A Survey of Radioactive Residues in Foods Before and After 1945: Evidence of Possible Fallout Contamination*

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Evidence based on a number of reports indicates that certain foodstuffs, notably dairy products, are receiving radioactive contamination from tropospheric fallout. Presumably, this invasion of the food chain by man-made radioactivity occurs in three ways: (a) direct deposition of the fallout on leaves and fruits of edible plants; (b) translocation by the plants of the fallout which has become mixed with soil and water; (c) consumption of contaminated plant materials by animals whose tissues and other products are subsequently used for food.

This report covers results of a survey conducted over the past two years to determine to what degree foods may have become contaminated with radioactive fallout. It has been assumed that no significant man-made radioactive contamination could have occurred prior to 1945, the year the first experimental and military nuclear devices were exploded. Foods produced before and after this critical date have therefore been examined for total beta radioactivity adjusted for the presence of potassium 40, a widely distributed naturally occurring radioactive isotope. It is possible that other naturally occurring radioactive substances may contribute to the total. This contribution is extremely small, and while it may vary from food to food there is no reason to expect it to vary with time. Consequently if we consider the radioactive content of all pre-1945 foods as a base line, any increase in foods produced since 1945 can be interpreted as man-made radioactive contamination. This contamination is presently contributed mainly by fallout from weapons testing, but it can be expected also to reflect the presence of nuclear power plants and other applications.

In January 1957, in response to requests by the Food and Drug Administration, nearly a thousand samples of food antedating 1945

were submitted. These foods came from private homes and the food industry; some even from the caches of the Shackleton and Byrd Antarctica expeditions dating back to 1906. In addition, an equal number of post-1945 samples were collected, mostly from retail outlets.

The following categories were examined for total beta radioactivity: vegetables, fruits, fruit juices, sea foods, dairy products, bread, meat products, wheat, sugars, jams and jellies, cocoa beans, tea, and coffee. Nearly half of the samples were fruits and vegetables, and the predominant number of these were canned. Of the post-1945 samples, the largest number originated in the first five years; comparatively smaller numbers of samples came from the years 1950 to 1957.

Experimental

Methodology

1. Sample preparation.—All foods were well mixed or homogenized to insure uniform distribution of any radioactivity present. An amount of sample in its original state of hydration or dryness (as is) was weighed out so as to yield approximately 200 to 500 mg of ash. Dry ashing was done in several kinds of containers (Vycor, porcelain, silica, glass, platinum, etc.) and care was taken to retire any vessels whose surface had become severely etched. Ashing was usually allowed to proceed for 18 to 24 hours at a temperature of about 550°C. Unoxidized carbon was removed by wetting down the ash with small quantities of water and repeated heating. Wherever possible, fusion of the ash was avoided because of the attendant difficulties of removal from the ashing vessel. The ash yield was weighed to the nearest milligram, and its relation to the original sample calculated as an ash ratio (mg ash per mg of original food). Ash samples were finely pulverized in an agate mortar and stored in stoppered glass vials.

2. Radioactivity measurements.—Total beta radioactivity was measured with a Tracerlab superscaler equipped with an automatic sample

changer and print-out which recorded the time necessary to accumulate 600 counts from each sample. The Geiger tube sensing element was a Tracerlab TGC 14 carbon-counter with essentially 100% response to any beta particles entering, but of low response to gamma photons. This tube was provided with a thin, aluminized mylar window (0.9 mg/cm²) and the internal atmosphere was maintained with a mixture of 99% helium and 1% isobutane (Geiger gas), flowing at the rate of one bubble per second. Because of the small sensitive volume and additional shielding within the lead pig, the background response of the tube averaged 11 counts per minute (c/m).

All ash samples were measured in 1″ stainless steel planchettes at fixed geometry. One hundred mg of ash was used for each determination and special care was taken in adjusting the thickness and surface uniformity of the sample. One standard and two empty (background) planchettes spaced 180° apart were carried with each revolution of the 25 position sample changer. Depending on the amount of radioactivity present, the time for one revolution of the turntable varied from 12 to 24 hours.

3. Standard.—The reference standard was potassium chloride. According to Nier (1) potassium contains 0.011% of K⁴⁰, the naturally occurring radioactive isotope, and so far as can be ascertained, this species is uniformly distributed in nature. Suttle and Libby (2) have determined the absolute numbers of beta and gamma emissions to be, respectively, 29.6 disintegrations and 2.96 disintegrations per gram of the metal. Because of the low counting efficiency of the Geiger tube for gamma photons, this component was ignored, and the standard was applied as a pure beta standard.

One hundred milligrams of dried and finely powdered reagent grade KCl was used as the working standard. This amount, deposited in a 1″ stainless steel planchette, reproduced the geometry of all ash samples very closely. The mass absorption error of 100 mg of salt was of the order of 5%. The density of most ash samples closely approached that of KCl; hence, mass absorption error of the ash was assumed to be of the same order. For ash weights greater or less than 100 mg, corresponding weights of KCl standards were applied.

As derived from 29.6 disintegrations/second/g of potassium, 100 mg of KCl produces 93.13 disintegrations/min. A number of determinations of 100 mg samples of KCl has established that the Geiger tube "sees," on the average, 28.9 counts/min. This is an overall efficiency of

31%. In expressing all readings on ash samples in terms of the potassium standard, counts per min. (c/m) have been converted to disintegrations per min. (d/m) by multiplying c/m by an average factor of 3.22.

Potassium Analyses

All ash samples were analyzed for potassium (flame photometer) with an accuracy of ±5%. Milligrams potassium per gram of original sample were converted to disintegrations per minute per gram by multiplying by 1.776. This is the factor derived from the data published by Suttle and Libby (2).

Net Radioactivity

This was derived by subtracting from the total radioactivity the contribution due to the presence of potassium. On the average the net value approached zero except in those cases where other radioactive substances or fission products were present. Wherever the term "total beta radioactivity" has been used subsequently, a net or potassium corrected value is meant.

Errors

1. Mass absorption.—From experiments with a variety of ash samples wherein radioactivity as read was plotted against varying weights of sample, it was determined that mass absorption at 100 mg was of the order of 5%. However, since a 100 mg KCl standard also exhibited mass absorption of the same order, these errors were considered to cancel out. Hence with strictly fixed geometries, no corrections for mass absorption were applied.

2. Counting.—Theoretically, the probable counting error for 600 counts, regardless of the time necessary to accumulate this number, is ± 3.2%. However, an error of this low order obtains only when the ratio of total count to background is 10 to 1 or better. Such a favorable situation rarely occurred because most of the samples were low in radioactive content. Generally the ratios varied from 1:1 to 4:1. Under these conditions the probable error was much greater, and at times as great as ± 20%.

3. Net radioactivity.—Errors in measuring net radioactivity are not directly determinable, but it is clear that they will be significantly influenced by the errors of the analytical operations, viz., (a) flame photometric determination of potassium, and (b) total radioactivity. In extreme instances the errors may be additive and a range of error of ± 25% is possible.

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Table 1. Average total beta radioactivity of fruits and fruit products harvested before and after 1945

Fruit[a]	No.	Before 1945 d/m/g	s.e.(±)	No.	From 1945 to 1957 incl. d/m/g	s.e.(±)
Pears	36	0.01	0.033	12	0.00	0.056
Cherries	44	0.00	0.042	17	0.16	0.037
Peaches	51	0.00	0.040	38	0.04	0.053
Apricots	11	0.22	0.089	5	0.00	0.111
Apples[b]	19	0.10	0.032	9	0.00	0.073
Prunes and plums	40	0.24	0.054	21	0.00	0.071
Tomatoes[c]	83	0.01	0.047	51	0.00	0.051
Miscellaneous[d]	36	0.01	0.054	21	0.00	0.080
Berries[e]	60	0.04	0.041	13	0.00	0.084
Citric fruit juices[f]	17	0.00	0.047	21	0.03	0.054
Fruit juices, other[g]	29	0.06	0.056	11	0.00	0.046

[a] Exclusive of dried fruits.
[b] Includes apple sauce.
[c] Includes juices, relishes, catsup, soup.
[d] Includes grapes, pineapple, rhubarb, currants, pumpkins, quinces, figs, olives, nectarines, but no citrus fruits.
[e] Includes predominantly strawberries, raspberries, and blackberries.
[f] Includes lemon, lime, grapefruit, and orange.
[g] Includes predominantly grape and apple.

Sensitivity

The counting equipment was capable of detecting one count per minute total beta radioactivity above background in a 100 mg sample of ash. Depending on the ash content, this represents from 0.02 to 0.5 c/m in terms of the original food.

Results

Table 1 gives the results for fruits and fruit products. No significant increase in radioactivity of the post-1945 over the pre-1945 samples is demonstrated. Reference to the raw data from which these averages were constructed shows that a predominant number of samples originated in the years 1945 to 1950, inclusive. During this period nuclear weapons testing with resultant fallout had not yet reached the proportions that followed in later years. It might be assumed therefore

that the conclusions indicated from Table 1 should be subject to revision.

Accordingly, the data were analyzed by comparing the average of all samples collected in 1945 with the average of all samples collected in 1956 and 1957. For 78 samples collected in 1945 the average was 0.05 d/m/g and for 44 samples collected in 1956 and 1957 the average was 0.00 d/m/g. No significant increase in radioactivity by year could therefor be demonstrated.

Since only a very small number of dried fruit samples were available for analysis, no significant comparisons between pre- and post-1945 periods could be made. There is some indication, however, that a trend toward increased radioactivity in current samples of dried fruits may be occurring.

Table 2 gives the results for vegetables. No significant increase in radioactivity of the

Table 2. Average total beta radioactivity of vegetables harvested before and after 1945

Vegetable	No.	Before 1945 d/m/g	s.e.(±)	No.	From 1945 to 1957 incl. d/m/g	s.e.(±)
Potatoes	20	0.13	0.185	9	0.00	0.192
Corn	43	0.00	0.062	47	0.01	0.047
Beans[a]	79	0.00	0.043	53	0.05	0.096
Peas	35	0.00	0.064	57	0.06	0.042
Beets and turnips	28	0.00	0.093	12	0.03	0.104
Carrots	21	0.00	0.113	19	0.06	0.116
Spinach[b]	10	0.01	0.125	17	0.00	0.119
Miscellaneous[c]	54	0.00	0.050	25	0.11	0.080

[a] Includes lima, soy, etc.
[b] Three samples, only, from year 1956 showed an average value of 0.30 d/m/g.
[c] Includes asparagus, mustard greens, onions, pimentos, okra, mushrooms, squash, cabbage, broccoli, cauliflower.

**Table 3. Average total beta radioactivity of miscellaneous foodstuffs
produced before and after 1945**

| Foodstuff | Before 1945 | | | From 1945 to 1957 incl. | | |
	No.	d/m/g	s.e.(±)	No.	d/m/g	s.e.(±)
Wheat[a]	19	0.00	0.151	28	0.01	0.087
Sugar and jams[b]	21	0.00	0.049	17	0.03	0.005
Meat products[c]	13	0.19	0.111	13	0.00	0.125
Bread[d]				13	0.00	0.119
Cocoa beans[d]				16	0.33	0.234
Misc. sea food[e]	16	0.00	0.130	12	0.00	0.196
Coffee[d]				8	0.00	0.874

[a] The unprocessed wheat grain.
[b] Includes sirup and a variety of fruit jams.
[c] Includes beef, pork, poultry, lamb; also dried and cured products.
[d] Fresh: All 1957; no pre-1945 samples available.
[e] Includes shrimp, crab, lobster.

post-1945 over the pre-1945 samples is demonstrated. Again it was noted from the raw data that the preponderance of samples was drawn from the years 1945 to 1950. Taking the 107 samples originating from the year 1945 and comparing with 46 samples from the years 1956 and 1957, the averages are as follows: 1945, 0.05 d/m/g; and 1956 and 1957, 0.00 d/m/g. There is no trend toward increased radioactivity in current samples. Unfortunately, only a very limited number of vegetables are represented by the leafy variety, such as cabbage and lettuce; therefore, in the light of recent findings (3) further analyses must be conducted to confirm present trends (1948 *et seq.*).

Table 3 lists results for a number of miscellaneous foodstuffs. No significant increase in radioactivity is demonstrated for wheat, sugar and jams, meat products, and miscellaneous sea foods. No pre-1945 samples of bread, cocoa beans, and coffee were obtainable; hence a comparison is not possible. However, unless cocoa beans store some other natural radioactive substance, it is possible that this product reflects contamination from fallout. Further work is necessary.

With respect to wheat which has been shown to be radioactive in certain areas of Minnesota in the growing years of 1956, 1957, and 1958, it should be emphasized that practically all our post-1945 samples came from the 1956 harvest. Of these, 16 samples originated from California, 3 from Texas, 2 from New York, and one each from Indiana and Michigan. This may account, therefore, for our non-confirmation of increased contamination in this product to date.

The products which have shown significant increases in radioactivity, or at least well defined trends, are shown in Table 4. Of these, the dairy products show a highly significant increase in total radioactivity in the post-1945 period. Tea shows an outstanding increase. When plotted by years, the individual values for the dairy products described a line having a significant positive regression. A similar plot for shellfish is shown in Fig. 1.

Identification studies on two samples of tuna fish which exhibited marked elevation in total radioactivity showed that the principal nuclide was zinc 65. Two samples of tea, similarly studied, revealed the presence of significant amounts of zirconium 95. These analyses were made by gamma spectrometry (4).

Two samples of tea showing relatively high total beta counts were subjected to radiochemical analyses (5). Of the total beta radioactivity in one sample, 6.7% and 4.5% was accounted for, respectively, by total

**Table 4. Foodstuffs which have shown
increased total radioactivity when
compared with pre-1945 samples**

| Foodstuff | Before 1945 | | From 1945 to 1957 incl. | |
	No.	d/m/g	No.	d/m/g
Fish[a]	25	0.00	26	0.32[b]
Shellfish[c]	15	0.00	32	0.36[d]
Dairy products[e]	26	0.00	46	0.55[f]
Tea[g]	36	0.00	88	31.4

[a] Includes mainly salmon, sardines, and tuna fish.
[b] Significance level, p = 0.1.
[c] Includes oysters, clams, and mussels.
[d] Significance level, p = 0.1.
[e] Includes fluid, dry milk, cheese.
[f] Significance level, p = 0.01.
[g] Tea leaves only, not the beverage.

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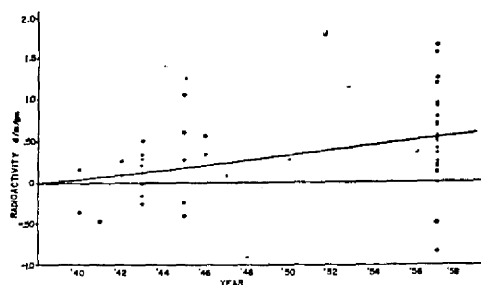
Fig. 1—Change in radioactivity of shellfish.

strontium (89 and 90) and cesium 137. In the other sample the percentages were, respectively, 8.4 and 4.9.

Because of the surprisingly high radioactivity in the tea, it became of interest to determine how much radioactivity would be conveyed to the beverage. Under conditions designed to produce an infusion approximately 10 times the strength of that commonly consumed, (assume normal brew 1 g/100 ml) 100 grams of infusion was made: (a) Sample 13 contained 154 d/m/g tea leaves; the extract from it, 10% of the original radioactive concentration, (b) Sample 21 contained 34 d/m/g tea leaves; the extract

Table 5. Resume of total beta radioactivity of tea for August, September, October, November, and December, 1958

Country	No. of Samples	Total Beta Radioactivity per Gram Tea Leaves d/m	Micro-micro-curies
	Far East		
Japan	66	79	36
Formosa	104	30	14
India	59	28	13
Malaya	8	28	13
Ceylon	30	13	6
Sumatra	22	11	5
Indonesia	20	10	5
Java	39	7	3
Viet Nam	4	6	3
	South America		
Brazil	37	12	5
Peru	4	10	5
	Africa		
Kenya	8	11	5
Portuguese East Africa	4	8	4
Belgian Congo	4	6	3
Tanganyika	4	7	3
	Middle East		
Iran	14	9	4

from it, 5% of the original radioactive concentration. From these results it appears that a relatively small transfer of radioactivity from the leaves to the infusion occurs.

Conclusion

The results presented in this report comprise approximately 2000 samples, half of which originated before 1945. Of those sampled after 1945 until the end of 1957,
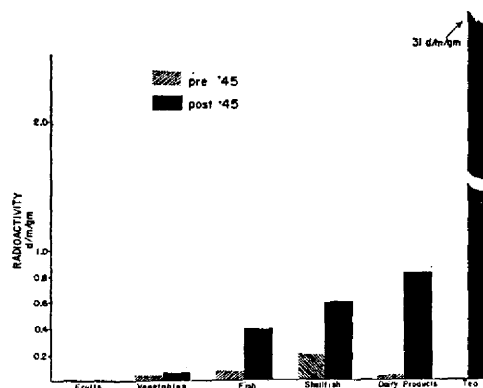

Fig. 2—Increase in radioactivity of certain foods since 1945.

(the cutoff date for all samples except tea) it can be seen that fish, shellfish, dairy products, and tea showed significant increase of radioactivity above the 1945 base line. These results are graphically summarized in Fig. 2. It can be seen that fruits and vegetables showed no noteworthy increases in radioactivity. Since nearly all of the latter had been processed by canning or freezing, there is some question whether significant removal of surface contamination may have occurred.

Addendum

At the time this report was made to the A.O.A.C. in October 1958, a total of 88 samples of tea from various parts of the world had been analyzed; they gave an average total beta value of 31 d/m/g (see Table 4). As of January 1, 1959, nearly 400 additional samples of tea have been examined; the results are given in Table 5. Listed by country of origin, it can be seen that Japanese, Formosan, Malayan, and Indian tea

exhibit the highest radioactivity. By comparison, South American, African, Iranian, and some West Indian tea show considerably less radioactivity. Nevertheless, contamination appears to be more or less worldwide. It cannot be explained by any special predilection of the tea plant for translocation of natural radioactivity from the soil, since no pre-1945 teas have ever shown any radioactive content.

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(4) Gamma spectrometry analyses were made at the Division of Nuclear Medicine, Walter Reed Hospital.
(5) Analyses furnished by the Health & Safety Laboratory, Atomic Energy Commission.