PESTICIDE RESIDUES IN FOODS

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Man no longer has direct control over many such essential factors affecting his life as the production and preservation of food, clothing, and shelter. By both design and accident, we have reached a stage of civilization and technology where society acquiesces in the deliberate addition of large quantities of toxic chemicals to the environment. The total is the sum of many individual contributions, each with a limited objective. Both assessment of the total hazard and control of that hazard pose problems requiring highly specialized knowledge, comprising a complex body of many laboriously gathered facts evaluated for their relative importance and analyzed for their implications. The health and well-being of man and animals depend upon this knowledge. Problems of pesticide residues in foods are not confined to a single nation. International trade and modern transportation make pesticide residues a matter of international concern. Many international organizations are striving to bring some uniformity into the controls required to ensure safety and the best interests of society.

Many reports and publications are concerned with pesticides, their uses, toxicology, persistence, and residual characteristics. This body of literature has been developed during the past quarter century. During this same period of time, man has managed to distribute these pesticide chemicals literally over the surface of the world and into the flesh of man and animals.

Food chains are the most probable source of these residues, and several investigators have postulated that the major source of pesticide chemicals in humans is to be found in the diet. Although this may be a true evaluation of the situation, many other avenues of exposure affect the cumulation of pesticide chemicals in humans. There are many uses for pesticide chemicals other than in food production. Large amounts are used in forest conservation, in control of household pests, in disease control, and in fiber production. Residues remain in the soil and are suspended in the air. They are distributed further through water systems, either from direct application or from runoff. The individual can no longer exercise a choice concerning exposure to pesticide chemicals.

There is a substantial body of literature showing that residues remain in food under controlled experimentation. At the same time, relatively little information has been published on pesticide residues in food in market channels. This report will be confined to foods in market channels. Regulatory control is effected through enforcement of established tolerances for residues on the raw agricultural commodity. Regulations provide that processed foods prepared from raw foods containing residues within legal tolerances will not be illegal if the residues have been removed to the extent possible in good manufacturing and if the remaining residue does not exceed the tolerance on the raw product. Again, few data have been published on commercially processed food. Data gathered by the various government units are usually collected and published at intervals. Since there is little uniformity in the presentation of the data, some difficulty is encountered when comparisons intended to show the total pesticide residue in foods are attempted.

Durham[1] comprehensively reviewed pesticide residues in food as related to

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human health in 1963 and expressed the opinion that the available evidence did not indicate that the levels of pesticide residues in the general food supply were increasing. He compared reports involving seizures with reports of governmental agencies that test products for compliance with tolerances. He reported that the State of California found that between 6.5–8.8 per cent of the samples of raw agricultural produce contained excessive residues. Recent reports from the University of California[2] indicate that the percentage of excessive residues found on fruits and vegetables declined from 3.1 in 1962, to 2.2 in 1963, and to 0.7 in 1964. During the same period, these reports showed substantial increases in the percentages of samples of animal feed containing excessive residues. Milk samples from California dairies containing residues increased from 70.6 per cent in 1962 to 92.8 per cent in 1964.

Milk and dairy products are of particular concern throughout the world. The first formal tolerance for DDT residues in milk and dairy products was approved under Section 408 of the Federal Food, Drug and Cosmetic Act in March 1967 on petition from the State of California. The approved levels in milk are 0.05 ppm for DDT, DDD(TDE), and DDE, singly or in combination. At the same time, a food additive tolerance was set at 1.25 ppm on the fat basis for these same chemicals in manufactured dairy products. These reports also show concentrations of DDT and its metabolites in coastal and deep sea marine foods ranging from 0.01 ppm to 4 ppm. There are similar studies[3] of residues in oysters and fish from Louisiana waters during 1964, where 42 per cent of the oysters sampled contained one or more chlorinated pesticides at levels of 0.01 ppm or greater.

Reports[4] by the State of Florida show samples of food (including milk, eggs, animal feed, and meat) containing excessive residues of 4.9 per cent in 1963, 8.0 per cent in 1964, 10.1 per cent in 1965, and 10.0 per cent in 1966. Canadian authorities[5] report 6.3 per cent of domestic samples (3,167 total samples examined) and 3.0 per cent import samples (461 total samples examined) as unsatisfactory because of pesticide residue in 1966. Egan[6] and Hammence[7] have reported residue studies on market samples of meat, dairy products, and vegetables in the United Kingdom. Egan's survey resulted in the imposition of restrictions on the use of several persistant organochlorine compounds. Hammence points out that only those samples giving positive results through the drosophila screening procedure were subjected to chemical assay. Lyarsky[8] reports that, of 125 agricultural chemicals used in Russia, 38 are found as residues in foods. He states that 350 laboratories are engaged in sampling at regular intervals for the presence of agricultural chemicals.

Results obtained in the Food and Drug Administration's (FDA) regulatory control programs show excessive residues in 4.3, 3.0, and 3.6 per cent of domestic samples in 1964, 1965, and 1966, respectively. The relatively high incidence of excessive residues in the above reports is biased with respect to the total food supply. In most cases, varying degrees of selectivity were used by the regulatory agencies as a means of controlling further distribution of food containing excessive residues. Regardless of the bias introduced by selecting samples from sources likely to contain excessive residues, such information on the pesticide content of the food supply is valuable even though the full limits of the problem are not defined. The information does show beyond question that pesticide residues are widely distributed throughout the world in most food sources. During the period July 1, 1963 through June 30, 1966, FDA examined 53,194 objective samples. Objective samples are those in which there is no reason to suspect residues; for

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all practical purposes, they can be considered as random samples. This class of samples included 3,836 import samples. An additional 25,728 samples were examined because of suspected residues. During the fiscal years 1965 and 1966, the U. S. Department of Agriculture[9] examined 5,036 objective samples of animal tissue collected from animals slaughtered in federally inspected establishments.

In our opinion, the above number of samples is sufficiently large to justify a high degree of confidence in general observations concerning the distribution, incidence, and levels of the various pesticide chemicals present in the food supply. For the purposes of this discussion, the samples have been divded into eight classes, as follows: (1) raw agricultural products; (2) fluid milk; (3) manufactured dairy products; (4) processed animal feeds; (5) shell eggs; (6) fish and shellfish; (7) meat; and (8) all others. The total number of samples of such products as canned or frozen foods, vegetable oils, and special dietary foods was not regarded as sufficiently large to consider separately, and these categories are reported under "all others." All data on meat were furnished by the Livestock Slaughter Inspection Division, Consumer and Marketing Services, U. S. Department of Agriculture.[9] Results on meat are not included in averages or percentages except as noted.

TABLE 1 shows that the distribution of FDA samples by food classes was relatively uniform during each year. The major exceptions were a decrease in the sampling of domestic manufactured dairy products each year and an increase in the sampling of imported fish and shellfish after the first year. Residues were found in the above samples as follows:

Fiscal Year	*1964*	*1965*	*1966*	*Total*
Percentage with residues	44.3	53.6	52.6	49.5
Percentage with multiple residues	23.8	33.3	32.8	29.3
Percentage with excessive residues*	3.6	3.0	3.0	3.2

* Classed as illegal if subject to the Food, Drug and Cosmetic Act.

TABLE 1

DISTRIBUTION OF SAMPLES AMONG FOOD CLASSES

	Fiscal Year							
	1964		1965		1966		Total	
	Domestic	Imports	Domestic	Imports	Domestic	Imports	Domestic	Imports
No. Samples	20,088	1,222	14,462	1,216	14,806	1,398	49,356	3,836
	%	%	%	%	%	%	%	%
Food Class								
Raw agricultural products	63.0	45.8	64.6	51.7	66.2	54.3	64.4	50.7
Fluid milk	18.3		16.5		16.8		17.3	
Manufactured dairy products	9.5	21.9	7.4	14.1	4.1	17.9	7.3	18.0
Processed animal feeds	3.4	4.0	4.1	5.1	4.6	2.5	4.0	3.8
Shell eggs	2.5	3.6	3.2	2.1	3.4	2.9	3.0	2.9
Fish & shellfish	0.6	1.8	2.2	9.7	2.0	9.5	1.5	7.1
All others	2.7	22.8	2.0	17.2	3.0	12.9	2.5	17.4

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The increase noted from 1964 to 1965 may be due to improvements in methodology that occurred about this time rather than to an actual increase in the residues present.

About 71 per cent of the objective samples of animal tissue examined in the U. S. Department of Agriculture program contained residues. There is no significant difference in the incidences of residues found in domestic and imported foods. Residues are found in about half of the FDA samples, and nearly a third of the samples contain more than one residue. The findings are generally the same when computed on an annual basis.

The same general picture is observed when the incidence of residues is calculated on each major class of foods. The data do not indicate that there has been a major change in the general situation during the three-year period. It is of interest to note that, with the high incidence of residues, the proportion of excessive residues remains at about 3 per cent. The percentages of excessive residue vary widely among food classes.

A total of 56,940 residues representing 81 different chemicals was found in the 26,326 FDA samples in which residues were detected. Thirty-three chemicals were common to each year. However, 11 chemicals account for 95 percent of the residues found. The 10 residues most frequently found in domestic samples, in import samples, and animal tissue are, in order of importance:

Domestic	*Import*	*Animal Tissue*
DDT	DDT	DDT and metabolites
DDE	DDE	BHC
Dieldrin	Dieldrin	Toxaphene
TDE	TDE	Lindane
Heptachlor epoxide	BHC	Dieldrin
Lindane	Lindane	Methoxychlor
BHC	Aldrin	Heptachlor epoxide
Endrin	Kelthane	Endrin
Aldrin	Heptachlor epoxide	Chlordane
Toxaphene	Endrin	Aldrin

Nine of the chemicals are common to import and domestic samples; these same chemicals are reported in animal tissue. Endrin, aldrin, and chlordane were found infrequently, 0.1 per cent or less. Methoxychlor was found in 2 per cent of the animal tissue samples. Although it is beyond the scope of this report to give a statistical analysis, these data indicate that these chemicals are generally distributed throughout all food classes.

The levels at which the residues were found is important in assessing the significance of the findings. TABLE 2 summarizes the findings on all residues in domestic samples by pesticide chemicals. These data show that 95 per cent of the residues are below 0.5 ppm: that 78 per cent are below 0.1 ppm: and that 57.9 per cent are below 0.03 ppm. Except for toxaphene and kelthane, the levels found for specific chemicals are generally of the same order of magnitude within each range. Excessive residues or combinations of residues amounted to 3.6 per cent, as compared to 3.0 per cent when calculated on a sample basis, indicating that some samples contain more than one excessive residue. The levels of residues in animal tissues were slightly higher in that only 79.6 per cent were below 0.5 ppm and 54.8 per cent were below 0.1 ppm.

The several studies of residues in prepared meals and in food ready for consumption during the past decade are not easily compared. Walker and his

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TABLE 2

PESTICIDE CHEMICALS.
RESIDUES IN DOMESTIC SAMPLES, 1963-1966, IN PERCENTAGES*

	DDT	DDE	Dieldrin	TDE	Heptachlor Epoxide	Lindane	BHC	Endrin	Aldrin	Toxaphene	Kelthane	All Residues
No. Residues	13,132	12,566	8,775	4,896	3,716	2,397	1,394	1,212	979	754	354	52,892
Range (ppm)												
T–0.03	50.4	62.0	65.2	60.7	45.7	76.7	68.2	72.0	79.7	16.8	17.8	57.9
0.04–0.10	22.8	19.9	19.3	21.8	24.7	14.1	18.2	20.1	14.3	4.8	22.9	20.3
0.11–0.50	20.2	15.2	14.0	14.2	27.2	6.0	11.5	7.5	4.6	28.0	42.4	16.8
0.51–1.00	3.4	1.7	1.2	1.6	1.8	2.4	1.3	0.2	0.9	18.6	10.2	2.5
1.01–1.50	1.1	0.5	0.1	0.5	0.4	0.3	0.4	0.2	0.3	8.0	2.8	0.8
1.51–2.00	0.6	0.2	0.1	0.4	0.1	0.1	0.1	—	0.1	3.8	1.4	0.4
Above 2.01	1.5	0.5	0.1	0.8	0.1	0.4	0.3	—	0.1	20.0	2.5	1.3
Excessive residues	1.5	1.0	4.9	1.1	7.5	1.1	0.6	29.8	4.5	9.0	0.3	3.6

* Residues in animal tissue not included.

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co-workers[10] analyzed restaurant and institutional meals in 1954, reporting a daily average DDT intake of 0.184 mg DDT. In 1956, Hayes *et al.*[11] reported that 16 meals from a penal institution contained an average daily intake of 0.202 mg DDT. Hayes *et al.*[12] also examined meatless meals in 1958 and reported a mean daily DDT content of 0.041 mg. In 1961, Durham[13] found 0.184 mg DDT in one day's meals from an Alaskan hospital.

Since the above period, significant improvements have been made in methodology, permitting reliable measurements at much lower levels. The most extensive studies of food ready for consumption are the total-diet studies carried out on a continuous basis by the FDA since May 1962. The results obtained on samples collected prior to June 1964 have been reported, but not otherwise summarized. Inspection of the data reported in 1963, 1964, and 1965 indicates levels and frequency rates similar to those of current findings. The results of these studies have been reported annually [14-17] since 1963.

Using the data reported by Williams,[15] Campbell *et al.*[17] calculated that the 23 diet samples contained a mean of 0.02 ppm DDT. A detailed report of the 46 samples collected during the period June 1964 through April 1966 has now been published.[10] These samples, collected, prepared, and examined under uniform conditions, are as reliable an index to kinds and amounts of pesticide residues consumed during the past several years in the United States as is available.

As might be expected, the pesticide chemicals most commonly found are the same as those found in the surveillance samples mentioned previously. Calculations show the following averaged levels of the chemical classes in a well-balanced diet:

chlorinated organics	– 0.025 ppm
organic phosphorus	– 0.003 ppm
chlorophenoxy compounds	– 0.003 ppm
carbamate compounds	– 0.05 ppm

There was no statistically significant difference between the first and second years of the studies under discussion based on the chlorinated organic compounds. The ranges and the averages are:

Period	*Average* (ppm)	*Range* (ppm)
1st year	0.021	0.001-0.042
2nd year	0.030	0.003-0.059
Combined	0.025	

The frequency rates and levels of other classes of pesticide chemicals are low and have not been statistically evaluated. It should be mentioned that an improved method for determining organic phosphates was employed in the second year of the study. No organic phosphorus residues were reported during the first year, and it is believed that the findings during the second year of the study may be attributed to this factor rather than to a change in the residues present.

FIGURE 1 shows the relative proportions of the different chemical classes of residues, expressed as percentage incidence of the number of positive findings and the total number of composites examined, as well as the relative amount of each chemical class expressed in percentage of the total amount of organic pesticide residues found.

The distribution of chlorinated residues among the 12 food classes is shown in FIGURE 2. The proportion of DDT and its analogs to the total chlorinated residues

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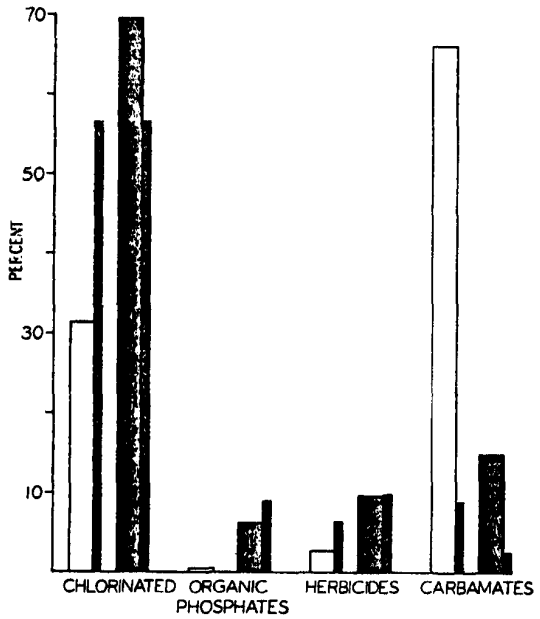


FIGURE 1. Distribution of Residues by Chemical Class.

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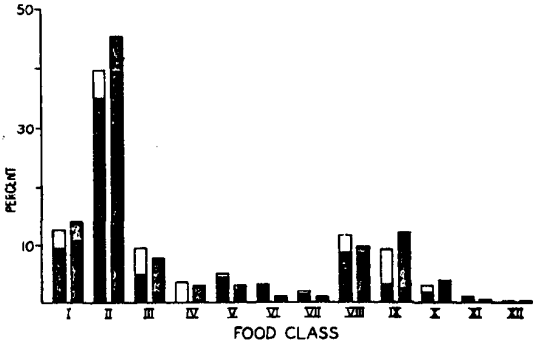


FIGURE 2. Distribution of Total Chlorinated Organic Pesticides and DDT Compounds Among Food Classes. [I: Dairy products (8-13% fat); II: Meat, fish & poultry (17-23% fat); III: Grain & cereals; IV: Potatoes; V: Leafy vegetables; VI: Legume vegetables; VII: Root vegetables; VIII: Garden fruits; IX: Fruits; X: Oils, fats & shortenings (83-88% fat); XI: Sugars & adjuncts; XII: Beverages.]

% 1964 total chlorinated residues

% 1965 total chlorinated residues

% DDT and analogs

TABLE 3

AVERAGE INCIDENCE AND DAILY INTAKE OF 15 PESTICIDE CHEMICALS FROM 516 DIET COMPOSITES

	Positive Composites (percent)	Daily Intake (Mg)
DDT	37.4	0.037
DDE	32.4	0.024
TDE	23.1	0.016
Dieldrin	20.2	0.006
Lindane	13.6	0.004
Heptachlor epoxide	12.6	0.003
BHC	6.2	0.003
Malathion	4.5	0.009
Carbaryl	4.7	0.082
Aldrin	4.5	0.002
2,4D	3.5	0.004
Diazinon	3.0	0.001
Kelthane	2.5	0.008
PCP	2.5	0.004
Endrin	2.3	T

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in each food class is shown. It is obvious from the above that the same chemical compounds appear in the finished food, although there is a difference in order of frequency and in the levels of residues found in objective samples of raw foods. Methoxychlor, found in 2 per cent of animal tissue samples, was not found in the meat, fish, and poultry composites of ready-to-eat foods. The relative amounts of specific chemicals present are important because of wide variations in toxicity.

TABLE 3 shows the incidence and calculated average intake of the 15 most commonly found pesticide chemicals in milligrams/day. The average intake is based on consumption of 4 kg food/day, which is almost twice that consumed by the "average" man. It can be seen that DDT and its analogs, DDE and TDE, at a level of 0.019 ppm in the diet, account for about 75 per cent of the daily intake of chlorinated organic compounds. DDT alone accounts for a third of the total intake. Residues of dieldrin, lindane, and heptachlor epoxide follow in order of frequency and, when combined with the DDT group account for 90 per cent of the total intake of chlorinated compound. Robinson and McGill[20] report an average daily intake of 0.0008 mg/kg body weight DDT compounds and 0.00026 mg/kg body weight dieldrin in their study in 1965 of prepared meals in the United Kingdom.

TABLE 4 compares "acceptable" daily intakes of pesticide chemicals with findings from FDA studies from June 1964 through April 1966. Acceptable daily intakes have been approved by the joint FAO-WHO committee[21] on pesticides. The potential daily intake is considered in establishing tolerances.[22]

In summary, residues of pesticide chemicals are present to some degree in about half of the lots of food entering market channels. There is some variation in incidence within broad categories of foods entering these channels, but no food category is free of residues. These residues are reduced, but not removed, during processing and preparation for consumption. The overall incidence, levels, and character of pesticide residues in foods have remained comparatively uniform since 1964. No significant trends have developed, though it can be seen that the greatest contribution of residues to man's diet is not food treated with pesticide chemicals. Residues found in fish, shellfish, dairy products, milk, and meat are unavoidable and indirect in that they result from environmental influences. It can be concluded that current levels of pesticide residues in the nation's food supply are not approaching the danger level. The frequency with which residues are

TABLE 4

COMPARISON OF AVERAGE DAILY INTAKES OF PESTICIDE CHEMICALS

Pesticide	FAO-WHO Acceptable Intake (mg/kg)	Total Diet Calculated Intake (mg/kg)
DDT	0.01	0.0005
(combined DDT, DDE, TDE)		0.0011
Lindane	0.0125	0.00006
Malathion	0.02	0.0001
Carbaryl	0.02	0.0012
	Calculated from tolerances	
Dieldrin	0.006	0.00009
Heptachlor epoxide	0.0006	0.00004
2,4-D	0.017	0.00005

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found suggests that further increases are undesirable and that attention should be directed toward reducing the levels, especially in food items such as dairy products, fish and meat, which acquired their residues indirectly.

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