Environmental Health Perspectives
Vol. 19, pp. 83–87, 1977

Levels of Arsenic in the United States Food Supply

by C. F. Jelinek* and P. E. Corneliussen*

At the present time, the Food and Drug Administration (FDA) accords the highest priority to mercury, lead, cadmium, arsenic, selenium, and zinc in its program on toxic elements in foods. The only regulatory levels for arsenic in foods in the U. S. are the tolerances which have been established for its residues in specified foods, resulting from the application of arsenical pesticides on food and feed crops and from animal feed additives. FDA has monitored for arsenic in its Total Diet Survey since the inception of this program. The data from this program indicate that the average daily intake for arsenic (as As_2O_3) has decreased from about 130 μg/day in 1968 to about 20 μg/day in 1974. Most of the arsenic is found in the meat-fish-poultry food class of the total diet. In individual foods, the highest levels were found in fish, with a mean level of about 1.5 ppm (as As_2O_3) in the edible portion of finfish. Much lower levels were found in all the other food types analyzed; of these, the highest levels found were a mean level of 0.08 ppm in chicken and 0.16 ppm in rice. FDA toxicologists do not believe that the average daily intake of arsenic, or its levels in the different food commodities, pose a hazard to the consumer.

The Food and Drug Administration has conducted various studies on toxic elements in foods for many years. At the present time, the Agency accords highest priority to mercury, lead, cadmium, arsenic, selenium and zinc.

This report summarizes the results FDA has obtained in recent years in monitoring for arsenic in foods. Neither the toxicological aspects of arsenic nor details of analytical methodology will be discussed in detail, but will be referred to only as they are related to our monitoring activities.

The only regulatory levels for arsenic in foods in the U. S. are the tolerances that have been established for its residues in specified foods, resulting from the application of arsenical pesticides on food and feed crops and from animal feed additives. Generally, the tolerance for arsenic residues (as As_2O_3) on various fruits and vegetables resulting from pesticidal use of copper, magnesium and sodium arsenates is 3.5 ppm As_2O_3 (Table 1). Tolerances for residues resulting from the use of lead arsenate, sodium arsenite, and the sodium salts of methane arsonic acid and of cacodylic acid are also given.

A number of arsenic compounds are also permitted for use in food producing animals either as growth stimulants or for prevention or treatment of diseases. Among these are arsanilic acid and its sodium salt, 4-nitrophenylarsonic acid, 3-nitro-4-hydrophenylarsonic acid, and 4-nitrophenylarsonic acid. Tolerances for total combined arsenic residues (calculated as As) resulting from these uses are given in the Code of Federal Regulations, Title 21, part 556.60. Essentially, the tolerances are as follows: 0.5 ppm arsenic in eggs; 0.5 ppm in the muscle of chickens, turkeys, and swine; and 2 ppm in edible by-products of chickens and turkey and the livers and kidneys of swine. The tolerance for arsenic in other edible by-products in swine is 0.5 ppm. A 5-day withdrawal period is required when arsenicals are used in these species to allow for excretion, so that residue levels in the tissues will not exceed these tolerances.

The Food and Drug Administration has monitored for arsenic in its Total Diet ("market basket") Survey since the inception of this program, to determine the levels of this element in the average diet. The composition of the "market basket" used in FDA's Total Diet Program (*1*) is based in large part on the survey of U. S. household food consumption conducted by the U. S. Department of Agriculture (USDA) in 1965. Diet guides are provided for each of four geographical regions of the U. S. as defined by USDA: South, Northeast, North Central, and West. Each market basket rep-

*Bureau of Foods, Food and Drug Administration, 200 C St., S.W., Washington, D. C. 20204.

Table 1. Tolerances for residues of arsenic-containing pesticides.

Code of Federal Regulations, Title 40	Pesticide	Tolerance
Part 180.192	Calcium arsenate	3.5 ppm As_2O_3, numerous fruits and vegetables
180.193	Copper arsenate	3.5 ppm As_2O_3, numerous vegetables
180.194	Lead arsenate	7 ppm lead, numerous fruits and vegetables 1 ppm lead, citrus fruits
180.195	Magnesium arsenate	3.5 ppm As_2O_3, beans
180.196	Sodium arsenate	3.5 ppm As_2O_3, grapes
180.289	MSMA (methanearsonic acid, monosodium salt)	0.7 ppm As_2O_3, cottonseed
	DSMA (methanearsonic acid, disodium salt)	0.35 ppm As_2O_3, citrus fruit
180.311	Cacodylic acid (dimethylarsenic acid)	2.8 ppm As_2O_3, cottonseed 1.4 ppm As_2O_3, beef kidney and liver 0.7 ppm As_2O_3, cattle meat, fat, by-products, other
180.335	Sodium arsenite	2.7 ppm As_2O_3, beef and horse kidney and liver 0.7 ppm As_2O_3, beef and horse meat, fat and by-products, other

resents the typical 2-week diet of a 15 to 20-year-old male for the region of the country in which it is collected. The total diet samples are collected from retail markets in representative urban areas of the U. S., and are shipped to FDA's Kansas City District laboratory. Food samples which require cooking or other processing before consumption are then prepared in a dietary kitchen as they would be in a household. Composites of the 12 Total Diet categories listed in Table 2 are prepared by the FDA Kansas City laboratory, and each of the composites is analyzed for the residues specified. All arsenic findings are calculated as As_2O_3 because residue tolerances for most arsenic-containing pesticides are expressed in terms of As_2O_3. As_2O_3 can be converted to As by multiplying by 0.76.

The average levels of arsenic (as As_2O_3) found in the different food composites from 1967 to 1974 are shown in Table 2. It can be seen that the levels in all food categories have decreased significantly since 1967–1970, to the extent that the meat-fish-poultry composites now are the only major source of arsenic in the total diet.

Table 3 shows the estimated average daily intake of arsenic (as As_2O_3) represented by the different food composite categories. Again, the levels of arsenic intake from all the food categories have significantly decreased, with the meat-fish-poultry category contributing most to the daily intake. It is believed that much of this drop may be due to the decreased use of arsenic-containing pesticides on food crops since the late 1960s. To explain the apparent increase in arsenic levels in the meat-fish-poultry and fruit categories from 1973 to 1974, it will be necessary to evaluate the data from the Total Diet Studies for several more years to determine whether this represents a trend, or if it is merely the random variation one may expect from year to year.

The estimated average daily intake of As_2O_3 from the total diet has varied from about 100 μg/day in 1967–1969 to about 10–20 μg/day in 1971–1974. FDA toxicologists believe that these levels do not represent a hazard to the consumer, but that they do warrant continued surveillance. On a priority basis, mercury, lead, and cadmium have been of more concern to FDA in recent years than has arsenic.

To obtain a better idea of the levels of arsenic in individual food items, during the past three years (Fiscal Years 1974–1976), FDA has carried out exploratory surveys for arsenic in various food commodities in interstate commerce, which are obtained primarily at the retail level.

The data obtained on arsenic levels in meats, eggs, and milk are shown in Table 4. The mean levels in meats were all less than 0.1 ppm As_2O_3. The highest levels were found in chicken, with a mean value of 0.08 ppm and a maximum level of 0.5 ppm. Generally the levels were about the same in the samples of muscle and liver examined.

Table 2. Average levels of arsenic, total diet composite (as As_2O_3).[a]

As, ppm

Food class composite	Fy 1967	Fy 1968	Fy 1969	Fy 1970	Fy 1971	Fy 1972	Fy 1973	Fy 1974
Dairy products	0.017	0.010	0.007	0.007	ND	ND	0.003	ND
Meat, fish, and poultry	0.110	0.166	0.141	0.180	0.057	0.046	0.020	0.059
Grain and cereal products	0.011	0.063	0.027	ND	ND	ND	0.003	ND
Potatoes	0.015	0.026	0.010	0.007	0.003	ND	0.003	ND
Leafy vegetables	0.013	0.026	0.013	ND	ND	ND	T	ND
Legume vegetables	0.009	0.007	0.010	ND	ND	ND	T	ND
Root vegetables	0.012	0.007	0.010	0.007	ND	ND	T	T
Garden fruits	ND	0.020	0.013	0.007	ND	ND	ND	T
Fruits	0.013	0.047	0.017	0.007	0.007	ND	T	0.017
Oils, fats, and shortening	0.007	0.020	0.007	ND	ND	ND	T	ND
Sugars and Adjuncts	0.015	0.013	0.017	ND	ND	ND	T	ND
Beverages (including water)	0.008	0.020	0.010	ND	ND	ND	ND	ND

[a]ND = not detected; T = <0.001 ppm As_2O_3 average for the given food class composite in the given year (30 composites of each of the 12 food classes each year). The limit of quantitation for a single analysis was approximately 0.1 ppm (As_2O_3). Detections below that level are estimations only.

Table 3. Average daily intake of arsenic, total diet.[a]

As_2O_3, μg/day

Food class composite	Fy 1967	Fy 1968	Fy 1969	Fy 1970	Fy 1971	Fy 1972	Fy 1973	Fy 1974
Daily products	13	8	5	6	ND	ND	2	ND
Meat, fish, and poultry	29	45	34	48	15	12	6	16
Grain and cereal products	4	29	11	ND	ND	ND	1	ND
Potatoes	3	7	2	1	T	ND	T	ND
Leafy vegetables	1	2	1	ND	ND	ND	T	ND
Legume vegetables	1	1	1	ND	ND	ND	ND	ND
Root vegetables	1	1	T	T	ND	ND	T	T
Garden fruits	ND	4	1	1	ND	ND	ND	T
Fruits	4	12	4	1	2	ND	T	4
Oils, fats, and shortenings	1	2	T	ND	ND	ND	T	ND
Sugars and adjuncts	1	2	1	ND	ND	ND	T	ND
Beverages (including water)	10	24	15	ND	ND	ND	ND	ND
Total	68	137	75	57	17	12	10	21

[a]ND = Not detected. T = <1 μg As_2O_3/day. Detections below that level are estimates only.

The U. S. Department of Agriculture (USDA) conducts a continuing monitoring program for various contaminants in red meat and poultry obtained in slaughterhouses. USDA has the responsibility for assuring the safety of meat products in the U. S. and has, therefore, examined many more meat samples than FDA has. The mean levels of arsenic that USDA found in chicken, swine, and cattle meat products in 1975 agreed fairly well with the FDA findings presented in Table 4. One difference was that USDA found somewhat higher levels in liver than in muscle; the highest was an average of about 0.8 ppm arsenic as As_2O_3 in chicken livers. Actually, we would expect to find somewhat higher values in the liver, since it is the target organ for arsenic. As in the FDA survey, levels in chickens were higher than those found in swine or cattle. The higher levels in chickens may result partially from

Table 4. Arsenic (as As_2O_3) in meats, eggs, milk, FDA surveys.[a]

Product	Number of samples	As, ppm Mean	As, ppm Range
Beef, muscle and ground	56	0.02	ND–0.1
Beef, liver	24	0.03	ND–0.05
Chicken, muscle	21	0.08	ND–0.5
Chicken, liver	6	0.08	ND–0.4
Pork, muscle	29	0.02	ND–0.1
Pork, liver	2	0.02	ND–0.05
Frankfurters, sausages	28	0.03	ND–0.2
Protein meat extender	22	0.04	ND–0.1
Eggs	28	0.03	ND–0.2
Milk	29	0.02	ND–0.2

[a]Limit of quantitation approximately 0.1 ppm As_2O_3 in an individual analysis. Detections below that level are estimations only.

the greater use of arsenical additives in poultry feed.

Table 5 presents the results that FDA obtained for arsenic levels (as As_2O_3) in vegetable and fruit products in the 1974–1976 period. The mean levels in all types examined were well below 0.1 ppm. The highest value in any single sample was 0.3 ppm As_2O_3 in a potato product.

Table 5. Arsenic (as As_2O_3) in fruit and vegetable products.[a]

Product	Number of samples	As, ppm Mean	As, ppm Range
Applesauce	28	0.03	ND–0.1
Tomatoes, fresh and canned	28	0.03	ND–0.1
Carrots and carrot products	28	0.02	ND–0.1
Lettuce	28	0.02	ND–0.05
Pork and beans	28	0.03	ND–0.1
Onions and onion products	256	0.02	ND–0.1
Potatoes and potato products	268	0.01	ND–0.3

[a]Limit of quantitation approximately 0.1 ppm As_2O_3 in an individual analysis. Detections below that level are estimations only.

The levels obtained for arsenic (as As_2O_3) in cereal, nut, and sugar products are shown in Table 6. The mean values are again all well below 0.1 ppm, with the exception of rice, where the mean level was 0.16 ppm. The highest individual level in the foods in Table 6 was 0.4 ppm in a sample of rice.

During Fiscal Years 1973 and 1974, the Food and Drug Administration carried out a comprehensive fish survey to determine the levels of pesticides, PCBs, and heavy metals in fish. The species were selected either on the basis of their commercial significance, or on the fact that bottom feeders are indicators of pollution. The samples were generally obtained at the wholesale level in order to determine the quality of fish available to the consumer. All analyses were carried out on the edible portion of the fish. Arsenic was added to this surveillance program in 1974. However, this survey was cut short in order to divert resources to the botulinum-in-mushrooms crisis at that time. As a result, only 105 samples were analyzed, all of which were finfish, except for 10 samples of shrimp.

Table 6. Arsenic (as As_2O_3) in cereal, nut, sugar products.[a]

Product	Number of samples	As, ppm Mean	As, ppm Range
Flour	15	<0.01	ND–0.05
Corn meal	29	0.04	ND–0.2
Rice	27	0.16	ND–0.4
Breakfast cereals	29	0.04	ND–0.2
Bakery products	202	0.01	ND–0.05
Protein meat extender	22	0.04	ND–0.1
Peanut butter	27	0.03	ND–0.2
Sugar	28	0.04	ND–0.3
Soft drinks	18	0.04	ND–0.1

[a]Limit of quantitation approximately 0.1 ppm As_2O_3 in an individual analysis. Detections below that level are estimations only.

FDA also conducts a continuing surveillance program for residues of heavy metals in samples of oysters and clams obtained from approved commercial harvesting areas.

Table 7 shows the results obtained by FDA in its Fiscal Year 1974 Comprehensive Fish Survey and in its Fiscal Years 1974 and 1975 Heavy Metals in Shellfish Program. Since it was possible to analyze only 105 samples in the comprehensive fish survey, we do not feel that we can draw conclusions about species likely to have higher arsenic contents, and thus we have classified the species from this survey only as "finfish" and "shrimp."

Table 7. Arsenic (as As_2O_3) in finfish and shellfish.[a]

Species	Number of samples	As, ppm Mean	As, ppm Range
Finfish	95	1.47	ND–19.1
Shrimp	10	0.67	0.3–1.5
Eastern oyster	627	0.09	ND–1.2
Pacific oyster	86	0.07	ND–0.4
Soft shell clam	164	0.14	ND–0.9
Quahog	225	0.12	ND–1.4

[a]Limit of quantitation approximately 0.1 ppm As_2O_3 in an individual analysis. Detections below that level are estimates only.

As can be seen, there is very wide variation between the levels found for individual samples. The

highest finding in an individual sample of finfish (19 ppm) was more than 10 times the mean level (1.47 ppm). Similarly, some individual samples of molluscs contained more than 10 times as much arsenic as the mean level for the species.

With respect to the results for arsenic in clams and oysters, we should point out again that all samples were obtained from approved commercial harvesting areas. We do not know whether the same general levels would be found in molluscs in all areas where they may be caught.

The nature of the sampling in FDA's comprehensive fish survey precludes the ability to determine the precise origin of catch, since entire lots of fish are sampled primarily at the wholesale level.

Even though analysis of a lot is an appropriate method of determining the average intake of arsenic for the general public, it will be beneficial to learn more about the levels of specific species caught in specific locations. In this regard, surveys being conducted by the National Marine Fisheries Service will provide valuable additional information, since their sampling population is much larger, and they obtain their samples at specified locations.

In summary, the total diet monitoring program carried out by FDA since 1967 shows that the average daily intake of arsenic (as As_2O_3) has decreased drastically from about 130 μg/day in 1968 to about 20 μg/day in 1974. Most of the arsenic is found in the meat-fish-poultry composite of the total diet survey. In analyses carried out on individual foods, the highest levels were found in fish, with a mean level of 1.47 ppm in finfish. Much lower levels were found in all other food types; of the latter, the highest levels found were a mean level of 0.08 ppm in chicken and 0.16 in rice. We do not believe that this average daily intake, or the levels in the various foods, pose a hazard to the consumer. However, we do believe that continued surveillance for arsenic in our food supply is necessary.

For the future, FDA plans (1) to continue to measure arsenic in its total diet studies to determine whether there are any trends in arsenic levels in the average U. S. diet, and to follow up on any unusually high total diet findings; and (2) to carry out special surveillance programs on individual food commodities when unexpected levels warrant.

REFERENCES

1. Duggan, R. E., and McFarland, F. J. Residues in fish and food. Pestic. Monit. J. 1: 1 (1967).