDD</th>
<th>Dieldrin</th>
<th>Total Residues</th>
</tr>
</thead>
<tbody>
<tr>
<td>D</td>
<td>0.16</td>
<td>0.35</td>
<td>0.09</td>
<td>0.03</td>
</tr>
<tr>
<td>D</td>
<td>0.12</td>
<td>0.41</td>
<td>0.055</td>
<td>0.05</td>
</tr>
</tbody>
</table>

A new technique, thin-layer chromatography, is also being developed to a point where it is a valuable screening tool and for confirmation of questionable results obtained by the electron-capture procedure. The spots on the plate can be removed manually and checked by chemical or other techniques.

The above methodology has been discussed since the change in “administrative tolerance” is based on the sensitivity of methodology. The “zero” or administrative tolerance remained at 2.5 ppm on the fat basis until October 11, 1963 when the Food and Drug Administration announced that improvements in methodology had made it possible to positively identify residues at the following levels:

Separately or in total

- DDT: 0.05 ppm in milk
- DDE: 1.25 ppm in fat
- DDD:...

On January 16, 1964 lindane, BHC and methoxychlor were added to the list at 0.05 and 1.25 ppm in milk and fat respectively.

The October 11, 1963 announcement added 5 other residues that previously had not received much attention outside of FDA:

- Aldrin
- Dieldrin
- Heptachlor
- Heptachlor-epoxide
- Endrin

In determining compliance or action levels the following interpretations are made:

- Aldrin or dieldrin, individually or as an aggregate, total at levels of 0.01 ppm in whole milk or 0.25 ppm on a fat basis.
- Heptachlor or heptachlor-epoxide, individually or as an aggregate, total at levels of 0.01 ppm in whole milk or 0.25 ppm on a fat basis.
- Endrin total at levels of 0.01 ppm in whole milk or 0.25 ppm on a fat basis.

The above group of toxic compounds was referred to in the “President’s Report on Pesticides” (Wiesner Report) and their toxicity was emphasized (8). This report and perhaps Rachel Carson’s “Silent Spring” apparently stimulated the recent concern for this group which is now receiving special attention in pesticide residue analysis. I suspect that FDA tests every sample of milk and dairy products for this group and that their findings, together with the improved methodology, prompted the announcement of October 11, 1963.

A survey made by FDA showed that of approximately 3,000 samples of milk analyzed during the period July 1, 1963 through March 10, 1964, 49 or 1.6% had actionable levels of residues. Only 8 samples exceeded 1.25 ppm in DDT whereas 29 samples exceeded 0.25 ppm in dieldrin and 16 samples exceeded 0.25 ppm of heptachlor epoxide. The “actionable level” samples were largely confined to three areas—20 in Denver, 14 in Baltimore and 10 in the Minneapolis district. The Baltimore area has recently received considerable publicity because of heptachlor epoxide values in milk that exceeded current “actionable level.” It has been reported that farmers treated alfalfa with heptachlor as recommended in
the USDA Handbook 120. This treatment, using tests available at the time of approval, did not produce “actionable” residues. The current more sensitive methods used by regulatory authorities showed that residues present exceeded expiration date for payment is January 31, 1965.

cause the cows were fed hay treated according to the tests available at the time of approval, did not health hazard use on agricultural crops and on animals. The milk lost in the Baltimore area due to residues. The expiration date for payment is January 31, 1965.

WHERE DO RESIDUES COME FROM?

The USDA and FDA each have a responsibility at the Federal level to regulate the use of insecticides to the degree that they will not create a public health hazard and at the same time permit their use on agricultural crops and on animals. The USDA administers the Federal Insecticide, Fungicide and Rodenticide Act of 1947. This Act requires that insecticides shipped in interstate commerce be registered with USDA. Before an insecticide product can be registered, the manufacturer must submit proof that the chemical will safely and effectively accomplish the purpose for which it is manufactured when used in accordance with directions developed for its use. The burden of proof is placed on the manufacturer.

The Food and Drug Administration administers the Federal Food, Drug and Cosmetic Act of 1938. The Miller Amendment of this Act (Section 408), often referred to as the Pesticide Amendment of 1954, is specifically concerned with tolerances. When a pesticide chemical is to be used on a food crop, both agencies may be involved. If the compound leaves a residue, the USDA delays registration until a tolerance has been established by FDA. In order to secure a tolerance the manufacturer must file a petition under Section 408e of the Food, Drug and Cosmetic Act. The manufacturer must furnish experimental evidence on toxicity to establish what tolerance, if any, will be safe.

When a tolerance has been set by FDA, the USDA registers the insecticide which can then be marketed interstate with approved labeling. Most states have a “uniform state act” or other legislation requiring that pesticides conform to Federal Standards.

With controls established at Federal and some state levels why do we have the pesticide residues now found in milk? When electron-capture procedures are used it is doubtful that any milk will show “zero” or negative tests — a trace at least can usually be found.

Prior to the current interest in chlorinated hydro- carbons in milk, that is, before 1960, much of the contamination was due to improper use on the dairy farm. The spraying in barns, in milk houses and on cows with insecticides that should not be used for these purposes has resulted in residues in the milk. It was the contention of many regulatory personnel that this was the principal source of contamination and that since it was under the control of the dairymen, it like the antibiotic problem, could be quickly solved. The feeding of “trash” materials, such as apple pumice, trimmings of lettuce, cabbage and other plants, sweet corn stover, etc., contributed much to the contamination. An educational program that began in 1960 and warned against the mishandling of sprays on the dairy farm and the dangers from feeding trash materials, resulted in a marked reduction in pesticide levels but not the incidence. As methodology improved, fewer and fewer “zero” or negative tests were reported.

The residues persisted at lower levels and much research indicated that feed, contaminated by drift from aerial application of insecticides on fields of other crops resulted in contamination of forage and pastures, was one of the major sources of residues. The relationship of low level ingestion of insecticides in feed to the levels of residues in milk has not been well established by research work. Trials conducted at the University of California on feeding DDT at low levels indicate the relationship between DDT in the feed and DDT in the milk (9). Pairs of dairy cows were fed 0 to 5 ppm DDT based on the feed intake. The regimes were maintained for six weeks. The results indicated that the maximum level of DDT in the feed that did not produce detectable residues in the milk was 0.5 ppm. The State of California requires that alfalfa offered for sale have less than 0.5 ppm DDT. This level was established when the FDA “actional level” was 2.5 ppm on the basis of the fat. The 0.5 ppm in hay provides for some safety factor but with an “actional level” of 1.25 ppm a lower level in the feed is indicated, perhaps 0.25 ppm.

Dr. Witt of the University of Arizona has reported that when DDT dust is applied at the rate of two pounds per acre, the spray applied as dust can contaminate feed with 0.25 ppm approximately 3½ miles downwind from the border of the target crop. In the form of a spray, the same concentration of DDT was found at little less than one-half mile from the border of the target crop. Of course the drift found in any one trial will be affected by wind direction, wind velocity, temperature inversion, humidity, type of formulation, particle size, dosage rate and many other factors.

The feeding of contaminated feeds to growing heifers will result in residues in the milk when she
freshens. In California a new law prohibits the sale of heifers fed contaminated feed or milking cows that have pesticide residues in their milk. The DDA test of urine is used to administer the law. In Los Angeles where the cow replacement rate is high it is important that heifers purchased from other areas do not have residues in the milk when they freshen.

The heptachlor epoxide problem on alfalfa has indicated that because of the improved sensitivity of analytical methods all recommendations of the USDA and the University Extension Departments should be re-examined.

What can be done to reduce residues?

1. Since most of the control must occur at state levels, it is essential that state programs be designed to control the use of spray materials.
   a. A law governing the registration and sale of insecticides is essential.
   b. The licensing of applicators, both private and commercial, and continued inspection of their performance is necessary.
   c. Many toxic insecticides should only be applied after a permit is secured indicating the composition of the material, the crop to be treated, the rate of application, list of adjacent crops and the name and license number of the applicator. For administrative purposes it is desirable that the County Agricultural Commissioner or similar authority issue the permits.
   d. A state agency (usually the Department of Agriculture) should maintain an inspection force to secure milk samples and work with the dairymen to assist in locating sources of contamination. A laboratory or laboratories should be maintained to test the samples supplied by the inspectors.
   e. Samples of hay and grains offered for sale in markets should be tested at intervals for compliance with residue tolerance where established.
   f. A tolerance level (usually the current FDA "actionable level") should indicate the point where corrective action will be taken.

The above program is best administered by one state agency. A strong Department of Agriculture is recommended with the Department of Public Health acting in an advisory capacity.

2. Restrict the use of chlorinated hydrocarbons if residues cannot be controlled in other ways.
   a. Two states have taken steps to restrict the use of DDT as a dust. In October 1963 California adopted a law that required a permit be secured from the County Agricultural Commissioner for each application. Arizona held a hearing August 6, 1964 to make legal a voluntary prohibition on the use of DDT as a dust for one year.
   b. Cancel the approval of compounds such as chlordane, heptachlor, dieldrin and endrin when they are used on any crop that could cause residues when fed to dairy cattle. Restrictions are now in effect on many crops normally fed to cows.

3. Prohibit the sale of heifers or dairy cows that have been exposed to insecticide contaminated feeds until tests prove the milk will not contain residues in excess of "actionable levels".

What about Tolerances?

The new "actionable level" issued by the Food and Drug Administration on October 11, 1963 alerted the dairy industry to the fact that further improvements in methodology could result in levels that could not be met by prevailing agricultural methods even when the best practices were followed. The Food and Drug Administration Commissioner, Mr. George Larrick, when testifying at the Ribicoff Senate Committee meeting indicated that as methodology improved, it will be necessary to establish finite tolerances - even for milk, he added. The Miller Amendment provides a mechanism under Section 405e for securing tolerances. The dilemma is: At what level shall tolerances be set? What is "safe" and what is below "pharmacological insignificance"? At one time 2.5 ppm DDT in the fat must have been considered "safe". Is 1.25 ppm "safe" or below "pharmacological insignificance"?

References