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CESIUM-137 AND STRONTIUM-90 RETENTION
FOLLOWING AN ACUTE INGESTION
OF RONGELAP FOOD

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ABSTRACT

Marine and plant foods used by natives living on Rongelap in the Marshall Islands contain higher levels of long-lived fission-product radionuclides than do diets of people living in the United States due to residual contamination from fallout in 1954. During the 1963 medical survey of the Rongelap population, three food items indigenous to the Rongelap diet were brought back to the United States and consumed over a seven-day period by a member of the medical team. The ingestion of these foods introduced levels of ^{90}Sr and ^{137}Cs which were 20 and 60 times higher, respectively, than in the normal diet and was therefore considered in terms of an acute intake of two fission-product radionuclides that are important from a radiological standpoint. Urinary and fecal collections were analyzed separately, and whole-body ^{137}Cs measurements were made with a whole-body counter. The urine was the principal excretory route for the ^{137}Cs , whereas the feces was the main removal means for the ^{90}Sr . The retention of ^{90}Sr could be represented by a series of exponentials, whereas the retention of cesium as determined by whole-body counting indicated that a single long-term component with a biological half-life of 74 days describes the removal process. Reasonably good agreement was obtained between retention as determined by whole-body counting and by excretion measurements. It is estimated that about 25% of the ^{90}Sr from the Rongelap food was retained by the body at the end of 190 days. The

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average urinary to fecal excretion ratio was 3.5 for ^{137}Cs and 0.06 for ^{90}Sr . These findings are in agreement with other studies during which ^{90}Sr and ^{137}Cs were ingested under a variety of experimental and accidental conditions.

INTRODUCTION

Natives of Rongelap in the Marshall Islands consume indigenous plant and sea foods that contain long-lived fission-product radionuclides from fallout that occurred in 1954 during Operation Castle. The body burdens of these people, particularly with regard to ^{90}Sr and ^{137}Cs , are higher than those of U. S. inhabitants. Although estimates of total-body concentrations of these two radionuclides have been made directly for ^{137}Cs with a portable whole-body counter and indirectly for ^{90}Sr from urine analyses, the amount of foods consumed varies at different times of the year, and therefore information on the assimilation and excretion of specific nuclides as related to body burden has been difficult to evaluate. It was felt that a controlled intake and excretion study with the use of the ^{90}Sr and ^{137}Cs naturally present in Rongelap food would provide valuable data. Since controlled intake and excretion studies on Rongelap natives was not feasible in the field, it was decided by one of the authors (Robert A. Conard) that useful information in this regard might be obtained from an intake and excretion study carried out on himself using Rongelap foods brought back to Brookhaven National Laboratory (BNL), where a whole-body counting facility would be available for the study, and consumed under controlled conditions. Although these foods did not represent a typical native diet, it was felt that the relatively high levels of ^{90}Sr and ^{137}Cs present in them would make it possible to study quantitatively the excretion rates of these nuclides after an acute ingestion.

BACKGROUND

As part of the annual medical examinations of Rongelap natives who have been exposed to fallout radiation following the detonation of a high-yield thermonuclear device at Bikini Atoll in March 1954, 24-hr urine specimens are collected. These samples have been analyzed for ^{90}Sr at the Health and Safety Laboratory (HASL) during the past several years. The purpose of these analyses has been to attempt to relate excretion to total-body burden of ^{90}Sr resulting from the long-term chronic exposure of the natives to this radionuclide through ingestion. A portable whole-body counter was used during the 1961 survey, as in previous surveys, to measure the ^{137}Cs body burdens of the natives directly, but this instrument has not been available for

subsequent surveys. Therefore HASL was requested to analyze the urine specimens collected in 1963 and 1964 for ^{137}Cs as well as for ^{90}Sr .

In 1962, the year of the last reported survey,¹ the mean urinary ^{90}Sr level for individual adult 24-hr specimens was about 12 pc/liter, from which the body burden was estimated^{1,2} to be 12 nc. In comparison adults in a metabolic ward at Hines Hospital in Chicago, Ill., in 1962 were excreting an average of 0.5 pc of ^{90}Sr per liter of urine and, based on bone ^{90}Sr levels,³ probably had body burdens of around 0.5 nc. In 1961 the average ^{137}Cs body burden of the Rongelapese was 14.7 nc per kilogram of body weight compared to 0.048 nc per kilogram of body weight measured in U. S. medical-team personnel.^{2,4} The ^{90}Sr and ^{137}Cs body burdens of Rongelapese in 1961 and 1962 were therefore about 24 and 300 times higher, respectively, than those of individuals living in the United States. The average ^{137}Cs body burden of Rongelap natives in 1961 was as high as or higher than the burdens of Lapps and Eskimos, who have unusually high body burdens. Assuming that 50 kg is the average weight of a Rongelap native, one can calculate, based on the reported data, an average body burden in 1961 of 735 nc of ^{137}Cs . This value may be compared to the measurements⁵ made on Finnish Lapps in May 1962 which showed an average body burden of 508 nc of ^{137}Cs . Whole-body counts⁶ of Alaskans at Anaktuvuk Pass showed an average of 421 nc of ^{137}Cs during the summer of 1962. It is now clear that the high Rongelapese body burdens are the result of consuming the various types of contaminated food which come from the sea or are produced on Rongelap.² This is reasonable since the 1961 whole-body-count data showed that the mean ^{137}Cs levels of Rongelapese who were exposed to the heavy fallout in 1954 were not significantly different from the body burdens of those who were not exposed.⁴

During a single 24-hr period in September 1959, nine Rongelap total-diet samples were collected by the University of Washington Laboratory of Radiation Biology and subsequently analyzed for ^{137}Cs and ^{90}Sr as well as for other nuclides.⁷ The ^{137}Cs and the ^{90}Sr daily intakes averaged 2.4 and 0.084 nc, respectively. Although individual foods were not analyzed in this study, it was reported that pandanus was one of the highest contributors of these radionuclides to total diet.

MATERIALS AND METHODS

During the 1963 Rongelap medical survey, several kilograms each of pandanus pulp, coconut meat, and coconut milk were collected, frozen, and brought back to the United States. Over the seven-day period from July 2 to 8, 1963, one of the authors consumed 4.85, 1.75, and 3.20 kg of the pandanus, coconut meat, and coconut milk, respectively, in addition to a normal diet. Total fecal and urinary samples were collected for two consecutive three-day periods prior to the

consumption of the Rongelap food, for three- and four-day periods while the food was being consumed, and for two consecutive three-day periods after the last day on which the Rongelap food was consumed. Thereafter, for the next 184 days, 24-hr excreta samples were collected on Friday of each week. Whole-body counts were done at regular intervals.

Food aliquots and excreta specimens were sent to HASL for ^{90}Sr and ^{137}Cs analyses. The food was dry-ashed in a muffle furnace at approximately 450°C and solubilized by fusing with sodium carbonate and dissolving the pulverized melt in hot water and finally mineral acid. Cesium-137 was separated by the hot-water leach and then extracted from solution with ammonium phosphomolybdate. The cesium was purified by sorption on the selective elution from a cation-exchange resin and then precipitated as the tetraphenylborate for counting. A complete description of this procedure is given in the HASL procedural manual.⁸ The mineral-acid portion of the dissolved melt was evaporated to dryness in dilute acid. The dehydrated silica was filtered, and a carbonate-collection precipitation was carried out on the solution. After filtration the carbonate precipitate was dissolved in nitric acid, and the solution was evaporated to dryness. Successive fuming nitric acid separations were used to separate strontium from calcium and other interfering ions. Radium and lead were removed by scavenging with barium precipitated as the chromate. Traces of other fission products were scavenged with yttrium hydroxide. After equilibration of the ^{90}Sr with its daughter, ^{90}Y was precipitated as the hydroxide and converted to the oxalate for counting. This procedure is also described in the HASL manual.⁸ All counting was done with low-level beta scintillation counters designed at HASL.⁹ The urinary and the fecal samples were wet-ashed with nitric acid, and the residues were fused to effect complete dissolution. From this point, identical procedures to those previously described were used to separate ^{90}Sr and ^{137}Cs .

Cesium-137 body burdens were measured with the BNL whole-body counter. Measurements were taken at convenient intervals during and after the consumption of the Rongelap food. These ^{137}Cs values were corrected for contributions of ^{137}Cs present in the normal diet. This correction was made with the assumption that the subject's normal diet during the experimental period was similar to that of non-milk-drinking BNL personnel during the same period. The normal body burden was assumed to have increased linearly¹⁰ from 6.35 nc in July to 10.8 nc at the end of December 1963.

RESULTS

The amount of ^{90}Sr and ^{137}Cs ingested via the Rongelap food over the seven-day period is shown in Table 1. Pandanus contributed the

Table 1—INTAKE DATA FOR ^{90}Sr AND ^{137}Cs IN RONGELAP FOOD, JULY 2 TO 8, 1963

Food item	Intake for seven-day period		
	^{90}Sr , pc	^{137}Cs , pc	Calcium, g
Pandanus	4,379	57,327	0.992
Coconut milk	56	5,656	0.329
Coconut meat	48	2,824	0.460
Total intake	4,483	65,807	1.781
Average intake per day	640	9,401	0.254
Picocuries of ^{90}Sr per gram of calcium	2,517		

major portion of the activity of both radionuclides. The average ^{90}Sr and ^{137}Cs intake per day from these Rongelap foods was about 20 and 60 times higher, respectively, than from a normal New York City area diet during this time.¹¹ Initially, the activity from the Rongelap food essentially masked the contribution from the normal diet, but later in the study the effect of the normal diet on the excretion rates became increasingly important. In retrospect, it was unfortunate that the normal diet of the subject was not measured because the ^{90}Sr and ^{137}Cs levels in foods were increasing during this period.

The ^{90}Sr and the ^{137}Cs levels found in the urinary and the fecal collections are given in Table 2 and are plotted as a function of time in Figs. 1 and 2. Calcium measurements were made on the food and excreta samples, but, since the calcium intake from the normal diet was not known, the urinary and fecal calcium excretion data are difficult to interpret quantitatively. The average urinary calcium was 170 ± 40 mg/day, and the average fecal calcium was 360 ± 150 mg/day. These values do not indicate a metabolic abnormality, although the low fecal calcium reflects a very high absorption.

In Fig. 1 the ^{137}Cs excretion via the urine and the feces is plotted. The smoothed curves through the data points were drawn by eye. As has been observed in other studies,¹²⁻¹⁷ the main means of ^{137}Cs excretion is urine. Fecal excretion fell off very rapidly, indicating that only a relatively small amount of the ^{137}Cs ingested was not absorbed. Neither the urinary nor the fecal excretion rate fell to preexperiment levels at the end of 190 days. This was probably due to the increase in the normal diet ^{137}Cs which took place during this period.

The excretion pattern of ^{90}Sr as shown in Fig. 2 contrasts markedly with that of ^{137}Cs . The fact that the majority of the ingested ^{90}Sr was not absorbed by the body is evidenced by the high fecal excretion levels that were observed almost immediately following the consumption of

Table 2—⁹⁰Sr, ¹³⁷Cs, AND CALCIUM IN EXCRETA

Sampling period, 1963	Series	Urine, ml	Feces, g	⁹⁰ Sr, pc		Ca, g		¹³⁷ Cs, pc		⁹⁰ Sr, pc/day		Ca, g/day		¹³⁷ Cs, pc/day		Pc of ⁹⁰ Sr per g of Ca	
				Liter of urine	Kg of feces	Liter of urine	Kg of feces	Liter of urine	Kg of feces	Urine	Feces	Urine	Feces	Urine	Feces	Urine	Feces
June 26-28	Pre	2520	645	3.1	48	0.25	0.84	58	50	2.6	10.2	0.21	0.18	49	11	12	57
June 29- July 1	Pre	3430	573	2.6	110	0.19	1.88	43	73	3.0	21.0	0.21	0.35	49	14	14	58
July 2-4	Rongelap food	4240	1081	10.8	991	0.11	1.53	431	707	15.3	357	0.15	0.55	609	255	98	618
July 5-8	Rongelap food	4420	809	8.6	1406	0.06	1.97	617	1835	9.5	284	0.06	0.39	682	371	143	714
July 9-11	Post	3610	482	10.8	264	0.13	1.28	439	1102	13.0	42.4	0.15	0.20	528	177	83	206
July 12-14	Post	3900	544	*	153	*	2.49	*	709	*	27.8	*	0.45	*	128	*	61
July 19	Post	1150	327	4.7	*	0.14	0.57	508	306	5.4	*	0.16	0.18	584	100	34	*
July 26	Post	609	211	5.9	166	0.17	1.48	710	514	4.1	35.0	0.12	0.31	490	108	35	112
Aug. 8	Post	905	186	4.7	138	0.17	1.31	513	581	4.2	25.7	0.15	0.24	464	108	28	105
Aug. 9	Post	1900	156	1.7	121	0.09	2.12	218	591	3.3	18.9	0.17	0.33	414	93	19	57
Aug. 16	Post	1350	297	3.0	104	0.15	2.22	341	*	4.0	31.9	0.20	0.66	460	*	20	47
Aug. 23	Post	1280	270	2.4	131	0.11	2.66	273	350	3.1	35.4	0.14	0.72	349	95	22	49
Aug. 30	Post	770	221	3.6	75	0.16	1.15	297	357	2.8	16.5	0.12	0.25	229	79	23	65
Sept. 6	Post	660	441	3.9	46	0.19	0.79	*	135	2.6	20.2	0.12	0.35	*	60	20	58
Sept. 13	Post	†	176	†	97	†	2.22	†	557	†	17.0	†	0.39	†	98	†	44
Sept. 20	Post	678	†	3.7	†	0.29	†	295	†	2.5	†	0.20	†	200	†	13	†
Sept. 21	Post	1040	†	2.9	†	0.19	†	282	†	3.0	†	0.20	†	293	†	15	†
Oct. 4	Post	920	160	3.0	105	0.26	2.30	236	386	2.7	16.7	0.24	0.37	217	62	11	46
Oct. 21	Post	1180	230	2.6	109	0.19	2.30	219	274	3.0	25.1	0.22	0.53	258	63	13	47
Nov. 5	Post	680	320	4.7	†	0.24	1.22	217	299	3.2	†	0.16	0.39	148	96	20	*
Dec. 6	Post	1100	60	2.5	172	0.21	3.33	128	417	2.7	10.3	0.23	0.20	141	25	12	52
Jan. 7†	Post	780	94	3.2	149	0.18	2.76	196	356	2.5	14.0	0.14	0.26	152	34	18	54

*Sample lost.

†No sample received.

‡This sampling date was in 1964.

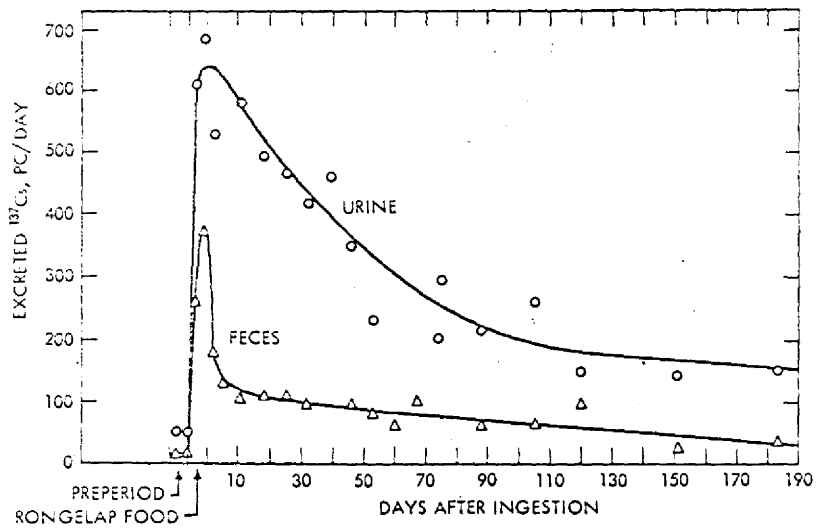


Fig. 1—Excretion of ^{137}Cs following ingestion of Rongelap food.

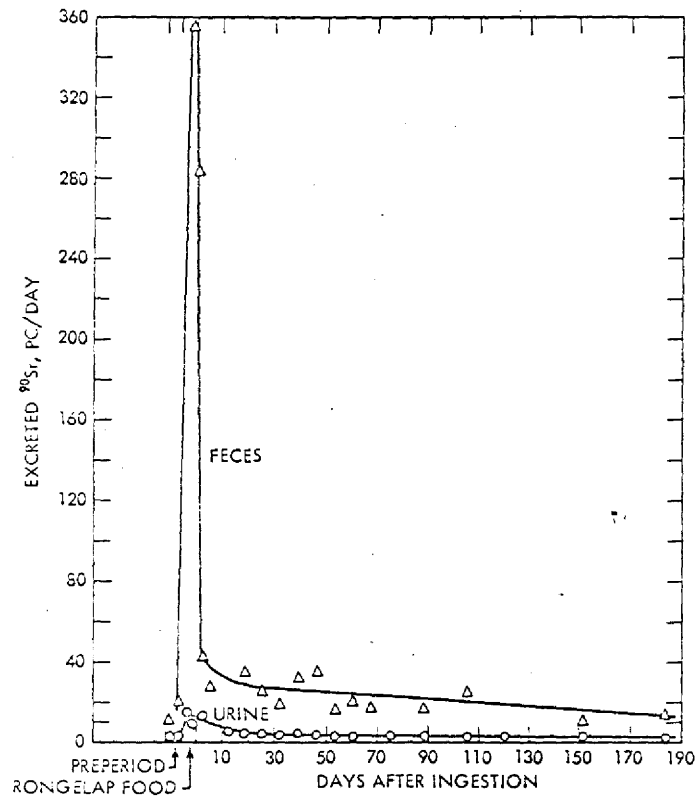


Fig. 2—Excretion of ^{90}Sr following ingestion of Rongelap food.

the high-activity food. Only a small increase in urinary ^{90}Sr excretion was observed, and both fecal and urinary elimination rates fell off sharply within 10 days after the acute-ingestion period. By 180 days both urinary and fecal excretion of ^{90}Sr had dropped to pre-high-intake levels.

Through integration of the smoothed excretion-rate curves, the amounts of ^{137}Cs and ^{90}Sr excreted per 10-day interval were determined. Excretions during the 7-day acute-ingestion period and the first 10 days after the high-activity-food consumption terminated were summed over the entire 17-day period. With the assumption that the increase in the ^{137}Cs excretion rate of the subject was proportional to the increase in the normal body burdens of laboratory personnel, a background correction was determined and applied to each 10-day excretion value. Since the ^{90}Sr excretion rate returned to preexperiment levels toward the end of the study, it was assumed that any increase in the excretion rate due to an increase in the normal ^{90}Sr diet level was not measurable. Therefore only the preexperiment ^{90}Sr level was subtracted.

The background-corrected cumulative excretions expressed as percent of intake are plotted against time in Figs. 3 and 4. Fifty percent of the ^{137}Cs ingested via the Rongelap food had been excreted in urine after 85 days, whereas only 14% had been excreted in feces during the same time. In contrast, almost 50% of the ^{90}Sr dose had been excreted in feces at 10 days, whereas only 2.5% had been eliminated in urine.

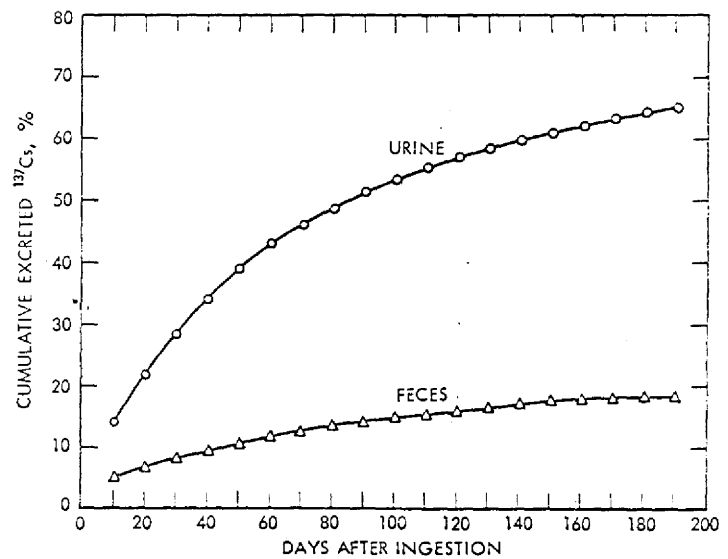


Fig. 3—Cumulative urinary and fecal excretions of ^{137}Cs following ingestion of Rongelap food.

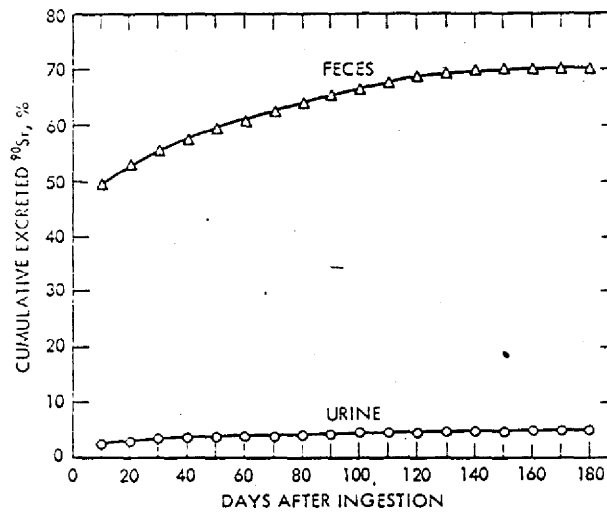


Fig. 1—Cumulative urinary and fecal excretions of ^{90}Sr following ingestion of Rongelap food.

Since the integration limits were relatively large initially compared to the rate of change of excretion level, the cumulative curves were not extrapolated beyond the point at 10 days after termination of the high-activity-food consumption. The urinary to fecal ^{137}Cs excretion ratio averaged 3.5, ranging between 2.8 and 3.8, and the ^{90}Sr excretion ratio averaged 0.06, ranging between 0.05 and 0.07.

The cumulative excretion data were subtracted from the total intake to obtain the amount of each radionuclide retained. These results along with the BNL whole-body ^{137}Cs measurements are given in Table 3. When the two sets of ^{137}Cs data are compared, they appear to be in reasonably good agreement with each other. The fact that the whole-body measurements are the more accurate, however, is seen from the semilog plot in Fig. 5. The expected single exponential fit demonstrates that the ^{137}Cs from Rongelap food behaves in a similar metabolic manner to that observed under other intake conditions.¹²⁻¹⁷ The deviations from a single exponential function of the retention data as derived from excretion measurements may be attributed to inaccuracies in the estimation of the contributions from the normal dietary intake and the accumulation of errors inherent in the procedure. The biological half-life as measured by the whole-body counter is 74 days, whereas the apparent half-life as measured on the initial straight-line portion of the excretion-data curve is 64 days. When it is considered that the excretion data underestimate the retention slightly in the early stages of measurement, the agreement is not unreasonable. It should be pointed out that the short-lived (one to two days) component that has

Table 3— THE RETENTION OF ^{137}Cs IN RONGELAP FOOD
(MEASURED BY WHOLE-BODY COUNTER AND FROM EXCRETION)
AND THE RETENTION OF ^{90}Sr (MEASURED FROM EXCRETION)

Days after ingestion of Rongelap food	^{137}Cs retention, nc		^{90}Sr retention, nc
	Whole-body counter	Excretion	
1	57.90		
2	57.10		
3	55.71		
4	55.64		
7	53.21		
10		53.34	2.17
11	51.00		
18	46.83		
20		47.22	1.98
25	45.38		
30		42.10	1.86
32	42.56		
39	38.88		
40		37.71	1.76
46	36.80		
50		33.73	1.66
53	36.16		
60	32.88	30.42	1.60
70		27.53	1.52
74	27.73		
80		24.97	1.44
88	25.68		
90		22.96	1.37
100		21.02	1.32
105	22.11		
110		19.38	1.28
120	19.19	17.91	1.21
120		16.56	1.20
140		15.26	1.17
150	14.34	14.22	1.16
160		13.22	1.14
170		12.33	1.14
180		11.50	1.14
184	9.37		
190		10.81	

been observed by other investigators^{14,16,17} was not measured in this study since the acute-ingestion period was longer than the expected half-life of the short-term component.

The ^{90}Sr retention curve as determined from excretion data (Fig. 6) does not describe a single exponential function as is the case with ^{137}Cs . The whole-body retention of ^{90}Sr is more closely represented by a sum of exponentials^{18,19} or a power function.^{19,20} In this

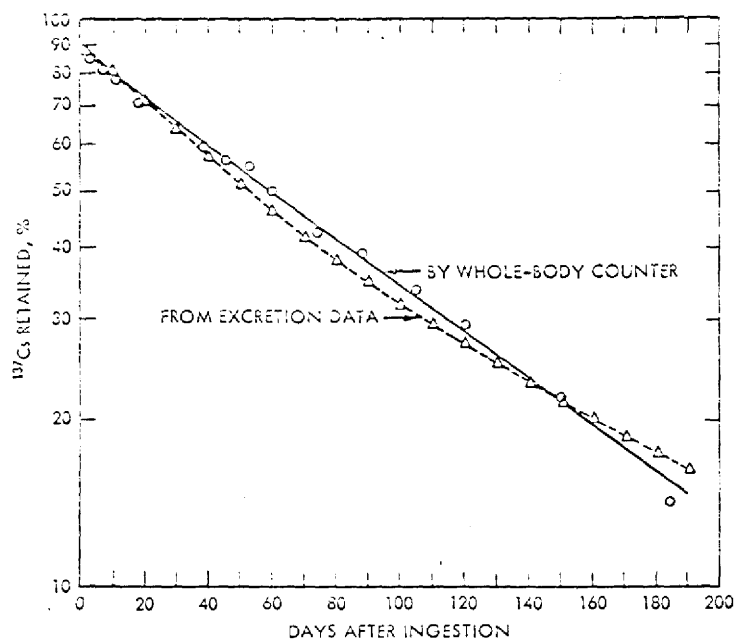


Fig. 5—Percent retention of ^{137}Cs from Rongelap food.

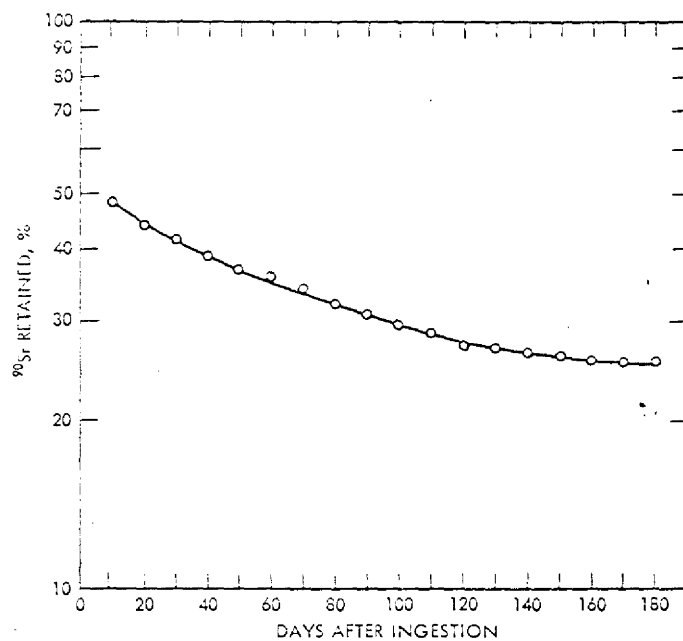


Fig. 6—Percent retention of ^{90}Sr from Rongelap food as measured from excretion.

study the retention curve is best described as a series of exponentials that level off after 140 days and approach a value of 25%. This is a somewhat higher retention value than that reported in other studies.²¹

DISCUSSION

A number of investigators have shown that the whole-body retention for a single administration of ¹³⁷Cs can be expressed by a two-component exponential function of time:^{14,16,17}

$$R_t = (1 - a) e^{-(0.693/T_1)t} + a e^{-(0.693/T_2)t}$$

where R_t is the fractional retention at t days, a is a constant, and T_1 and T_2 are biological half-lives in days. The short-term component has a half-life, T_1 , of about one day,^{14,16,17} and the principle component has a half-life, T_2 , that varies from study to study, ranging from 50 to 150 days (Refs. 12 to 16 and 22 to 25). The comparison of biological half-lives under conditions of both acute and chronic exposures is justified based on the work of Rundo.²⁶ The short-term component was not calculated in this study since the acute ingestion took place over a seven-day period and the initial excretion collection periods were over three-day intervals. The long-term component as determined by both whole-body counting and excretion data (74 and 64 days, respectively) was within the range of biological half-life values found by other investigators. Rundo¹⁴ has pointed out that a large part of the variations in body burden can be attributed to the variability in biological half-life rather than to differences in dietary habits. The low excretion of ¹³⁷Cs in feces relative to urine showed that the ¹³⁷Cs in the Rongelap food was rapidly and almost completely absorbed. Since the finding that a single long-term component describes the excretion from the whole body is in agreement with other studies, it can be said that the ¹³⁷Cs in Rongelap food is not in a unique chemical form with respect to the various accidental and experimental intake conditions under which the behavior of ¹³⁷Cs in man has been studied.

The urinary to fecal ratio was found to be reasonably constant and was within the range of 3 to 10 found by others.¹²⁻¹⁶ Rundo¹⁶ has used the urinary to fecal ratio to estimate the uptake of ¹³⁷Cs ingested in food. The fraction of ¹³⁷Cs ingested in food which is transferred from the gastrointestinal tract to blood was at least 0.9. The data obtained in this study do not indicate that the ¹³⁷Cs in Rongelap food behaves in a significantly different manner.

The excretion pattern of the ⁹⁰Sr from the Rongelap food was markedly different from that of ¹³⁷Cs. Most of the ⁹⁰Sr was not absorbed, as evidenced by the much higher fecal than urinary excretion.

The net absorption (intake-fecal excretion expressed as percent of intake) was 30%, a factor of 2 higher than the average for 10 normal adults on a high-calcium diet reported by Spencer et al.²¹ The low fecal calcium excretion found in the present study was indicative of a high ⁹⁰Sr absorption.

Although the retention curve for ⁹⁰Sr shows that the biological half-life varies with time after exposure, the data do not fit a power function as found by others.¹⁹⁻²⁰ If it had been possible to measure excreta over a longer period of time, a power function may have been described.

SUMMARY

The ¹³⁷Cs and the ⁹⁰Sr body burdens of people living on Rongelap Island are high compared to most other populations of the world. The reason for this is that the natives consume foods that are contaminated with long-lived fission-product radioactivity resulting from a fallout incursion in 1954. Their ¹³⁷Cs body burdens are comparable to those of people living in other limited areas such as Lapland and northern Alaska where unique ecological conditions are conducive to high ¹³⁷Cs concentrations in indigenous foods. The metabolism of ¹³⁷Cs and ⁹⁰Sr has been studied in the Lapland and Alaskan groups but not in the Rongelap natives.

Since facilities for a metabolic-balance study were not available on Rongelap Island, several native food items were brought back to BNL and consumed by one of the authors under controlled conditions. Urinary and fecal specimens were collected and whole-body counting measurements were made over a period of 180 days. The intake of ⁹⁰Sr over a seven-day period was 20 times higher than normal and that of ¹³⁷Cs was 60 times higher than normal.

Fifty percent of the ingested ¹³⁷Cs in the Rongelap food had been excreted in urine after 85 days, whereas 14% had been eliminated in feces during the same time. In contrast, most of the ⁹⁰Sr was unabsorbed. Fifty percent had been excreted in feces at 10 days, whereas only 2½% had been excreted in urine. The retention of ¹³⁷Cs as determined by both whole-body counting and excretion measurements showed a biological half-life of 74 days. Strontium-90 retention as a function of time was best described as a series of exponentials and approached a value of 25% after 140 days.

These findings fall within the range of results of many other studies conducted under a wide variety of natural, accidental, and experimental conditions.

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