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Dietary Considerations of the Radionuclide Contamination of Nonmilk Foods

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Several factors and concepts that may contribute to an understanding of the food contamination problem with Sr^{90} and Cs^{137} are reviewed. In this country at the present time, milk and milk products contain the highest relative proportion of these fission products; however, nonmilk foods, especially vegetables and cereals, contribute more Sr^{90} to the total diet when considered as an entity. In the future, assuming no further testing, nonmilk foods will become even more important as sources of Sr^{90} for the reasons discussed. Since Cs^{137} in milk and meat arises from a similar precursor (bovine serum), and since there is no apparent discrimination between these compartments, the relative contribution from these two major dietary sources of Cs^{137} is not expected to change with time. The variability in the radionuclide content of foods was emphasized by reference to the "Minnesota wheat" situation of several months ago.

AT THE PRESENT TIME, milk and milk products, on the average, are the largest single contributors of environmental radiocontamination to the diet of man in this country. However, nonmilk foods, when considered as an entity, may represent the source of from 40 to 60% of the Cs^{137} and Sr^{90} ingested by the human. Under certain conditions and at future times, nonmilk foods may well assume an even greater relative significance in this respect.

For an understanding of processes that determine relative routes of entry, a brief description of the mechanism of radionuclide transfer may be in order as well as an indication of levels of Cs^{137} and Sr^{90} in various foods. Mention will also be made of factors that alter the magnitude of radionuclide intake from nonmilk and milk sources.

In the present discussion, emphasis will be given to Sr^{90} and Cs^{137} , since these are the radionuclides of most concern. At early times after the release of nuclear

debris, Sr^{89} would represent a significant fraction of the total activity and its general metabolic behavior would follow that of Sr^{90} . Since Sr^{89} has a physical half life of 54 days, its importance would rapidly decrease with time. Radioiodine and radiobarium are also major

sources of radioactivity immediately after nuclear detonation but, because of their short half lives, 8 and 13 days, respectively, their primary dietary vector to man is in fluid milk following consumption of surface-contaminated vegetation by the grazing animal. Other

Table I. Summary of Sr/Ca Observed Ratios

[Sr/Ca observed ratio as defined by Comar et al. (2) = Sr/Ca in product ÷ Sr/Ca in precursor]

Species	Precursor → Product	O.R.	Range	Ref.
Man	Diet→bone	0.25	0.17-0.54	(17)
Man	Diet→milk	0.10	0.08-0.13	(7)
Sheep	Diet→bone (meat) ^a	0.24	0.15-0.31	(17)
Goat	Diet→bone (meat) ^a	0.23	...	(14)
Goat	Diet→milk	0.09	...	(14)
Cow	Diet→bone (meat) ^a	0.20	...	(15)
Cow	Diet→milk	0.10	0.09-0.16	(17, 15)
Rat	Plasma→fetus	0.6	0.55-0.65	(13)
Rabbit	Plasma→fetus	0.5	...	(13)

^a In general, the O.R. from diet to plasma, muscle, and bone is about equal although small differences do exist (2).

Table II. Estimated Mean Content of Sr⁹⁰ in Human Diet in United Kingdom, 1958 (1)

(Total Ca intake was estimated as 1084 mg./day, giving a Sr⁹⁰/Ca ratio of total diet of about 6 μμc. Sr⁹⁰/gram Ca)

Food	Mean Ratio of Sr ⁹⁰ /Ca, μμc./G.	Estimated Sr ⁹⁰ Intake from Food, μμc./Day		Per Cent		
Milk products						
Milk and cream	7.0	3.47	4.20	65.7		
Cheese	7.7	0.73				
Nonmilk products						
Root vegetables	14.2	0.35	1.96	30.7		
Leaf and other vegetables	8.7	0.33				
Fruit, etc.	8.7	0.21				
Flour and other cereals	15.9	0.75				
Eggs	2.0	0.04				
Meat	14.0	0.27				
Fish	1.0	0.01				
Drinking water		0.23			0.23	3.6
Total		6.39			6.39	100.0

Table III. Diet Estimate of Annual Intake of Sr⁹⁰, 1959 (3)

Food	Estimated Sr ⁹⁰ Ingested, μμc./Day		
	Individual food	Sub-total	Per cent ¹
Milk and products	7.0	7.0	39
Nonmilk products			
Bakery products	1.6	11.0	61
Cereal products	1.4		
Meat, fish, etc.	0.3		
Potatoes, other roots	0.6		
Other vegetables	5.3		
Fruits	1.4		
Water	0.4		

radionuclides such as fissile materials, direct products of fission (Pm¹⁴⁷, Zr⁹⁵, Nb⁹⁵, Ce¹⁴¹), or those produced by neutron bombardment (Zn⁶⁵, Co⁶⁰, Fe⁵⁵, Fe⁵⁹, C¹⁴) do not contribute significantly to the body burden of man.

Strontium-90

Strontium, in general, moves through the biosphere in a manner similar to calcium. Detailed studies from several laboratories (17) have shown that various steps in the pathway from fallout material to man can differentiate between calcium and strontium; many of these quantitative differences in behavior have been well defined. Some of the more significant values are given in Table I; the values are presented as the Sr/Ca Observed Ratio (O.R.), which is, by definition, the Sr/Ca ratio in the product compartment (bone) divided by the Sr/Ca ratio in the precursor compartment (diet). There is a definite consistency in the O.R. values between species in similar over-all processes. The Sr/Ca O.R. from diet to bone is about 0.25 in man, sheep, goats, and cows; the O.R. for bone is also approximately the same as for plasma and soft

tissues. For movement of strontium and calcium from diet to milk, the O.R. appears to average about 0.1; recent information in the lactating human also gave a value averaging about 0.1 (7). As an example of another biological process, the rate of passage of calcium across the placental membrane is about twice that for strontium (O.R._{plasma-fetus} = 0.5).

The use of the Sr/Ca observed ratio (or the comparison of strontium and calcium behavior) has considerable usefulness for several reasons: The observed ratio gives basic information on differences in the metabolic behavior of calcium and strontium; the long-term effect of dietary additives, including calcium, on radiostromium retention may be predicted from the observed ratio; the concentration of Sr⁹⁰ in terms of calcium at actual sites of deposition in the skeleton may be calculated; and the Sr⁹⁰ body burden may be predicted with a fair degree of certainty from the Sr⁹⁰/Ca ratio in the diet and a knowledge of the observed ratios. However, studies by Thompson (8) and Kornberg (4) have shown that a direct correlation between the biological movement of calcium and strontium is not always observed under certain experimental conditions; yet, uncertainties in prediction would be greater in attempting to evaluate strontium retention values alone than by a consideration of O.R. values. Therefore, calcium is apparently a good standard of reference for strontium behavior until a better approach is devised, but for purposes of complete assessment, survey data should include analytical values for both calcium and Sr⁹⁰ in the sample.

Extensive analyses of the Sr⁹⁰ content in various foods have been, and are being, carried out in the United Kingdom. Representative values are also available from the United States, but little infor-

mation has been forthcoming from other countries. In Table II, the Sr⁹⁰ contribution from several food sources in the United Kingdom is presented (7); the contribution of the radionuclide from any source was calculated from the calcium ingested from the food (as derived from national food consumption statistics) and the average Sr⁹⁰/Ca ratio of that food. About 65% of the Sr⁹⁰ is consumed with milk products in the United Kingdom, whereas nonmilk sources, including drinking water, contribute 35% of the total Sr⁹⁰ intake. Of the nonmilk foods, cereals, fruits, and vegetables account for 26% and eggs, meat, fish, and drinking water, 9%. The over-all level of Sr⁹⁰ in the British diet in 1958 was calculated to be about 6 μμc. per gram of Ca. In Table III, an estimate of the Sr⁹⁰ contribution from different foods in the United States is given; the data are those compiled by Hardy *et al.* (3). The method of calculation of the Sr⁹⁰ originating from any source was the same in principle as already mentioned. In estimate, milk contributed about 39% of the Sr⁹⁰ and nonmilk foods, including drinking water, about 61%. As with the British diet, cereals and vegetables were next to milk in singular quantitative importance.

It may be well to consider the reasons for the existing relationships between milk and nonmilk foods in their comparative contributions to the human diet. The strontium unit (S.U.) level for vegetables is about the same as that for milk in the United Kingdom (Table II). In the United States, the Sr⁹⁰/Ca ratio in milk is one half to one third that reported in vegetable sources. Assuming that the S.U. for the vegetable plant as taken from the field was about the same as the vegetation grazed by the lactating cow, one would expect that the S.U. of milk would be about one tenth that of vegetables. This obviously was not the situation. Apparently, the processing of the vegetables caused a considerable reduction in their Sr⁹⁰ content.

Studies by Russell (10) have shown that the majority of the Sr⁹⁰ in vegetation is from direct foliar contamination in contrast to root absorption or plant-base absorption. Since the outer leaves of such plants as cabbage and Brussels sprouts are discarded during food processing, a large fraction of the original Sr⁹⁰ activity is also discarded. Similarly, a significant fraction of the Sr⁹⁰ in grains is also discarded in the milling process. Further, the Sr⁹⁰ content of foods may be decreased yet more in the washing and blanching of foods in the frozen food and canning industries. Thus, the accumulative effect of processing vegetables, grains, etc., is to provide a technological barrier against Sr⁹⁰ in the food chain; a significant biological barrier against Sr⁹⁰ ingestion is the

discrimination between calcium and strontium by the bovine mammary gland. If it were not for these processes, the S.U. in human bone would be several times greater than it now is.

Another factor that will alter the relative contribution of Sr⁹⁰ from various food sources is time. At present, an appreciable amount of Sr⁹⁰ in the human diet originates directly or indirectly from foliar deposition of fallout debris. Assuming that there is no further nuclear testing, the soil would in the future become the primary reservoir of radioactivity entering the food chain. At that time, the S.U. in vegetative sources of food for man (cereals, grains, vegetables) may be similar to the S.U. in the diet of lactating cattle. Using the accepted discrimination factors from diet to milk and to meat in the bovine, estimations have been made as to the contribution of Sr⁹⁰ from milk, meat, and vegetation under this situation (Table IV). About 82% of the Sr⁹⁰ would come from plant sources, 17% from milk, and a negligible amount from meat. Although this situation would not occur before considerable decay of Sr⁹⁰ has taken place, there will be a range of conditions in which the Sr⁹⁰ from plant sources will become increasingly important.

Dietary habits of a population will determine relative amounts of Sr⁹⁰ ingested from various sources. Obviously the greater the proportion of milk consumed, the greater the proportion of Sr⁹⁰ that would come from that source. However, the net retention of Sr⁹⁰ from an all milk diet would be less than that from an all vegetable diet. If it is assumed that the fractionation between strontium and calcium in movement from diet to bone is the same for milk and nonmilk foods, the Sr⁹⁰ to calcium ratio in the bone would then be directly related to the strontium units in the diet. Although there is evidence that milk drinking alters discrimination in the rat (2), there has not been any substantiating evidence for this in man. Since the Sr⁹⁰/calcium ratio in nonmilk foods (as a composite) exceeds the ratio in milk by a factor of about three (Table V), it is apparent that individuals subsisting entirely on milk would have a body burden one third that of nonmilk drinkers with the additional assumption that the skeletal size would be about the same. Thus, the greater the proportional intake of milk, the smaller would be the body burden of Sr⁹⁰. Important factors here are the high calcium content of milk and the significant discrimination against strontium in the lactating cow.

The contribution of Sr⁹⁰ from meat and fish is small as compared with milk and cereal sources in the Western countries. In other countries, such as Japan, Sr⁹⁰ from fish may assume greater significance. Salt water fish

Table IV. Estimated Contribution of Milk and Nonmilk Sources to Sr⁹⁰ in Human Diet at Equilibrium

(Estimation based on Sr/Ca observed ratio of diet to milk of 0.1 and diet to meat of 0.25; assume that major Sr⁹⁰ activity originates from soil and that Sr⁹⁰/Ca ratio in vegetation of human diet is about same as that of livestock)

Class of Food	Ca Intake, Mg./Day	Arbitrary Radiostrontium Units, Sr*		Per Cent
		Sr*/mg. Ca in diet	Sr* intake	
Milk	663	10	6,630	16.7
Meat	27	25	675	1.7
Cereals, vegetables, etc.	324	100	32,400	81.6
Total	1014	135	39,705	100.0

contain less Sr⁹⁰ per gram of Ca than do fresh water fish because of the higher calcium content of sea water and the greater volume of dilution.

Cesium-137

Cesium behaves metabolically somewhat like potassium in that it is readily absorbed from the gastrointestinal tract, accumulates in muscle and other soft tissues, and has relatively short biological turnover time. However, distinct differences in biological properties exist between cesium and potassium, particularly the different rates at which cesium and potassium move across cell membranes and the different affinities that intracellular materials have for cesium and potassium. As shown by studies of Relman *et al.* (9) and those in our laboratory, cesium can displace potassium from muscle cells, but the converse, the displacement of intracellular cesium by potassium, does not readily occur. The over-all consequence of this behavior is that cesium moves somewhat independently and noncompetitively in relation to potassium. Data to support this are presented in Table VI.

In this study, rats were given Cs¹³⁷ as a part of their drinking water for 35 days and maintained on complete diets that varied in their potassium content from 0.2 to 1.8%. First, the muscle Cs¹³⁷ levels decreased only about twofold (from 14.0 to 7.5% daily dose) as the potassium increased by a factor of 9; if there were complete interaction between cesium and potassium, one would expect that the ninefold increase in dietary potassium would result in a

Table V. Strontium-90 to Calcium Ratio (Strontium Units) in U. S. Diet

Source	Strontium Units, μc./Sr ⁹⁰ /G. Ca	
	Milk	Nonmilk
Langham, 1957 (6)	6.2	16.9
H.A.S.L., 1958 ^a (3)	10.6	29.7
Lough <i>et al.</i> , 1959 (7)	10.3	33.5
H.A.S.L., 1959 ^a (3)	10.6	31.0

^a H.A.S.L. = Health and Safety Laboratory, U.S. Atomic Energy Commission.

ninefold decrease in Cs¹³⁷ retention. When the discrimination factors between cesium and potassium were calculated on the various potassium intakes, the Cs/K ratio in muscle to the Cs/K ratio in the diet varied directly with the potassium intake. Thus, the discrimination between cesium and potassium is highly variable depending upon, as a primary factor, the amount of potassium that is consumed by an individual.

This point is particularly made since cesium data are oftentimes given in terms of potassium as a reference and the similarity in relationship has not been established. There is some value in employing Cs/K ratios for survey data because the potassium content of muscle and other tissues on a fat-free basis is acceptably constant from animal to animal but does not offer any more advantage beyond using net weight or dry weight of tissue as the basis of reference. One advantage for using Cs/K ratios is that potassium is employed as an internal standard in the radioassay of Cs¹³⁷.

Table VI. Effect of Dietary Potassium on Cs¹³⁷ in Muscle of Rat

(Values represent mean of 5 to 6 rats; animals on continuous Cs¹³⁷ intake for 35 days)

Level of Dietary K, %	% Daily Dose of Muscle Cs ¹³⁷		Dietary Cs ¹³⁷ /K, % Daily Dose/Mg.	Cs ¹³⁷ /K Muscle Cs ¹³⁷ /K Diet
	Per gram	Per mg. of K		
0.2	14.0 ± 0.7 ^a	3.9	3.9	1.0
0.6	10.0 ± 0.1	2.9	1.3	2.2
1.8	7.5 ± 0.4	2.1	0.4	5.3

^a Mean ± standard error of mean.

Table VII. Estimated Mean Content of Cs¹³⁷ in Human Diet in United States, 1956-1957 (5)

(Values estimated from K contribution from each food source and mean Cs¹³⁷/K ratio of respective food. Mean Cs¹³⁷/K ratio of diet is about 20 μμc. Cs¹³⁷/gram K)

Food	Total Intake, Grams/Day		Mean Cs ¹³⁷ /K, μμc./G. K	Cs ¹³⁷ Intake, μμc./Day	% K from Food	
	Dietary	Potassium			Individual	Class
Milk products	533	0.74	32	23.7	60.6	60.6
Nonmilk products						
Meat	223	0.29	32	9.3	23.8	
Flour and Cereals	189	0.12	24	2.9	7.4	
Vegetables	268	0.34	5	1.7	4.3	39.3
Fruits	223	0.06	25	1.5	3.8	
Potatoes	134	0.36	0
Total	1570	1.91		39.1	100	100

Table VIII. Concentration of Cs¹³⁷ and the Cs¹³⁷/K Ratio in Plasma, Milk, Muscle, and Kidney of Lactating Goat after 25 Days on Continuous Isotope Intake

(Values represent average of 2 goats)

Tissue or Fluid	Cs ¹³⁷ , % Daily Dose/G.	Cs ¹³⁷ /K, % Daily Dose/G. K	Cs ¹³⁷ /K Tissue Cs ¹³⁷ /K Plasma	Cs ¹³⁷ /K Tissue Cs ¹³⁷ /K Diet
	Plasma	0.00096	5.6	1.0
Milk	0.0089	5.1	0.9	1.3
Muscle	0.020	4.9	0.9	1.2
Kidney	0.036	19.5	3.5	4.9

The contribution of Cs¹³⁷ from various foodstuffs, as estimated by Langham (5), is given in Table VII. The method of calculation is similar to that for Sr⁹⁰ in that the potassium intake from any class of food was determined from food consumption tables and multiplied by the mean Cs¹³⁷/K ratio of that foodstuff. As indicated before, the majority of the Cs¹³⁷ intake comes from milk with 40% coming from nonmilk food sources. Of the nonmilk foods, meat is the primary food source and, alone, contributes about 24% of the total Cs¹³⁷ intake. Plant foods constitute a minor source of cesium.

Recent studies in lactating goats brought forth data that may be helpful in understanding the long-term behavior of Cs¹³⁷ (12). In this experiment, goats were given Cs¹³⁷ twice daily for 25 days. Over this period, the secretion or excretion of Cs¹³⁷ in milk, feces, and urine was followed. At termination, the levels of Cs¹³⁷ in various tissues were determined. In Table VIII, some of the more interesting values are summarized. Although milk and muscle contain a higher concentration of Cs¹³⁷ than plasma on a per-gram basis, there was no appreciable difference in Cs¹³⁷ content in terms of tissue potassium levels; kidney, however, concentrated Cs¹³⁷ to a greater degree than it did potassium. When the Cs¹³⁷/K tissue values are expressed as a function of dietary Cs¹³⁷/K ratios, (Table VIII), plasma, muscle, and milk contained about 1.2 to 1.4 times as much ingested Cs¹³⁷ as ingested potassium, whereas kidney concentrated Cs¹³⁷ by a factor of 4.9 over potassium. Since meat and milk obviously have the same pre-

cursor (diet), and the Cs¹³⁷/K ratios in these substances are about the same, the relative amount of Cs¹³⁷ in the human diet coming from these foods would not change with alteration in fallout conditions or with time.

Environmental "Hot Spots"

In the foregoing discussion, average levels of Sr⁹⁰ and Cs¹³⁷ in foods as theoretically consumed by the average American were presented. There are large variations in the radionuclide content of different food products obtained from different areas at different times. Of particular concern was the realization that wheat and milk from particular areas contained consistently higher levels of Sr⁹⁰ than comparable foods from other regions.

For example, wheat samples from Minnesota were found to contain an average of 153 μμc. of Sr⁹⁰ per gram of calcium or 51 μμc. of Sr⁹⁰ per kg. of wheat. These values are considerably higher than the strontium units detected in bread and other wheat products in other states. The conditions and circumstances that result in these wide regional variations are not known precisely. Several factors, all playing a part simultaneously, could conceivably offer an explanation. If the fallout were particularly high immediately before harvest, those plants and their products might show relatively high levels of Sr⁹⁰.

A localized, increased rate of fallout was implicated since the accumulated amounts of Sr⁹⁰ in the soil of Minnesota at that time were about the same as

the United States average. Although higher than average values are disconcerting in themselves, the American diet is a composite from several sections and, therefore, there is an averaging of the Sr⁹⁰ content from many food items. Also, the Sr⁹⁰ levels in Minnesota wheat only occasionally exceeded the maximum permissible concentration as recommended by the National Committee for Radiation Protection and Measurement, and the International Committee on Radiological Protection for the whole population. However, there are environmental "hot spots" and these should be considered in assessing the over-all problem of radionuclide contamination of foods.

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