Radioisotopes and Environmental Circumstances: The Internal Radioactive Contamination of a Pacific Island Community Exposed to Local Fallout

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A unique opportunity for the study of the internal radiation hazard associated with the contamination of an inhabited land mass by *local* fallout was afforded when several of the Marshall Islands were accidentally contaminated to varying degrees as a consequence of the fallout-producing nuclear detonation of March 1, 1954 (Cronkite et al, 1956). The area contaminated was thousands of square miles beyond the range of the thermal and blast effects. Two hundred thirty-nine Marshallese persons were exposed to levels of gamma radiation, ranging from 175 r on Rongelap to 14 r on Utirik. Further, the inhabitants of Rongelap and Utirik were also subjected to an acute inhalation and ingestion exposure during the 48-hour period that elapsed prior to evacuation. Their initial body burdens of internal emitters were estimated from analysis of their urine and also from data obtained on animals simultaneously exposed. These data indicate that the acute hazard from the internal emitters was very small as compared to the concomitant external dose. Medical surveys have been made yearly since the accident in order to follow up the recovery progress of the exposed people (Bond et al, 1955; Cronkite et al, 1955; Conard et al, 1956, 1957, 1959).

Within a month of the accident, islands of the following atolls were surveyed: Rongelap, Rongerik, Bikar, Likiep and Utirik. Numerous land animals, birds and marine specimens, and samples of plants, soil and water were collected for analysis of the content and distribution of radioactive material (Cohn et al, 1955). At the same time, a gamma dose rate survey was made over several typical land areas to provide data on the degree of the external radiation hazard.

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was similar to cow milk in that the CaKNa resin removed less than that removed from milk which had strontium added to it. In fact only 55 per cent of the strontium was removed, less than could be removed from cow milk. This would indicate that the binding capacity in goat milk is greater than in cow milk. A low calcium resin was effective in removing the strontium from goat milk. A resin mixture (1Ca:2K:1Na) removed 91.3 per cent of the strontium.

Conclusions

The experiments described in this paper indicate that strontium and cesium could be removed from milk without altering the flavour or the calcium composition of the milk.

The simplest treatment is that of a single treatment with equilibrated resin, e.g., CaKNa. This resin could be regenerated with equilibrating solution. In this case only 50 per cent could be removed in one treatment and about 70 per cent in two treatments. Resorting to a pair of mixed resins would remove 85 to 90 per cent of the strontium. The mixed resins are more efficient because some of the strontium is bound and is not available for exchange onto an equilibrated resin.

It appears that strontium and cesium removal from milk is possible. More work must be done before the process can be placed on a commercial basis.

Acknowledgment

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Contamination from Local Fallout

In order to ascertain the degree of radiation hazard associated with residual contamination, surveys of the fallout contaminated areas have been conducted over a 5-year period.

The most comprehensive studies were made at 1 and 2 years following the accident (Rinehart et al, 1955; Weiss et al, 1956). Data were obtained on the residual activity in soil and on the uptake and retention of fallout material by plants and land and marine animals. These data form the basis for an estimate of the radiation hazard associated with both an acute and chronic exposure to local fallout. It is perhaps the only long-term study of a community exposed to a significant local fallout, and thus is of considerable value. Further, since the Rongelap people were moved from their original habitat and then returned after a period of 3 years, the relationship between body burden and changing levels of environmental contamination can be studied. Unfortunately, subsequent weapon tests in the Pacific Proving Grounds during the past 5 years have to a degree interfered with the assessment of the relationship between environmental contamination and rate of equilibration of various of the fission products in the body, insofar as they have contributed small but detectable accretions to the contamination in this area. The procedures for sample collection, preparation and radiochemical analysis have been described (Rinehart et al, 1955; Weiss et al, 1956; and Shipman et al, 1955).

The most interesting new technical development in the field of fission product analysis in human beings has been the application of the technique of whole-body gamma spectroscopy. In 1957 it was determined by Miller that direct whole-body gamma counting of the Marshallese was feasible. A group of seven Marshallese was brought to Argonne National Laboratory and counted in their whole-body counter. In 1958 and 1959, a "portable" (20 ton) whole-body counter, designed and built at Brookhaven National Laboratory, was taken out to the Marshall Islands for use in the annual medical survey. Several hundred Marshallese people were counted (Conard and Robertson, 1958; Cohn, 1959). The technique of whole-body spectrometry has been described by Miller (1959).

Results and Discussion

The radiation hazard to human beings resulting from habitation in an area subjected to radioactive contamination resides in the fate of the radioactive material deposited thereon. Radioactive material settling down on the earth's surface is transported through the soil, air and water to plants, and thence to animals and finally to man. The levels of radioactivity in each of these components of the ecological chain from soil to man were measured and are reported below.

External Dose

The levels of radioactive contamination which fell on the Marshall Islands were reflected by the doses that were received over approximately 48 hours

(figure 1). Gejen, an uninhabited northern island, received the highest dose, 2,000 r; Rongelap (from which it was necessary to evacuate the inhabitants) received an intermediate dose of 175 r; and Utirik Island to the east received 14 r in the first 2 days. The dose rate on the Rongelap Atoll, as measured

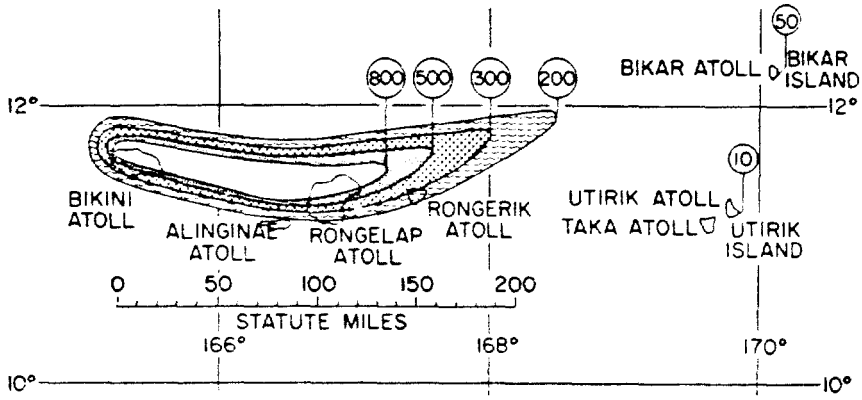

FIGURE 1. Isodose lines of estimated pattern of radioactive fallout, Pacific Proving Grounds, March 1, 1954. The numbers on the map represent the doses that would have been received over approximately 48 hours without shielding. The dose, above which survival is unlikely is 600 r, and below which survival is probable is 200 r (from Cronkite et al, 1956).

initially and over a 5-year period, is presented in table 1. The values of dose rate at 1 year varied from 0.5 mr/hr on Rongelap Island to 3.0 mr/hr on Kabelle Island. These values represent averages, as the levels of activity were not uniform on each island. The rapid fall in gamma activity over the first 2-year period reflects the high percentage of short-lived radioisotopes which contribute

TABLE 1. Average gamma dose rates on Marshall Islands following nuclear detonation of March 1, 1954 (from Held, 1958)

	Gamma exposure levels Rongelap Atoll (mr/hr at 3 ft above ground)		
	Rongelap Island	Eniactok Island	Kabelle Island
Jan. 1955	0.5	2.0	3.0
Oct. 1955	0.2		
July 1956	0.4		1.2
July 1957	0.07		0.20
March 1958	0.029	0.067	0.14
Aug. 1958	0.058	0.076	0.16
March 1959	0.035	0.048	0.10

the activity at early time intervals. The principal isotopes present at the early intervals are Sr^{89}, Sr^{90}-Y^{90}, Zr^{95}-Nb^{95}, Ru^{106}-Rh^{106}, $Te^{127\text{-}129}$, $I^{131\text{-}132\text{-}133\text{-}135}$, Cs^{137}-Ba^{137}, Ba^{140}-La^{140}, Ce^{144}-Pr^{144}, Pr^{147}, and Np^{239}.

Soil

The fallout material appeared initially to be associated with relatively large particulate matter and to be distributed uniformly over the soil. The residual activity on the islands was initially contained primarily in the top few inches of soil.

Radioanalysis of soil profiles at 1 year indicated that little translocation of the activity had occurred. Cs^{137}, Ce^{144}-Pr^{144}, and Ru^{106}-Rh^{106} constituted the largest portion of fixed contamination in the soil at this time.

Activity in the lagoon bottom silt at 1 year appeared to be distributed rather uniformly to a depth of 6 or 7 inches. Four samples of silt from the northeast corner of the Rongelap Atoll lagoon had levels of beta activity ranging from 8,000 to 12,000 d/m/g. The samples were collected at water depths of 40 to 120 ft. The activity in lagoon bottom silt is of interest, since it provides a reservoir of activity available for future incorporation into marine food specimens.

At 2 years following the contaminating event, most of the activity was still firmly fixed to the surface layer (0 to 1 inch) of soil. Loss of activity from the surface layer in the period 1 to 2 years was largely the result of radioactive decay rather than the result of leaching or erosion. At 2 years, the rare earths, Ru^{106}, Sr^{90}, and Cs^{137} constituted 84, 10, 5 and 1 per cent of the activity respectively on Rongelap Island.

The Sr^{90} concentration in the surface soil of the island at 2 years is shown in table 2. The level of Sr^{90}/Ca in the soil of Rongelap was 8.4×10^3 $\mu\mu$c Sr^{90}/g of exchangeable calcium. The level of a soil sample from Gejen (the northernmost island) was exceptionally high, 6.2×10^5 $\mu\mu$c Sr^{90}/g exchangeable calcium. These concentrations correspond roughly to the external gamma dose rates measured on the islands.

The extractability of Sr^{90} (i.e., the ability of plants to extract Sr^{90} from soil) depends on the soil type, the solubility of the Sr^{90} compounds, water depth, pH of soil, moisture and organic matter (Blume and Smith, 1954). In addition to the Sr^{90} extractability from the soil, the amount of exchangeable calcium in the soil is also a very important factor. The ability of plants to take up Sr^{90} is inversely related to the amount of exchangeable calcium in the soil (Fuller and Flocker, 1955). For example, plants grown in acid soils low in calcium take up the largest amounts of Sr^{90}, while plants in alkaline calcareous soil take up the least. In the last column of the table (table 2), the ratios of Sr^{90}/exchangeable calcium are tabulated. While the calcium content of the various soil types analyzed ranges from 23 to 35 per cent, the exchangeable calcium in the soil represents only a small fraction of the total calcium.

TABLE 2. Strontium[90] concentration in surface soils of Marshall Islands, March 1956 (from Weiss et al, 1956)

Island	Soil type	Sr^{90} (d/m/g soil)	"Exchangeable" calcium (mg/g soil)	$\mu\mu c$ Sr^{90}/g calcium
Gejen	large calcareous particles	1,886 ± 20	1.4	$6.2 \times 10^5 \pm 6.3 \times 10^3$
Eniaetok	small sand-like particles	400 ± 11	0.9	$1.9 \times 10^5 \pm 5.4 \times 10^3$
Sifo	small sand-like particles	25 ± 3	0.8	$1.3 \times 10^4 \pm 1.7 \times 10^3$
Rongelap	loamy	234 ± 4	12.7	$8.4 \times 10^3 \pm 150$
Eniwetok	small sand-like particles	62 ± 2	4.6	$6.1 \times 10^3 \pm 180$
Utirik	small sand-like particles	3 ± 1.5	1.9	660 ± 330
Likiep	loamy	11 ± 2	11.3	430 ± 60

Contamination from Local Fallout

Plants

The uptake of the fission products from soil by plants is the next step in the chain by which these radioisotopes reach man. It was found that, during the first 6 weeks, only a very small amount of the fission products is available to plants growing on contaminated soil. The fallout material appeared initially to be associated with particulate matter and to be uniformly distributed on earth, grass and food plants. Levels of beta activity of the order of 1 μc per plant and fissile material (1×10^{-4} μg) were present on the external surfaces of plants at one month (Rinehart et al, 1955). Only very low levels of beta activity and no alpha activity were detected in the edible portions of the plants. The one exception was the presence of high levels of beta activity in the sap of the coconut tree (1 μc/liter). The isotopic composition of this tree sap was very similar to that of the ground water, suggesting uptake of these fission products by the root system. The predominant fission product found in the tree sap at this time was Sr^{89}, presumably as a function of its abundance and relatively high solubility.

At the time of the 1-year resurvey of the island, extensive incorporation of radionuclides into plants had occurred. The gross beta activity in plants on Rongelap Island at 2 years after the detonation is shown in table 3. The level of internal contamination in the plants corresponds roughly to the level of activity in the soil and the gamma dose rate on the islands. However, wide variations occur in the levels of beta activity in the plants on any one island. The availability of the fission products to the plant is undoubtedly a function of the age of the plant, the stage of development of the fruit, the type and depth of soil and the physical-chemical properties and distribution of the fallout material. Since the fallout from the March 1, 1954 detonation consisted of fission products associated with large particles of CaO, $Ca(OH)_2$, $CaCO_3$ and NaCl particles, it was very soluble in the soil and could readily be absorbed through the root system of the plant. Moreover, it could be absorbed directly through the leaves.

The plant portulaca was found to have by far the highest level of activity of any of the plants analyzed. Leafy structures of plants were, in general, more radioactive than their fruit. The leaves of the plants contained primarily the rare earth elements together with small amounts of Ru^{106} and Sr^{90}. By contrast (table 4), the primary fission product in coconuts, papaya fruit and arrowroot tuber was Cs^{137}. Very small amounts of activity were contributed to these plants by the rare earths, Ru^{106} or Sr^{90}. The concentrating capacity of coconuts for Cs^{137} was especially marked when compared to the low levels of Cs^{137} in the soil, particularly in the area of the root system of the plant. The high Cs^{137} concentration may reflect foliar absorption and/or a potassium deficiency in the plants with the substitution of cesium for potassium. The Sr^{90} concentration was uniformly low in the edible portions of the above samples of plants.

TABLE 3. Gross beta activity in plants, April 1956 (from Weiss et al. 1956)

Plant	Part	Island						
		Gejen	Eniwetok	Eniaetok	Rongelap	Sifo	Utirik	Likiep
		(c/m/kg × 10^{-1})[a,b]						
Portulaca	whole plant	87.4	19.2	3.05	1.26		1.71	1.33
Arrowroot	stems, leaves	11.0	4.5	0.32	0.25	0.21		0.03
	tubers	2.32	0.57	0.69	0.55	0.08	0.14	0.03
Pandanus	air root	2.87	0.17	1.05	0.32	0.96	0.08	0.02
	leaves	2.64	1.02	5.26	0.38	0.15	0.21	0.03
	green keys	1.27	0.37	0.70	0.22	0.10	0.09	0.03
	ripe keys	.....	.....	0.53	0.17	.....	0.07	0.02
Papaya	ripe	.....	.....	.....	0.12	.....	0.11	
	green	.....	.....	.....	0.25	.....	0.09	0.04
	leaves, trunk	.....	.....	.....	0.09	.....	0.16	0.06
Ripe coconut	milk	2.87			0.54	0.63	0.12	0.57
	meat	1.90	0.36	1.97	0.24	0.17	0.08	0.06
	shell	4.98	0.38	0.72	0.44	0.28	0.06	0.02
	husk	1.83	0.65	1.57	1.31	0.77	0.21	0.09
Green coconut	whole	3.1						
	milk	.....	0.29	0.11	0.05	.....	.....	.....
	meat		0.33	0.25		0.13		0.05
	shell			0.80		0.08	0.07	0.02
	husk			0.48	0.12	0.37	0.08	0.09
	shell, husk		0.11			0.11	0.11	0.02

[a] All counts were corrected for the counting efficiency of Sr^{90}-Y^{90}.

[b] Gross beta activity of plant samples was determined in April 1956 and that of soil and water in May 1956.

TABLE 3 (Continued)—Gross beta activity in plants, April 1956 (from Weiss et al, 1956)

Plant	Part	Island						
		Gejen	Eniwetok	Eniaetok	Rongelap	Sifo	Utirik	Likiep
		(c/m/kg × 10^{-5})[a,b]						
Sprouting coconut	milk		1.61	0.76	0.79	0.71	0.11	0.09
	meat		0.38	0.40	0.12	0.30	0.07	0.06
	shell		0.29	0.41	0.35	0.18	0.04	0.02
	husk		0.73	1.57	0.88	0.68	0.26	0.07
Coconut	leaves		15.4	0.86		0.84	4.7	1.66
	frond		0.94	0.51		0.23	0.09	0.11
	leaves, frond	1.48						
Banana	fruit							0.06
	bark							0.07
	leaves							0.18
Taro	leaves, stalks							0.06
	tuber, roots with soil							0.19

[a] All counts were corrected for the counting efficiency of Sr^{90}-Y^{90}.

[b] Gross beta activity of plant samples was determined in April 1956 and that of soil and water in May 1956.

Sr^{90}/Ca ratios were highest (over 1,000 $\mu\mu$c Sr^{90}/g of calcium) in portulaca, coconut meat and milk and pandanus (Weiss et al, 1956). Portulaca had a particularly high Sr^{90}/Ca ratio, ranging from 6,000 to 25,800 $\mu\mu$c Sr^{90}/g of calcium. Average Sr^{90}/Ca ratios in common foods collected at 3 and 4 years are discussed later in considering the dietary estimate of body burdens in the exposed Marshallese.

Water

Ocean and lagoon water samples were collected initially off several islands in addition to the samples collected from cisterns and wells on the islands. Water from the cisterns and a well on Rongelap had high levels of activity (1 μc/liter) at 30 days. The predominant radionuclide present in the water at this time was Sr^{89}. Cistern water contained varying levels of contamination, depending on the original contamination, nature of watershed areas, etc. The ocean water samples contained very small amounts of beta activity, presumably derived from activity washed off the islands. The radiochemical composition of ground water,

TABLE 4. Average relative composition of nuclides in plants, soil and water, March 1956 (from Weiss et al, 1956)

Source		No. of samples averaged	Relative composition (per cent)			
Plant	Part		Cs^{137}	Total rare earths	Sr^{90}	Ru^{106}
		Plants				
Portulaca	whole	1	48.9	39.2	11.8	.....
Papaya	fruit	1	79.8	17.8	2.5	.....
Coconut	husk	3	98.2	1.1	0.7	.....
	meat	2	98.9	0.05	1.0	.....
	shell	2	99.5	0.4	0.1	.....
	milk	1	99.6	0.2	0.2	.....
	leaves	2	8.3	86.5	0.4	5.1
Pandanus	keys	2	92.6	2.2	5.5	.....
	leaves	2	72.7	13.3	5.1	8.9
	air root	2	88.9	10.3	0.8	.....
Arrow root	tuber	1	75.4	16.8	1.0	6.8
	leaves	1	11.7	83.9	3.0	1.4
		Soil				
Depth, 0-1 in.		2	0.34	83.8	5.6	10.0
		Water				
Cistern		2	.....	64.4	35.6	.....
Well		2	.....	100	0	.....
Lagoon		2	.....	94.5	5.5	.....
Ocean		2	.....	100	0	.....

lagoon bottom silt and lagoon water was similar to that of the soil at 1 year (Rinehart et al, 1955).

At 2 years the gross activity in water samples was of a very low level. The observable activity was (as determined at 1 year) due primarily to the rare earth elements (table 4).

Fish and Marine Specimens

The level of radioactivity found in the tissues of fish and marine invertebrates collected at 2 years, while only 3 per cent of that found in fish collected at 1 month (3 μc/kg) was nevertheless readily detectable (Weiss et al, 1956). Considerable variation existed in the concentration of activity per weight as a function of the geographic location of the fish and marine specimens. In the Rongelap Atoll, for example, fish and invertebrates caught in the northern part of the lagoon at 1 year contained, on the average, 3 to 4 times the amount of internally deposited fission products as was found in similar specimens from the southern part of the lagoon. This finding is consistent with the fact that the northern lagoon was exposed to higher concentrations of fallout material. (Average external gamma readings of the northern and southern Rongelap Islands at the time of sample collection were 5.8 and 0.7 mr/hr, respectively.) This difference in concentration of radioactivity in fish from the north and south lagoons was not noted at 2 years. Apparently the activity was completely diffused throughout the lagoon by this time. While there was considerable variation in the concentration of activity of individual fish from some areas, no correlation could be found between the levels of radioactivity and the eating habits of the fish (carnivorous, herbivorous, and omnivorous).

Snails concentrated radionuclides to a much greater extent per unit body weight than did the fish in the corresponding localities. This concentrating ability may be due to the fact that snails feed on the lagoon bottom where higher concentrations of nuclides are found. Crabs and clams also showed high fission product concentration at 1 year (Rinehart et al, 1955). Approximately 40 per cent of the activity of fish collected at one year in the Rongelap and Rongerik lagoons was fixed in the skeleton. Muscle and viscera contained approximately 15 and 20 per cent, respectively, of the total internal activity. The remainder of the activity was found on the skin and the gills. At 2 years the skeleton contained 50 per cent of the beta activity, the viscera 33 per cent, and the muscle 14 per cent. The distribution was very similar to that found after 1 year. Zn^{65}, a "neutron-induced" activity, accounts for the high gamma-to-beta ratio observed at 1 and 2 years. Zinc was fairly evenly distributed throughout the tissues, contributing about 90 per cent of the activity in the skeleton and muscle (Weiss et al, 1956). The level of Sr^{90} was uniformly low in the marine specimens. Sr^{90} constituted less than 1 per cent of the total, and was found chiefly in the skeleton. The muscle of fish caught in the Rongelap lagoon in 1955 had average values of 200-300 $\mu\mu$c Sr^{90}/g of calcium (Weiss et al, 1956).

The presence of Co^{60} in the soft tissue of clams was noted for the first time in the 2-year resurvey (Weiss and Shipman, 1957). The Co^{60} accounted for the major fraction of the total activity in the clams. The very marked ability of clams to concentrate Co^{60} selectively was verified in laboratory experiments (Gong et al. 1957).

Land Animals

Internal radioactive contamination of animals from Rongelap was determined by radiochemical analyses of the tissues of domestic animals (pigs and chickens) sacrificed at various times following contamination (Cohn et al, 1955). The internal contamination resulted from both inhalation and ingestion of contaminated food, with the latter being the more important route of entry. The body burden of fission products was roughly proportional to the gamma dose rate on each island. The internally deposited activity in the pigs living on Rongelap for 1 month after the detonation was tenfold higher than that of the human beings who were evacuated within 48 hours. The difference thus reflects the prolonged stay of the animals in the contaminated area.

Only a small percentage of the fission products initially present in the environment was readily absorbed from the lungs and G. I. tract and retained in the body of land animals. This G.I. tract activity was contributed chiefly by isotopes of short radiological and biological half-life and limited solubility, and thus the levels of activity in the tissues of the body were quite low. At 3 months, radiochemical analyses of tissues from Rongelap pigs indicated that Sr^{89}, Ba^{140} and the rare earth group constituted 75 per cent of the total internal beta ac-

TABLE 5. Radiochemical analysis of tissues and urine of Rongelap pigs exposed to fallout from the March 1, 1954 nuclear detonation[a] (from Cohn et al. 1955)

	Beta activity d/m/total sample $\times 10^{-3}$			
	Gross activity	Sr^{89}	Ba^{140}	Rare earths
Skeleton (total)	8,745 (100%)	5,380 (62%)	595 (6.8%)	850 (9.7%)
Lungs (alveolar)	1.3	0.24	0.22	0.57
Stomach	1.6	0.26	0.62	0.80
Small intestine	2.5	0.73	0.69	0.69
Large intestine	14	5.0	2.8	4.0
Liver	29	0.47	0.27	5.9
Kidney	3.2	0.18	0.30	0.61
Remaining carcass	455			
Thyroid dose	100-150 rep (estimated from early analysis of urine)			
Total external gamma dose	330 r			
Internal beta activity	4 μc			

[a] Values are the average of two young adult pigs which were analyzed 3 months after detonation.

tivity (table 5). The largest portion (99 per cent) of the internally deposited activity was fixed in the skeleton.

The biological localization of residual activity within skeletal tissue is shown in an autoradiograph of the femur of a pig that was exposed to the initial fallout for a period of 30 days following detonation (figure 2). It will be noted that there is a dense concentration of fission products in the epiphyseal region of the bone. For comparison, the autoradiograph of a tibia of a rooster collected at 2 years following detonation is shown in figure 3. Here is seen a concentrated deposition of activity in the diaphysis and a lighter deposition in the region of growth at the ends of the bone, indicating that the primary deposition occurred soon after detonation, while the animal was young and growing, and that subsequent bone growth incorporated much smaller amounts of radioactive material.

FIGURE 2. Autoradiograph of femur of Rongelap pig; sacrificed 38 days after detonation (from Cohn, 1956).

FIGURE 3. Autoradiograph of tibia of Rongelap rooster, March 1956 (from Cohn, 1955).

Rongelap People

The body burdens of internal emitters in the Marshallese were estimated from the data obtained by radiochemical analysis of the tissues of the above-mentioned pigs which were simultaneously exposed, and from a comparison of urinalysis data from animals and from human beings (Cohn et al, 1955). The animal data are indicative of the qualitative nature of the inhalation exposure to the people, even though they quantitatively reflect the greater ingestion by the animals of contaminated food during their prolonged stay on the island. The total amount of radioactive material in the G.I. tract of the Rongelap people at 1 day after exposure was estimated to be 3 mc.

The mean body burden of the Marshallese at 1 day following the 1954 fallout was estimated to be Sr^{89}, 1.6 μc; Ba^{140}, 2.7 μc; I^{131}, 6.4 μc and the rare

earth group together, 1.2 μc. The contribution of this amount of internal contamination is small as compared to the 175 r external gamma that the Marshallese people received.

In the first few months following exposure, Sr^{89} and I^{131} (plus the shorter-lived iodine isotopes) contributed the greatest internal radiation dose. In the acute exposure, I^{131} and the shorter lived I^{132}, I^{133} and I^{135} contributed the highest individual tissue dose, 100-150 rep to the thyroid. Sr^{89} contributed the major portion of the beta dose to the skeleton at this early time.

Estimation of the Internal Radiation Hazard

The potential radiation effects produced by specific quantities of internally deposited radioisotopes can be predicted from the clinically observed effects of known amounts of internally deposited radium. These effects do not appear, of course, until a period of years has elapsed, and observations made at early times following exposure yield no significant data concerning the damage. Thus, as anticipated, the people and animals on Rongelap Island who received an intermediate fallout dose exhibited only transitory changes in blood-cell levels over the 5-year period of observation since their exposure, but no other pathological changes that could be ascribed to radiation.

In terms of chronic exposure to internal radiation, Sr^{90} is clearly the critical element. Particular effort was therefore made to determine its levels in soil, plants and man himself, and also the inter-relationship of these levels. In the situation where people are exposed to additional fallout from continued weapon testing, the sustained high level of the shorter-lived Sr^{89} may result in a hazard of the same order of magnitude. Thus Sr^{89} has been a significant fission product up to the cessation of testing in 1958.

Of the gamma-emitting fission products, Cs^{137} is of the greatest interest, even though of minor significance as an internal radiation hazard. Like Sr^{90}, Cs^{137} has a gaseous precursor with a half-life sufficiently long to avoid early condensation in the fireball. Cs^{137} thus follows Sr^{90} into the stratosphere. Since the fission yields and the half-lives of the two radioelements are nearly equal, they are present in the fallout in like quantities. While they behave differently in the ecological cycle, based on their differing chemical properties, Cs^{137} nevertheless provides a useful tracer for studying the movement of Sr^{90} through the biosphere, since its gamma-emitting properties make it readily detectable.

There is also some interest in the neutron-induced radioelement Zn^{65}, even though it, too, does not appear in levels which are hazardous to human beings. The interest centers chiefly around the fact that it is definitely transmitted through marine life, and thus provides a clear example of the transmission of a radioclement through the food chain to man.

Other fission products also appear in small amounts in the food chain and thus appear ultimately in man, but their levels are so small that little effort

has been made in the past to detect them or to trace their movement. Thus only the above three fission products of greatest interest will be considered here.

The body burden of fission products can be determined in three ways. The method of choice is the direct in vivo measurement using whole-body spectrometry. The limitations of this method are that few whole-body counters are in existence, and their absolute calibration is quite difficult. More importantly, this method is, of course, restricted to analysis of gamma-emitting isotopes, since, to date, a whole-body beta counter has not been developed.

A second method for calculating body burden, particularly for counting beta emitters such as Sr^{90}, is the estimation of the internal deposition from data obtained by radiochemical analysis of the urine.

Finally, it is possible to make a completely indirect estimate of the human body burden of radioisotopes by what may be called the environmental approach. In this method, the estimate of the body burden is based on the concentrations of the fission products present in the environment, chiefly the soil and the important components of the diet. In order to make this estimate, data must be obtained on the transfer of the fission products between successive elements of the ecological chain leading from soil to bone. For example, although strontium and calcium are chemically similar and thus appear together in the various components of the ecological chain, calcium is taken up preferentially by plants and animals, so that it is necessary to determine the discrimination factor for each step. When these factors are known, it is possible to estimate the dose to the "critical organ" in man from the concentration of the isotope in any step of the chain.

Environmental Estimate of Body Burden

The environmental estimate of internally deposited Sr^{90} can be made in the following way. A number of rats were collected on Rongelap at 2 years following the 1954 accident. They had subsisted on a diet consisting primarily of plants. These animals were subjected to careful radiochemical analysis, as it was thought that they might serve as indicators of the internal radiation hazard that would result to human beings if they inhabited the same area during this time, and subsisted on essentially the same diet. The Sr^{90}/Ca ratios for various tissues of these rats were measured directly and these values were compared to the Sr^{90}/Ca ratios of the food and soil on Rongelap collected at the same time. Thus a comparison could be made between the environmental approach and the direct measurement. Extrapolation of the environmental data will give the equilibrium value, whereas the direct measurement give the value at the time of measurement. In this manner, the per cent of equilibrium of Sr^{90} internally deposited in the rat can be determined. It can be seen from the data that the Sr^{90}/Ca ratios for different foods on Rongelap varied greatly. Further, the diet of the rats on Rongelap was too uncertain to assume an "average" diet, so that it was necessary for this estimate to use the Sr^{90}/Ca values of the soil itself.

Contamination from Local Fallout

The "Strontium-Calcium Observed Ratio" (OR) of Comar et al (1956) was used to denote the preferential utilization of calcium in the following manner:

$$OR_{\text{sample-precursor}} = \frac{\text{Sr/Ca of sample}}{\text{Sr/Ca of precursor}}$$

The Sr^{90} discrimination ratio in the chain from soil (s) to bone (b) via plants (p) can be expressed as follows:

$$OR_{\text{bone-soil}} = (OR_{p\text{-}s})\ (OR_{b\text{-}p}) = (0.7)(0.25) = 0.18$$

The value $OR_{b\text{-}p} = 0.25$ is an approximate value obtained experimentally on rats fed a stock laboratory diet (Comar et al, 1956). This discrimination factor of 4 for calcium against strontium from diet to bone in man has been reported by Schulert et al (1959a) and Bryant et al (1958). A more appropriate value for the rats in this situation might be the $OR_{\text{bone-diet}} = 0.16$ obtained by a study of wild Kangaroo rats living in the Nevada desert (Alexander et al, 1956).

The body burden is the:

$$(Sr^{90}/Ca)_b = (Sr^{90}/Ca)_s (OR_{p\text{-}s})(OR_{b\text{-}p})$$
$$= (5.4 \times 10^3)(0.7)(0.16) = 924\ \mu\mu c\ Sr^{90}/g\ Ca$$

The value thus obtained is approximately twice the value of 470 to 545 $\mu\mu c$ Sr^{90}/g of calcium obtained by direct radiochemical analysis of the tissues of rats living on the island during the 2-year period following detonation. This difference in the indirect environmental estimate of the body burden of Sr^{90} Ca as compared with the direct analysis may reflect either errors in the discrimination ratios or a lack of equilibrium between the Sr^{90}/Ca in the animals with the soil at 2 years. Since 13 rats of the same average age analyzed at 4 years had values close to those of rats collected at 2 years, at 443 ± 181 $\mu\mu c$ Sr^{90} g of calcium (Held, 1958), it must be assumed that the discrimination ratios are not sufficiently accurate for this estimation.

It is obvious that applying the same technique of estimating the Sr^{90} body burdens of the Marshallese people is also difficult because of the uncertainty of their diet and the discontinuous nature of their habitation on Rongelap Island.

Dunning (1957) has estimated the future Sr^{90} body burdens in the Marshallese from the Sr^{90}/Ca in an "average" food supply which was about 360 $\mu\mu c$ Sr^{90}/g of calcium in 1956. This figure would be reduced to a daily intake of about 100 $\mu\mu c$ Sr^{90}/g of calcium if consumption of land crabs which have a high Sr^{90} content were eliminated.

These estimates of average dietary intake are very approximate since the diet is not well known. A more extensive study made in 1958 yielded Sr^{90}/Ca levels of 67.5 $\mu\mu c$ Sr^{90}/g of calcium somewhat lower than those of Dunning's in 1956 (table 6). This study was based on an analysis of the average diets of 14 males on Rongelap (Held, 1958) and a radiochemical study carried out by Harley (1959). It was assumed that half of the daily calcium (0.8 g) was from

TABLE 6. Estimate of strontium[90] in diet of Rongelap adult, 1958

	A Daily intake[a] (g)[b]	B Calcium content (mg Ca/g)	C Daily calcium intake in mg (A × B)	D Fraction of total calcium intake[c]	E Sr^{90} content ($\mu\mu c$/g Ca)	F Contribution to total daily Sr^{90} intake (D × E) ($\mu\mu c$/g Ca)
Meat from mature coconut	89	0.075	6.7	0.008	1,200	9.6
Meat from drinking coconut	75	0.14	10.5	0.013	210	2.7
Milk from green coconuts	116	0.15	17.4	0.022	1,000	22.0
Pandanus edible portion	79	0.15	11.9	0.015	930	14.0
Arrowroot	58	2.10	121.8	0.152	19	2.9
Breadfruit	45	0.60	27.0	0.034	260	8.8
Fish	139	0.13	18.1	0.023	280	6.4
Clams	45	4.00	180.0	0.225	5	1.1
Crabs, land	14	4.00	56.0	0.070	(4,000)	(280.0)
	660		449	0.56		67.5
Imported Food						
Rice						
Canned "C" rations						
Flour						
Tea						
Milk						
Salt and sugar						

[a] Based in part on average daily diet of 14 Rongelap males (Held, 1958)

[b] Wet weight

[c] Based on total calcium intake of 0.8 g/day

indigenous foods and half from imported foods. Using the above-mentioned discrimination factor of 4, the equilibrium body burden for the Rongelap people on 67.5 $\mu\mu$c/g of calcium daily intake (excluding land crabs in the diet) would be approximately 17$\mu\mu$c/g of calcium. This is about 68 per cent of the equilibrium value estimated by Dunning (1957) and 74 per cent of that estimated from urinalysis data in 1958 (Woodward et al, 1959). Another effort was made in the 1959 Medical Survey to gather samples of meals to be assayed for their Sr^{90}/Ca content. However, since the Marshallese were found to subsist to a large extent on foods not indigenous to the area, such as "C" rations, rice and tea, it is even more difficult now to extrapolate from Sr^{90}/Ca ratios in food to body burden.

It is obvious that further data on the transport of low levels of Sr^{90} and other fission products through the ecological cycle in this and other communities are required to assess the internal radiation hazard to human beings living in a fallout-contaminated area.

Estimate of Body Burden by Whole-Body Counting and Urinalysis

More reliable estimates of the Marshallese body burdens were obtained by whole-body gamma spectrometry and by radiochemical urinalysis. The urinary excretion levels of Sr^{90} for 5 years following exposure to fallout are shown in figure 4. The 4- and 5-year urinary Sr^{90} levels were much higher following the return of the Marshallese to Rongelap. The excretion rate may be expressed as the sum of two exponential functions. The major fraction of Sr^{90} is excreted with a half-life of 40 days, and a smaller fraction is excreted with a half-life of 500 days. These excretion rates correspond to those reported by Cowan et al (1952) in a case of accidental inhalation of Sr^{90}, and were used in extrapolating back to the 1-day Sr^{89} body burden of the Marshallese (Cohn et al, 1955).

The Cs^{137} urinary excretion levels of the Marshallese for the 6 months immediately following exposure can be expressed as a single exponential function with a half-life of 110 days (figure 5). This biological half-life for Cs^{137} is in good agreement with the value of 140 days obtained by Anderson et al (1957) in a clinical tracer study.

The Rongelap body burden in 1958, per cent of equilibrium and the equilibrium value has been estimated from these excretion data (table 7) (Woodward et al, 1959). These values are subject to some uncertainties since it was necessary to use a number of generalizing assumptions to derive them. It is possible to check these values using different approaches. For example, the estimated burden of Sr^{90} in March 1958 equals 2 $\mu\mu$c/g of calcium and appears to be of the right order of magnitude when compared with data provided by bone biopsies. Two bone biopsies of vertebra and scapula on a Rongelap male performed at this time indicated a level of about 3.8 $\mu\mu$c/g of calcium. Using the normalization factor from vertebra to average skeleton (Schulert et al, 1959b), an average skeletal value of 2 $\mu\mu$c/g is obtained. Thus, the mean body

burden of Sr^{90} for exposed Rongelap people in 1958 was estimated to be approximately 2 mμc or about 9 per cent of the calculated equilibrium value of 23 mμc.

The body burden at equilibrium for Cs^{137} was estimated to be 1.3 μc (Woodward et al, 1959) (table 7), or about one-half of the ICRP MPC for non-industrial populations. This value, however, is too high, since the 1959 Cs^{137} body burdens appeared to have already leveled off. Preliminary analysis of the 1959 whole-body counting data indicates that the Cs^{137} levels had dropped slightly (0.53 μc) as compared with the 1958 values (Cohn, 1959). However, it is possible that the Cs^{137} intake is less than estimated, since much of the Rongelap food is imported and of lower Cs^{137} content.

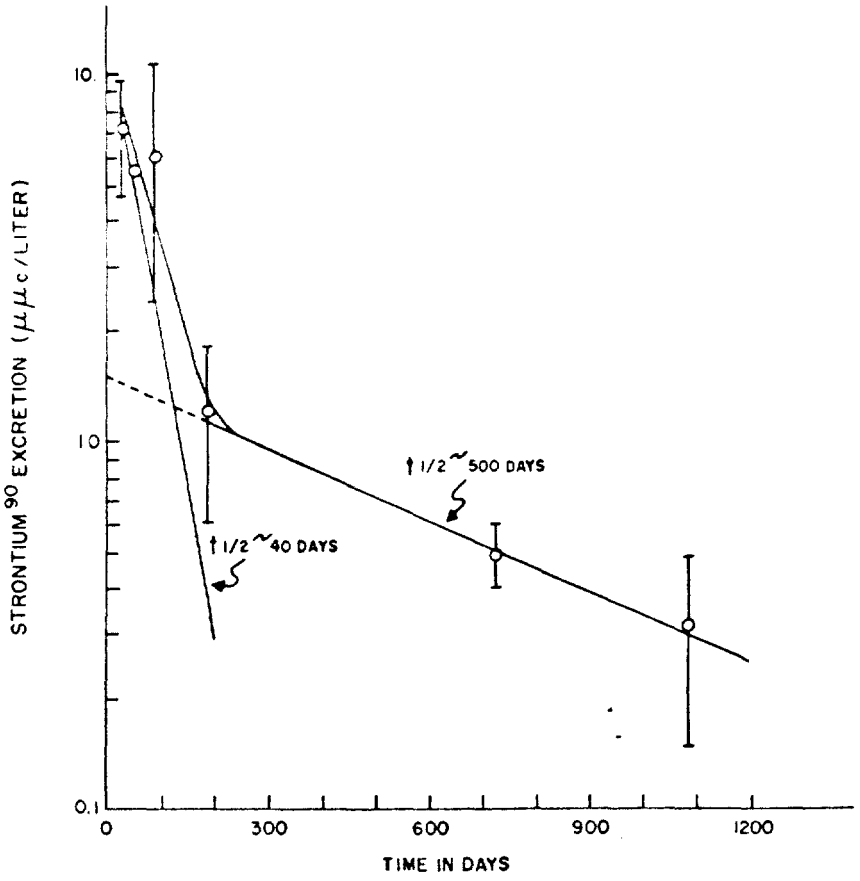

FIGURE 4. Strontium^{90} excretion in urine of exposed Rongelap people (from Woodward et al, 1959).

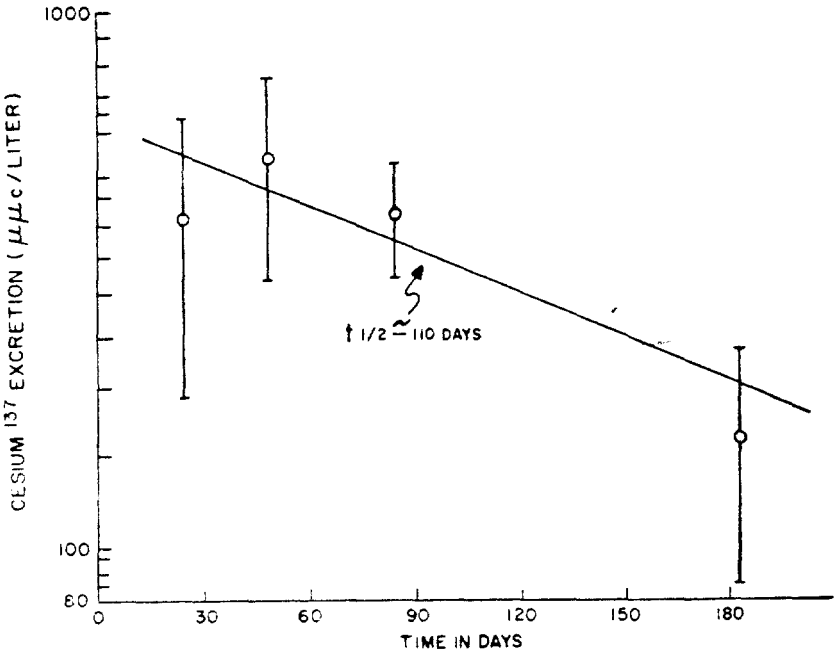

FIGURE 5. Cesium^{137} excretion in urine of exposed Rongelap people (from Woodward et al. 1959).

Zn^{65} was first detected by Miller (1957) in 1957 in the seven Marshallese examined by whole-body spectrometry, although it had been observed in high concentrations in fish as early as 1 year following the detonation (Rinehart, 1955). Body burdens of Zn^{65} in 1957, as measured directly, averaged 44 mμc for Rongelap inhabitants (figure 6), and 350 mμc in two Utirik inhabitants.

TABLE 7. Estimation of body burden of Rongelap population by urinary excretion levels, 1958 (data from Woodward et al, 1959)

	Sr^{90} (mμc)	Cs^{137} (mμc)		Zn^{65} (mμc)	
	Exposed	Exposed	Control	Exposed	Control
Body burden	2[a]	900	1,200	280	540
Equilibrated body burden	23	1,300	1,600	330	650
Per cent equilibrium	9	69	75	85	83
Daily intake	.015[b]	2(?)		2.1	4.1

[a] 3.7 Strontium Units (SU) determined by bone biopsy.
[b] 15 SU assuming daily calcium intake = 1 g.

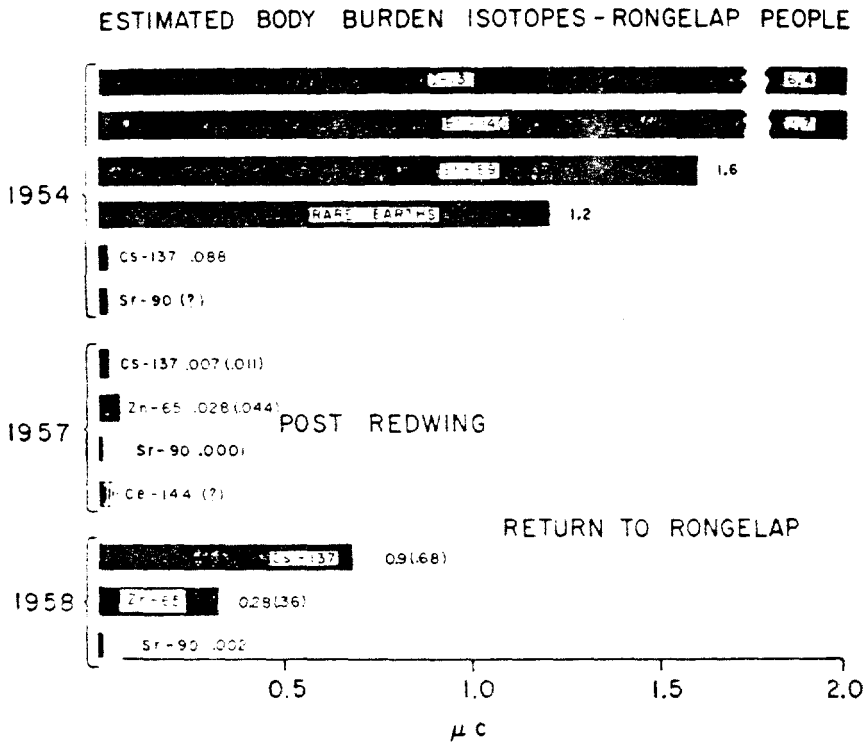

FIGURE 6. Estimated body burden of isotopes in Rongelap people; 1954-1958 (from Woodward et al, 1959).

Miller (1957) determined an effective half-life of 110 days for the elimination of Zn^{65} which gives a biological half-life of 200 days.

The mean body burden of Zn^{65}, estimated from the 1958 whole-body counting data, following the return of the Rongelap people to their island, was 8 times the 1957 whole-body measurement. The estimated intake of Zn^{65} of 2 to 4 mμc/day (Woodward et al, 1959) was derived from the fish in the diet. Zn^{65} levels in Rongelap fish muscle were 0.1 mμc/g in 1956 (Weiss et al, 1956). The directly measured mean body burden of Zn^{65} for Rongelap people in March 1958 (0.36 μc) is about 60 per cent of the estimated equilibrium value of 0.6 μc.

The estimates of body burden of Sr^{90}, Cs^{137} and Zn^{65} (1954-1958), derived both indirectly by urinalysis and directly by whole-body gamma counting, are presented in figure 6. The values in the parentheses are those obtained by whole-body gamma spectrometry, while the other values were obtained by extrapolation from urinalysis data. The body burden for Cs^{137} of 11 mμc (direct measurement) in 1957 indicates that the Rongelap people were exposed to a continu-

ing low level of Cs^{137} from stratospheric fallout during 1956 while they were living on Majuro. By contrast, the Cs^{137} mean body burden of the Utirik people (who were returned to their atoll in 1954) was 340 mμc, 30 times higher than that of the Rongelap people residing on Majuro (Woodward et al, 1959). This higher burden can be attributed to the higher level of Cs^{137} contamination in the environment of the Utirik people during this period of time.

The level of Cs^{137} in the body has fluctuated over the years since the original contaminating event. Unlike Sr^{90}, which is firmly fixed in the skeletal tissue, Cs^{137} has a relatively short biological half-life and thus readily reflects the environmental level. The slightly increased level of Cs^{137} during the 1956 and 1958 periods of weapon testing was thus rapidly reflected in an increased body burden in the Marshallese. A very marked increase in Cs^{137} was also observed in the Rongelap people after they returned to their original island in 1957. The Cs^{137} level in the Rongelap people in 1958 was about 0.68μc, about 60 times greater than the 1957 level, while the urinary Cs^{137} level rose 140 times.

The mean Zn^{65} body burden of the Marshallese, according to preliminary analysis of the 1959 whole-body counting data, is 0.44 μc, 8 per cent higher than the 1958 value (Cohn, 1959). These data would also appear to indicate that Zn^{65} deposition in the Rongelap people has not as yet reached equilibrium.

Summary and Conclusions

Since the nuclear fallout-producing detonation of 1954, in which a large land mass in the Marshall Islands and an isolated group of people were accidentally contaminated, studies have been in progress to determine the movement of fission products in the environment and in man himself.

While a large number of fission products are produced by a nuclear detonation, the majority have short half-lives, of the order of minutes or days. Still fewer gain entry into human tissues, as a result of the protective filtering mechanisms in the inhalation and ingestion systems of man. At early times following the contaminating accident, Sr^{89}, Ba^{140}, I^{131} and the shorter-lived iodine isotopes and some of the rare earth elements contribute the major portion of the internal dose. After a period of 1 year, the Sr^{90} contributes the greatest dose (to the skeletal tissue) and is the most critical fission product from the point of view of internal hazard. Cs^{137} and Zn^{65} have also been detected in tissues. These are present in small amounts, but are of interest because they yield information on the movement of the fission products from the environment to man. The dose from the internally deposited emitters was small compared to the concomitant external dose. Other than transitory changes in blood-cell levels, no pathological effects have as yet appeared as a result of this radiation.

Considerable data have been collected on the amount and kind of fission products in the soil, in various plants and numerous land animals and marine specimens. Radiochemical assays of specimens collected in yearly surveys have

yielded data on the transmission of fission products between successive members of the ecological chain.

Body burdens of gamma-emitting fission products (such as Cs^{137} and Zn^{65}), measured by use of a whole-body counter, have correlated rather closely with those estimated by radiochemical analysis of urine specimens. Estimates of the body burden may also be made from the Sr^{90}/Ca ratio in the dietary intake. The estimates of body burden derived from Sr^{90} g of calcium ratios in the soil through various steps of the ecological chain to man vary widely due to inherent difficulties in determining meaningful OR values from soil to plants.

The study of the fission products in the Marshallese people has been complicated in several ways. They were quickly evacuated from their island after the accident and did not return to their homes until 3 years later. In the interim 5 years since the accident, additional weapons tests have been held which have increased somewhat the amounts of the fission products in the environment. Finally, their diet now includes a variety of imported foods, so that they are not living in a "closed" environment, and therefore may not be rapidly approaching equilibrium with the fission products in it.

While the data collected in this study of a Pacific Island community give an indication of the internal radiation hazard resulting from acute and chronic exposure to *local* fallout, quantitative differences might be expected in different situations. For example, the physical and chemical properties of the fallout material, as well as meteorological conditions and properties of the biosphere, will influence strongly the uptake and retention of fission products in man via the soil-plant-animal cycle, and thus will determine the internal radiation hazard to man. For this reason, considerably more research is required on the transport of low levels of fission products (especially Sr^{90}) in food chains and biogeochemical cycles to determine the fate of radioisotopes in an environment, and thus to predict the potential internal radiation hazard to human beings living in fallout-contaminated areas.

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