Dietary Intake of Pesticide Chemicals

Calculated daily consumption of pesticides with foods are discussed and compared with currently accepted values.

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Thousands of samples of food are examined by the Food and Drug Administration each year to determine compliance with established pesticide tolerances on raw agricultural products. Residues of pesticide chemicals are found in about half of the samples, and generally about 3 percent of the samples contain residues in excess of, or not authorized by, legal tolerances. This relatively high incidence of residues in raw foods must be related, in proper perspective, to the amount of residues actually consumed as one measure of the effectiveness of legal tolerances in protecting the consumer. The continued investigation of the quantity of residues in foods ready for consumption is useful to other investigators who are primarily interested in the ultimate fate of residues in man.

The storage of chlorinated organic pesticide chemicals in human body fat has been the subject of several investigations (*1–3*). Campbell *et al.* (*4*) state that the total dietary intake accounts for most, if not all, of the DDT [1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)-ethane] and metabolite DDE [1,1-dichloro-2,2-bis (*p*-chlorophenyl)-ethylene] stored in the body fat of persons with no occupational exposure to the insecticides. Other investigators (*2*) have reached essentially the same conclusion.

Earlier reports (*4–10*) summarized results of investigations of pesticide residues in foods ready for consumption; these include data obtained by the Food and Drug Administration during previous "total diet" studies (*6*, *7*, *9*).

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Source of Data

This paper discusses the average daily intake calculated from the findings (*9*, *10*) of all pesticide chemical residues in or on "total diet" samples collected on 46 days in 25 different cities during the 699-day period of June 1964 through April 1966. As reported (*6*), this diet was constructed with the advice and assistance of the U.S. Department of Agriculture, Household Economic Research Division. The 16- to 19-year-old group was selected because members of this group consume greater quantities and kinds of food than other population groups. Each sample represented the total amount of foods and beverages consumed by one person for a 2-week period. The 46 diet samples, therefore, represented a food supply sufficient for a total of 644 days.

The limitations in interpreting data from a relatively small number of samples are well known. The samples collected for this study can be characterized as random. During the first year of the study (June 1964 to April 1965), samples were collected from retail food stores in three cities representing three geographic areas, with no restriction or designation of the food store. During the second year (June 1965 to April 1966), a total of five geographic areas were covered, and samples were collected in cities of different sizes ranging from major metropolitan centers to communities with a population of 50,000 or less. Food samples were collected in the same way a customer selects his food in a retail store, without regard to source or movement in interstate commerce. Undoubtedly a significant portion of the foods originated in producing areas outside the purchasing area. This belief is based on the known nationwide distribution of both raw and processed food products. Tables 1 and 2 summarize the annual findings in terms of the daily intake of specific chemicals found in each of the separate food classes. As reported (*9*), each diet sample was divided into 12 classes of similar foods and prepared for consumption by trained dieticians. The food items in each food class were composited and slurried for analysis. Each composited food class was analyzed separately by procedures capable of detecting 50 common pesticide chemicals.

The tabular summaries include the food-class composites from all diet samples. The daily intake in milligrams was obtained from the concentration in parts per million in the food class and the amount of food class prescribed for consumption during the 14-day period. Figure 1 shows the relative proportion of residues of the different organic chemical classes expressed as percentage of incidence of the number of positive findings and the total number of composites examined. The high proportion of chlorinated organic residues is not surprising, in view of their persistent character and the use patterns of this class of chemicals. Figure 1 also shows the relative amounts of each class of chemical, expressed in percent, in respect to the total amount, of organic pesticide residues found. The large proportionate amount of carbamate chemicals observed in the first year of the study was not found in the second year. In both years the proportionate amount of carbamate chemicals was much higher than the relative incidence.

Chlorinated Pesticide Chemicals

Residues of chlorinated organic pesticide chemicals were found in each of the 44 diet samples examined for this class of compounds. Except for beverages, such residues were found in each food class, although not in all composites of each class.

Although the tabular data are not presented on a regional or seasonal basis, Fig. 2 shows no substantial difference in the average intake in four of the five geographic regions. Results obtained on all diet samples collected in the Minneapolis area are quite low. Additional investigations are under way to determine whether there are significant differences in foods consumed in this particular region or if there was an experimental error. No recognizable seasonal influence on the daily intake is seen in these figures. The average daily intake of total chlorinated organic chemicals calculated from 18 diet samples during the first year was 0.082 mg. The individual diet samples ranged from 0.003 to 0.17 mg. The second-year average calculated from 26 diet samples was 0.12. The individual samples ranged from 0.013 to 0.23 mg. The combined average of both years was 0.10 mg. The range is not excessive when one considers the concentrations of combined residues present. The maximum intake on a sample basis found in each year was about double the average for all samples of that year. The order of magnitude has not changed.

The Student's *t*-test for statistical significance was applied to compare the data for intake of total chlorinated pesticide residues determined in each of the 2 years of our study. This class of pesticide chemicals was chosen because of the high incidence of postitive findings. In view of the fact that no significant difference existed between the two sets of data, they were combined for evaluation. With the data for both periods, it can be calculated, within 95 percent confidence limits, that any similar sample taken and examined during the 2-year period would contain chlorinated pesticide residues within the intake range of 0 to 0.22 mg per day. This calculated range is within the actual range found during this period. Further, it can be calculated, within 95 percent confidence limits, that the true value of any given sample should fall within the range of 0.08 to 0.12 mg per day.

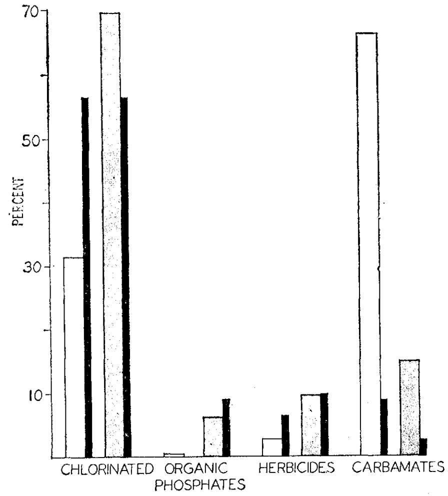

Fig. 1. Distribution of residues by chemical class. First pair of bars for each class, June 1964 to April 1965; second pair of bars, June 1965 to April 1966; solid bars, percentage of incidence; white and shaded bars, percentage of total organic residues.

The distribution of chlorinated residues, and the proportion of DDT and its analogs within this chemical class, among food classes is shown in Fig. 3. The major source of chlorinated organic residues was found to be the meat, fish, and poultry class. This class, combined with dairy products, accounts for more than half the intake of chlorinated residues. This is significant because there is little direct application of pesticide chemicals to these products, and therefore their presence must be due to indirect sources. Fruits, garden fruits, and grain each account for about 10 percent of the total intake of this class of chemicals. This pattern is not unexpected, in view of the fact that control is exercised over direct applications to raw agricultural products.

The qualitative makeup of the speci-

Table 1. Daily intake of pesticide residues, listed by food class (18 composites in each class) and expressed in milligrams per day from June 1964 to April 1965. T signifies trace (less than 0.001 mg). The average pesticide intake from beverages (class XII) was 0.104 mg of carbaryl per day. Percentage of fat content was 8 to 13 in dairy products; 17 to 23 in meat, fish, and poultry; and 83 to 88 in oils, fats, and shortenings.

Chemical detected	I Dairy products (8 to 13 percent fat)	II Meat, fish, and poultry (17 to 23 percent fat)	III Grain and cereals	IV Potatoes	V Leafy vegetables	VI Legume vegetables	VII Root vegetables	VIII Garden fruits	IX Fruits	X Oils, fats, and shortenings (83 to 88 percent fat)	XI Sugars and adjuncts
					Chlorinated chemicals						
DDT	0.002	0.011	0.004	T	0.001	0.002	0.001	0.006	0.002	0.001	0.001
DDE	.004	.012	T	T	0.001	T	0.001	T	T	T	T
TDE	.001	.005	0.001		0.002	0.001	T	0.002	T	0.001	T
Dieldrin	.001	.002	T	T		T	T	0.001	T	0.001	
Lindane	T	T	0.003	T	T			0.001	T	T	T
Heptachlor epoxide	0.001	0.001		T	T					T	T
BHC	.001	.001			T			T		T	T
Aldrin		T	T						0.001		T
Endrin			T	T				T		T	
Chlorbenside					T		T				
Heptachlor		T	T							T	
TCNB				0.002			T				
Tedion									0.001		
Kelthane									0.003		
Chlordane								T			
Perthane									T		
PCNB									T		
					Herbicides						
2,4-D										0.001	0.004
PCP		T	T								
MCP			0.002								
TBA										T	
					Inorganic residues						
Bromides*			4.20	0.19					0.12	1.19	0.33
Arsenic (As_2O_3)		T	0.002	0.063		0.001	0.001		0.002		
					Carbamates						
Carbaryl			0.021	0.003	0.004		0.001	0.002	0.008		0.005
Dithiocarbamates			0.012		0.014						

* Values reported are those in excess of 25 parts per million.

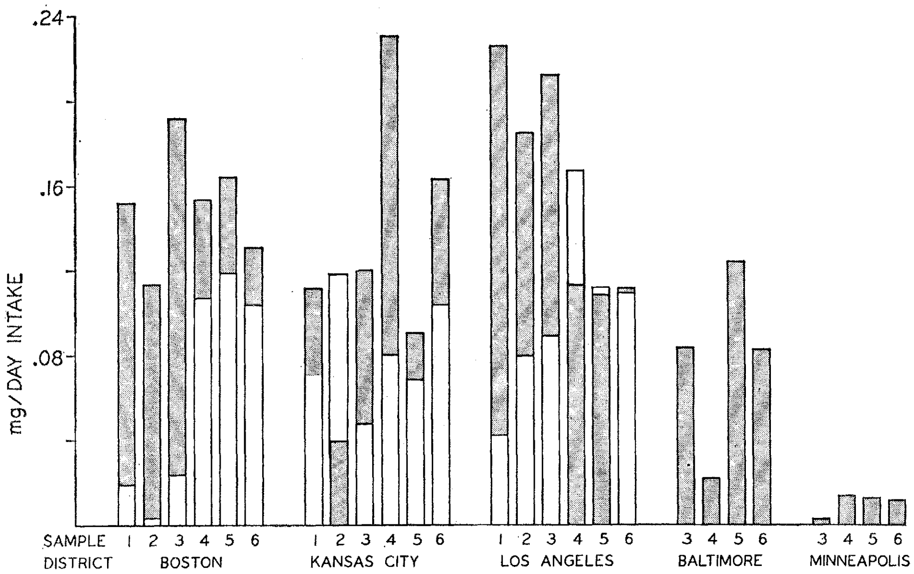

Fig. 2. Total chlorinated pesticide residues by geographic and sample distribution. White bar, 1964; shaded bar, 1965. Sample 1, June; sample 2. August; sample 3, October; sample 4, December; sample 5, February; sample 6, April.

age daily intake of these combined residues is less than 0.02 mg per day with more than half of the individual chemicals present in an amount below 0.001 mg per day.

fic chemicals in the chlorinated organic class of pesticide residues is of greater significance than the sum total present in the diet because of great differences in toxicity among the pesticides. The data in Tables 1 and 2 show a remarkable consistency in the intake of specific chlorinated organic chemicals. There is limited use of TDE and no direct use of DDE as pesticide chemicals. The frequency with which they are found is due to metabolism or conversion from the parent compound, DDT. The DDT and its analogs account for nearly three-fourths of the total calculated daily intake of chlorinated organic compounds based on this study and were found in all food classes except beverages. The DDT alone accounts for about one-third of the total intake.

Residues of dieldrin, lindane, and heptachlor epoxide follow DDT and its analogs in order of frequency. These six compounds account for approximately 85 percent of the total intake. There was no significant change in the frequency and amount of residues from the first to the second year of the study.

The incidence and amount of the remaining 14 chlorinated organic pesticide chemicals are too low to be meaningful as individual residues. The average daily intake of these combined residues is less than 0.02 mg per day with more than half of the individual chemicals present in an amount below 0.001 mg per day.

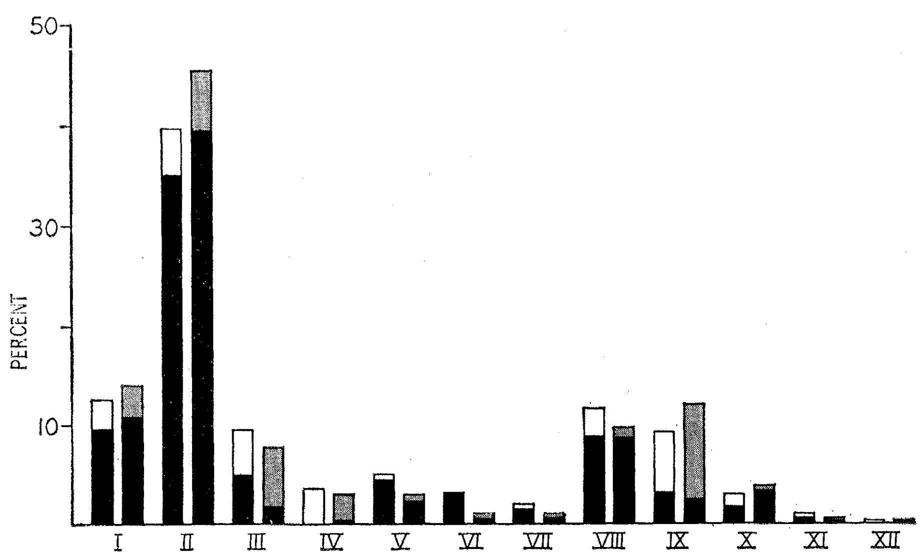

Fig. 3. Distribution of total chlorinated organic pesticides and DDT compounds among food classes. White bar, total chlorinated residues in 1964; shaded bar, total chlorinated residues in 1965; solid bar, DDT and analogs. Roman numerals I to XI are identified in Table 1; XII, beverages.

Organic Phosphate Pesticide Chemicals

No organic phosphate pesticides were detected during the first year of this study (Table 1). It is believed that the findings for this class of compounds during the second year of the study (Table 2) are due more to improved analytical methods than to a change in the residues present. The incidence and quantities of organic phosphate pesticide residues were generally low. A majority of the residues were found in the grain and cereal class. Malathion, the most common residue, was found at an average daily intake of 0.009 mg per day and accounted for over 80 percent of the calculated daily intake of organic phosphates from these samples. The incidence and intake of the remaining organic phosphates detected are too low to be meaningful.

Herbicide and Carbamate Chemicals

The incidence and quantity of herbicide chemicals were quite low and scattered through the food groups. The most common chemical, 2,4-D, was found most frequently in the sugars and adjuncts class and formed about one-third of the calculated daily intake of herbicide over the 2-year period. Residues of MCP and PCP combined account for about half of the calculated intake during the same time span. No significance is attached to the apparent increased intake of 0.01 mg per day during the second year because of the low incidence of herbicide residues in the various food classes.

The incidence and quantity of carbamate chemicals were the most variable observed. Carbaryl was found most frequently and at a calculated daily intake of 0.15 mg and 0.025 mg. The incidence and amount of carbaryl residues were much lower during the second year of the study. The recognition of this chemical as a component of the daily diet is questionable because of its infrequent occurrence.

Dithiocarbamate residues were found on a few samples collected the first year but not on samples during the second year. Inasmuch as dithiocarbamates

Table 2. Daily intake of pesticide residues, listed by food class (22 composites in classes I, II, and X; 26 composites in all others) and expressed in milligrams per day from June 1965 to April 1966. T signifies trace (less than 0.001 mg). The average pesticide intake from beverages (class XII) was 0.019 mg of carbaryl and 0.001 mg of PCP per day. Percentage of fat content was 8 to 13 in dairy products, 17 to 23 in meat, fish, and poultry, and 83 to 88 in oils, fats, and shortenings.

Chemical detected	I Dairy products	II Meat, fish, and poultry	III Grain and cereals	IV Potatoes	V Leafy vege-tables	VI Legume vege-tables	VII Root vege-tables	VIII Garden fruits	IX Fruits	X Oils, fats, and shortenings	XI Sugars and adjuncts
Chlorinated chemicals											
DDT	0.004	0.022	0.002	T	0.002	0.001	0.001	0.006	0.002	0.001	T
DDE	.008	.016	T	T	0.001	T	T	0.001	0.001	0.001	T
TDE	.002	.011	T	T	T	T	T	0.003	T	0.002	
Dieldrin	.002	.003	0.002	T	T		T	T	T	T	T
Lindane		.001	.003	T	T			T	T		
Heptachlor epoxide	.001	.002	T	T	T	T				T	
BHC	.001	.003		T							
Kelthane									0.011		
Aldrin	T		0.001			T		T	0.001		
Endrin				T			T	T		T	
Perthane			.001						T	T	
Thiodan					T			T	T		
Toxaphene					0.001			0.001			
Methoxychlor	T		T								
Tedion		T							T		
TCNB				0.003							
Chlordane								T			
PCNB			T								
Organophosphates											
Malathion			0.008		T		T			0.001	
Diazinon		T	.001		T			T			
Parathion				T	0.001						
Ethion									T		
Ronnel		T									
Herbicides											
PCP	0.001	T	0.001							0.003	
2,4-D					T						0.002
MCP	0.003				0.001						T
CIPC				0.005							
2,4-DB	T		T								
2,4,5-TP	T										
Dacthal					T				T		
Inorganic residues											
Bromides*			1.18	.57					0.002	0.078	0.41
Arsenic (As_2O_3)		0.003	0.001				T	0.001			0.001
Carbamates											
Carbaryl							T		0.006		0.001

* Values reported are those in excess of 25 parts per million.

decompose rapidly, such residues are not expected to be in foods after they are prepared for consumption. Therefore, dithiocarbamates are not recognized as being a regular constituent of the daily intake of pesticide chemicals.

Inorganic Bromides

An average of 80 percent of all food-class composites examined during the 2 years of this study contained some residues of inorganic bromide. The analytical method used does not differentiate between naturally occurring bromide residues and residues resulting from treatment with organic bromide fumigants. Results for the first year indicate an average total bromide intake of 27.0 mg per day, which drops to an average of 14.4 mg per day during the second year. During both years, the highest total bromide residues were found in the grain and cereals class. No specific information is available on the natural bromide content of the food classes examined in this study. Under certain conditions, untreated foods may contain as much as 25 parts per million of bromides (*11*); only those values exceeding 25 parts per million have been included in Tables 1 and 2.

Bromide residues in the range of 0 to 5 parts per million were found in 58.4 and 72.9 percent of the food-class composites during the first and second year of the study, respectively. Residues in the range of 5 to 25 parts per million were found in 33.3 and 24.3 percent of the composites. Residues above 25 parts per million were found in 8.3 and 2.8 percent of the composites.

Arsenic

The incidence and amount of arsenic residues (as As_2O_3) exceeding 0.001 mg per day in food-class composites were quite low. The analytical method used does not differentiate between naturally occurring arsenic and that resulting from application of pesticide chemicals containing arsenic. Tables 1 and 2 show a tenfold difference in the annual intake of arsenic. This difference is due to the arsenic contained in a single sample of potatoes collected in December 1964. There was no general change in the incidence or amount in potatoes or other food classes. The recognition of this chemical from a pesticide chemical source as a component of the daily diet is questionable because of the low incidence of residues.

Daily Intake Levels

This diet is representative of the food consumed by the average 16- to 19-year-old male. The average male (*12*) of this group weighs 69.1 kg (152 lb) and consumes 4.0 kg (8.8 lb) of food and drink daily. This dietary intake represents very great food consumption. The "average" food consumption of the "standard" individual is 2.2 kg (4.8 lb) or slightly more than half the quantity of food specified in our study. Therefore, the calculated daily intake in Tables 1 and 2 must be considered as maximum quantities found in a balanced diet. The average daily intake for the population generally will be about half the maximum; the influence of food classes on the daily intake is significant. For comparison with other values (*13*), the amounts of daily intake found in this study have been converted to milligrams per kilogram of body weight, based on the calculated weight and amount of consumption of the 16- to 19-year-old male.

At a joint meeting of the Food and Agriculture Organization of the United Nations and the World Health Organization Expert Committee on Pesticide Residues, acceptable daily intakes of certain specified chemicals were proposed (*13*).

The following comparison shows the acceptable daily intake and the calculated intake from these samples, respectively, in terms of milligram per kilogram of body weight: DDT 0.01, 0.0005; lindane 0.0125, 0.00006; malathion 0.02, 0.0001; carbaryl 0.02, 0.0012.

During the process of establishing legal tolerances on raw agricultural products, data must be furnished on amounts resulting in acute and chronic toxicity and in "no effect." Among the factors used in establishing tolerances on foods are the proportion of the food in the daily diet and the potential consumption of the pesticide chemical at the tolerance level (*14*). The tolerance concept does not anticipate, as a practical matter, that all foods will contain residues at the tolerance level for all chemicals for which a tolerance has been established, or even that all of a single food will always contain a residue at the tolerance level. Actual experience has proved this to be a valid concept. Such contributions to the dietary intake are considered safe, based on our present knowledge. Daily intake of specific pesticides, based on established tolerances where acceptable daily intakes have not been proposed, have been calculated to milligrams per kilogram of body weight. The following comparison shows the daily intake calculated from tolerances and the intake calculated from these samples, respectively, in terms of milligram per kilogram of body weight: dieldrin 0.006, 0.00009; heptachlor epoxide 0.0006, 0.00004; 2,4-D 0.017, 0.00005; total bromides 1.45, 0.30; arsenic (as As_2O_3) 0.049, 0.001.

The calculated values for arsenic and bromides include all findings and represent the natural content and any additions resulting from applications of pesticide chemicals.

Summary and Conclusions

Residues of chlorinated organic pesticide chemicals were commonly found in all diet samples and all food classes within samples except beverages at a daily intake of 0.0014 mg per kilogram of body weight. Meat, fish, and poultry were the major source of pesticide residues and, when combined with dairy products, account for more than half of the intake of chlorinated organic pesticide chemicals. Fruits, garden fruits, and grain foods each accounted for about 10 percent of the intake of chlorinated pesticides. The DDT, its two analogs, dieldrin, lindane, and heptachlor epoxide account for 85 percent of the total intake of chlorinated pesticides. A single pesticide, DDT, accounts for one-third of the total. There was no statistically significant change in frequency or quantities of these compounds during this study. Frequency and quantities of the 14 additional chlorinated pesticides were too low to be meaningful.

Organic phosphate pesticide chemicals not found in the first year of study are reported in the second year at a daily intake of 0.01 mg. The most common residue, malathion, accounts for more than 80 percent of the daily intake of this class of chemicals. The frequency and quantity of other organic phosphates are too low to be meaningful. Herbicide chemicals were found infrequently; they averaged about 0.01 mg, of which one-third was 2,4-D and one-half was MCP and PCP combined.

Carbaryl was found infrequently during both years, and the amount dropped from 0.15 to 0.025 mg per day for the second year. Dithiocarbamate findings were too limited to consider these pesticide residues a regular constituent of the diet. Arsenic and bromides were found at daily intake at or below the maximums found as naturally occurring elements in foods. This study was based on an amount of food consumption almost double that of an average individual. The findings on these samples are much lower than the acceptable daily intake or potential consumption at the tolerance level for DDT, lindane, malathion, carbaryl, dieldrin, heptachlor epoxide, 2,4-D, bromides, and arsenic.

The residues of pesticide chemicals consumed in a normal well-balanced diet are substantially below the limits set for acceptable daily intakes by the World Health Organization and United Nations Committees and below the safe levels anticipated when legal tolerances are established on the raw foods. Although this conclusion is reassuring, it cannot be said that pesticide residues in foods are not a matter for continued concern. Unpublished data, obtained from objective samples of raw food examined to determine compliance with tolerances, show (i) that for each of the last 3 years almost half the lots examined contain residues, mostly below tolerance levels, and (ii) that over half these lots contain more than one residue. Even though this investigation does not show a statistically valid difference, there is a finite increase in the daily intake of total chlorinated organic compounds. From these findings, we conclude that continued surveillance and attention to pesticide residues in foods are required.

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