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METABOLISM OF FISSION PRODUCTS IN MAN: MARSHALLESE EXPERIENCE

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Abstract — Résumé — Аннотация — Resumen

METABOLISM OF RADIOACTIVE FISSION PRODUCTS IN MAN: DATA FROM THE MARSHALLESE. The medical study of the Marshallese accidentally exposed to local fall-out in 1954 is unique in that, along with the Japanese fishermen study, it provides the only data existing on the metabolism of mixed fission products in a human population. Early diagnosis of the internal radioactive contamination was made by radiochemical analysis of the excreta of the exposed people and by radiochemical analysis of the tissues of animals simultaneously exposed. Initially, $\text{Sr}^{90}\text{Ba}^{140}$, I^{131} and its shorter-lived daughters and a number of rare-earth elements contributed the major portion of the internal radiation dose.

After a year, the principal radioisotopes were Sr^{90} , Cs^{137} and Zn^{65} . Subsequently these radionuclides and more recently, Co^{60} , have been measured periodically. Since 1958, the γ -spectra of a number of Marshallese have been obtained with a portable whole-body counter.

The report discusses the findings of these studies for the past eight years. The results of an early attempt to alter the rate of removal of the mixed fission products in the Marshallese with calcium disodium EDTA are presented. The metabolism of the radionuclides and their relationship to levels present in the environment is also discussed.

MÉTABOLISME DES PRODUITS DE FISSION RADIOACTIFS CHEZ L'HOMME. DONNÉES RECUEILLIES AUX ÎLES MARSHALL. L'enquête médicale à laquelle ont été soumis les habitants des îles Marshall qui, en 1954, ont été exposés accidentellement à des retombées locales présente un caractère unique en ce sens que, tout comme l'enquête sur les pêcheurs japonais, elle fournit les seules données que l'on possède sur le métabolisme d'un mélange de produits de fission chez l'homme. Un premier diagnostic de la contamination radioactive interne a été fait par analyse radiochimique des excréta des sujets exposés et par analyse radiochimique des tissus des animaux exposés simultanément. Au début, ^{90}Sr , ^{140}Ba , ^{131}I et leurs produits de filiation de courte période, ainsi qu'un certain nombre de terres rares, produisaient la majeure partie de la dose de rayonnements internes.

Après une année, les principaux radioisotopes étaient ^{90}Sr , ^{137}Cs et ^{65}Zn . Par la suite, on a mesuré périodiquement ces radionucléides et, plus récemment, ^{60}Co . Depuis 1958, on a pu obtenir, à l'aide d'un appareil portatif de dosage de l'activité du corps humain, les spectres gamma d'un certain nombre d'habitants des îles Marshall.

L'auteur étudie les résultats de ces enquêtes, obtenus au cours des huit dernières années. Il présente également les résultats d'une tentative faite au début pour modifier la vitesse d'élimination des produits de fission mixtes chez les autochtones au moyen d'un sel double de calcium et de sodium de l'EDTA. Il étudie enfin le métabolisme des radionucléides et leurs rapports avec les concentrations présentes dans le milieu ambiant.

МЕТАБОЛИЗМ РАДИОАКТИВНЫХ ПРОДУКТОВ ДЕЛЕНИЯ В ОРГАНИЗМЕ ЧЕЛОВЕКА. (ДАНИЕ ОБСЛЕДОВАНИЯ ЖИТЕЛЕЙ МАРШАЛЬСКИХ ОСТРОВОВ.) Медицинское обследование жителей Маршалльских островов, случайно подвергшихся действию местных радиоактивных осадков в 1954 году, является уникальным в том смысле, что оно, наряду с обследованием японских рыбаков, дает возможность получить единственные в этой области данные по метаболизму смешанных продуктов деления в организмах людей. Ранняя диагностика внутреннего радиоактивного заражения осуществлялась с помощью радиохимического анализа выделений облученных людей и методами радиохимического анализа тканей животных облученных одновременно. На начальной стадии большую часть внутренней дозы радиации составляли стронций-89, барий-140, йод-131, их более короткоживущие дочерние продукты и ряд редкоземельных элементов.

Через год основными радионуклидами были стронций-90, цезий-137 и цинк-65. Позднее периодически измерялась активность этих радионуклидов, а еще позднее - и кобальта-60. Начиная с 1958 го-

да гамма-спектры у жителей Маршалльских островов получают с помощью портативного счетчика для измерения активности всего тела.

В докладе обсуждаются данные этих исследований за последние восемь лет. Приводятся результаты ранних попыток изменить скорость удаления смешанных продуктов деления из организма с помощью двуназиевого кальция EDTA. Обсуждается также метаболизм радионуклидов и связь их с уровнями в окружающей среде.

METABOLISMO DE PRODUCTOS DE FISIÓN RADIACTIVOS EN EL HOMBRE: DATOS OBTENIDOS EN LOS HABITANTES DE LAS ISLAS MARSHALL. El estudio clínico de los habitantes de las islas Marshall que en 1954 quedaron expuestos accidentalmente a precipitaciones radiactivas posee gran valor científico porque, junto con el caso de los pescadores japoneses, constituye la única fuente de datos sobre el metabolismo de mezclas de productos de fisión en el hombre. Al principio, el diagnóstico de la contaminación radiactiva interna se efectuó por análisis radioquímico de los tejidos de animales expuestos simultáneamente. Se encontró que la mayor parte de la dosis de radiación interna se debía al ^{89}Sr , ^{140}Ba , ^{131}I y sus descendientes de período breve, así como a algunos de los elementos de las tierras raras.

Al cabo de un año, los principales radioisótopos presentes eran el ^{90}Sr , ^{137}Cs y ^{65}Zn . Después se siguieron midiendo periódicamente estos radionúclidos y, más recientemente, el ^{60}Co . Desde 1958, se han obtenido los espectros y de un cierto número de pobladores de aquellas islas por recuento del organismo entero con un aparato portátil.

El informe discute los datos que durante los últimos ocho años se han obtenido gracias a estos estudios. Presenta los resultados de un tratamiento con la sal cálcico-disódica del EDTA, que se efectuó poco después de la contaminación con el propósito de modificar la velocidad de eliminación de las mezclas de productos de fisión en los habitantes de las islas Marshall. También discute el metabolismo de los radionúclidos y su relación con la radiactividad ambiente en aquella zona.

INTRODUCTION

Information on the metabolism of fission products in man has been obtained largely from studies carried out with parenterally administered soluble salts of radioisotopes administered in medical treatment or in tracer studies. The recent development of the whole-body γ -spectrometer with its highly sensitive detection system has been of considerable value in extending these studies by providing data on very low levels of isotopes in man over long periods of time.

The evaluation of the internal radiation hazard resulting from acute exposure to mixed fission products requires more data than are available from these research studies, however. One point of divergence rests on the fact that while in most clinical and laboratory work the isotope is administered parenterally, an exposure to mixed fission products would occur as an inhalation and ingestion exposure. In the latter case the isotopes are filtered by the lungs or by the membranes of the gastro-intestinal tract. It is also necessary to determine whether an isotope administered singly will behave in like manner when administered in combination with numerous other isotopes. Further, it is necessary to determine the effects of the varying physical forms and particle sizes encountered with the isotopes formed in fall-out.

Because of the difficulties involved, little research has been performed with the entire spectrum of mixed fission products. It has therefore been of very particular importance to keep under surveillance the group of Marshallese people accidentally exposed to fall-out in 1954. These people and the Japanese fishermen who were simultaneously accidentally exposed are unique in that they constitute the only groups of people who have been

exposed to radioactive contamination by mixed fission products from a nuclear detonation.

In the Marshallese experience, 239 individuals were accidentally exposed to local fall-out for periods up to two days. The levels of the associated radiation fields were estimated to vary between 14 and 175 r for the different Marshallese groups [1-3]. The radiation exposure from the internal emitters following this acute exposure was insignificant compared with the dose received from the external radiation, but nevertheless it is of great interest and importance to follow the course of the isotopes in humans. As part of the continuing medical care and follow-up of these people, a study of the metabolism of internal emitters, inhaled or ingested, by the inhabitants of Rongelap Island (the group receiving the highest exposure) has been carried out [4, 5].

In addition to the investigation of the acute exposure situation, it is of considerable importance to obtain data on the metabolism of fission products and neutron-induced radionuclides encountered in low-level chronic exposures. This report will review the pertinent findings of the Marshall Island studies on the levels of the various radionuclides in the Marshallese in relationship to their environment over the past eight years. This relationship is of particular interest inasmuch as the Marshallese, since their return to Rongelap in June 1957, have lived in an environment of persisting low levels of contamination. These levels are far below the value considered to be hazardous, but are nevertheless readily detectable with the whole-body counter.

Laboratory and field studies have indicated that relatively few of the 250 radioisotopes formed in the fission process are significant from the biological point of view. Only those products which are produced in significant quantity and which are sufficiently long-lived to become integrated into the ecological environment are listed in Table I. Since fall-out consists of various neutron-induced radionuclides in addition to fission products, they are also listed in the table. The presence of radionuclides in man depends first on their physical availability in the environment. All of the radionuclides listed in Table I were found in plants and animals on the Marshall Islands or in the lagoon. The biological availability of an element to a plant or animal and ultimately to man is also dependent to a large extent upon the solubility and physical state of the element. Of these fission products listed in Table I, only the first seven are absorbed from the gastro-intestinal tract in amounts exceeding 1% of the quantity available. The remaining fission products have been assigned (ICRP-1959) a high absorption via the respiratory tract even though the GI absorption is quite low.

In the present situation, in which the nuclear device was detonated on or near the ground of a Pacific Island, the bulk of the radioactivity resided as insoluble oxides on particles of CaO or Ca(OH)₂ made by the heat of the fireball acting on the coral of the Islands. A large amount of activity was also carried on NaCl particles. A different physical-chemical nature of fall-out in another situation would modify the metabolism of the fission products to some extent.

In general, the radionuclides that form strong complexes with organic matter (such as the transition elements) and those present as particulate or colloidal matter are found in marine organisms. These are Co^{57, 58, 60},

TABLE I
BIOLOGICALLY IMPORTANT FISSION PRODUCTS IN FALL-OUT

Radionuclide	Mass yield		Type of radiation	Half-life (d)			Fraction entering body	
	U ²³⁵ *	U ²³⁸ *		Physical	Biological	Effective	by ingestion	by ingestion
	%	%		T _f	T _b	T	fw	fa
I ¹³¹	3.1	-	β, γ	8	7	7.6	1.0	0.75
Cs ¹³⁷	6.15	6.2	β, γ	1.1x10 ⁴	140	138	1.0	0.75
Sr ⁹⁰ -Y ⁹⁰	5.77	3.2	β	10 ⁴	1.8x10 ⁴	6.4x10 ³	0.30	0.40
Sr ⁹⁰	4.79	2.9	β	50.5	1.8x10 ⁴	50.4	0.30	0.40
Ba ¹⁴⁰ -La ¹⁴⁰	6.4	5.7	β, γ	12.8	65	10.7	0.06	0.28
Ru ¹⁰⁶ -Rh ¹⁰⁶	0.38	2.7	β, γ	365	7.3	7.2	0.03	0.27
Ru ¹⁰⁶	3.0	6.6	β, γ	4.1	7.3	6.2	0.03	0.27
Pm ¹⁴⁷	2.7	-	β	920	656	383	10 ⁻⁴	0.25
Ce ¹⁴⁴ -Pr ¹⁴⁴	6.0	4.9	β, γ	290	563	191	10 ⁻⁴	0.25
Zr ⁹⁵ -Nb ⁹⁵	6.2	5.7	β, γ	63.3	450	55.5	10 ⁻⁴	0.25
Ce ¹⁴¹	6.0	-	β, γ	32	563	30	10 ⁻⁴	0.25
<u>Neutron-induced activities</u>								
Zn ⁶⁵	0	0	β, γ	245	350	144	0.10	0.30
Co ⁶⁰	0	0	β, γ	1.9x10 ³	9.5	9.5	0.30	0.40
Mn ⁵⁴	0	0	β, γ	300	17	5.6	0.10	0.30
Fe ⁵⁵	0	0	β, γ	1.1x10 ³	800	463	0.10	0.30

*U²³⁵ - Thermal neutron fission - nuclear power reactors

U²³⁸ - Fast neutron fission - nuclear weapon testing

Fe^{55,59}, Zn⁶⁵, Mn⁵⁴ and the fission products Zr⁹⁵ - Nb⁹⁵, Ru¹⁰⁶ - Rh¹⁰⁶, and Ce¹⁴⁴ - Pr¹⁴⁴. In the terrestrial environment, the fall-out material taken up in greatest amounts by land plants and man are those which are most highly soluble, i. e. Cs¹³⁷ and Sr⁹⁰.

While few of the neutron-induced activities were present in the island soil or in plants, they make up to 100% of the activity in plankton and fish. Thus the neutron-induced activities and the fission products listed above are taken up by marine forms and get into the diet of man via fish, clams and crabs [5]. Both these radionuclides are incorporated into plants via root or foliar absorption, or as external contamination on the leaves of plants. There are many factors that determine the uptake of these two nuclides by plants, such as solubility, soil chemistry and soil structure, competing ions, etc. The levels of these radionuclides in various plants and animals vary considerably, and consequently the body burdens of the people living in this environment vary greatly as a function of the composition of their varied diets.

Since the half-lives of the various fission products differ widely, the time of exposure after fission will determine the composition of the remaining fission product mixture. Immediately following the acute exposure, most of the radioactivity is contributed by short-lived radionuclides.

A. ACUTE EXPOSURE

In the Marshallese experience, the acute exposure lasted less than two days. The diagnosis of internal radioactive contamination was made early from the appearance of high levels of radioactivity in urine assays of the Marshall Islanders. The highest levels were found in the 82 inhabitants of Rongelap Island. During the two days that the people remained on Rongelap, before their evacuation, they lived in a radioactively contaminated environment and took no precautionary measures to avoid ingestion or inhalation of the material. The initial body burdens of internal emitters in the Rongelap people were estimated from data obtained by radiochemical analysis of the tissues of two pigs, which had been simultaneously exposed, and subsequent comparison of human and animal urinalysis data [4]. The mean body burden at one day was estimated (in μc) as Sr⁸⁹ -1.6, Ba¹⁴⁰ -2.7, I¹³¹ -6.4 and the rare-earth group together -1.2.

Information on the tissue distribution of fission products was obtained from the radiochemical analyses of the pigs living on Rongelap for one month and sacrificed two months later (Table II). 95% of the internal activity is localized in the skeleton. The alkaline earths, Sr⁸⁹ and Ba¹⁴⁰, and the rare earths together constitute 75% of the gross β -activity at 82 d. The activity in the pig was tenfold higher than that estimated in the people, reflecting the 30-d longer exposure of the animals. The pattern of deposition of the fission product mixture in the skeleton, as seen in an autoradiograph of a tibia of one of these pigs, resembles that seen after administration of alkaline earths, i. e. dense deposition in the epiphyseal region (Fig. 1).

In the first few months following this acute exposure, Sr⁸⁹ and I¹³¹ (plus the shorter-lived iodine isotopes) contributed the greatest internal radiation dose to the Marshallese. Sr⁸⁹ contributed the major portion of

TABLE II
INTERNAL RADIOACTIVE CONTAMINATION
OF MARSHALLESE PIGS EXPOSED TO
FALL-OUT FROM THE 1 MAR. 1954 NUCLEAR DETONATION*

	β -activity dpm/total sample $\times 10^{-3}$			
	Gross activity	Sr ⁹⁰	Ba ¹⁴⁰	Rare-earths
Skeleton (total)	8745 (100%)	5380 (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			

* These values are the average of two young adult pigs which were analysed three months after detonation.

the β -dose to the skeleton at this early time. The highest dose to an individual tissue (100 to 150 rep) was delivered to the thyroid by the iodine isotopes. This was estimated from the urinary data of the Marshallese in comparison with clinical data.

In accord with theoretical estimates, only a small percentage of the fission products initially found in environmental samples was found in the tissues of the body. The level of radioactivity found in the GI tract of the pigs, however, was high (3 mc), but because of its insoluble nature or the large size of the particles, only a small fraction of the fall-out was able to enter the systemic circulation [4].

Recently Van DILLA [6], by using a spectral analytical technique, was able to demonstrate that Pu²³⁹, Ce¹⁴⁴, Ru¹⁰⁶ and Zr⁹⁵, in the form of insoluble oxides from fall-out, were found in the rumen of cattle located near the Nevada test site, but were essentially absent (except for a trace of Zr⁹⁵ in the liver) in the tissues of the animals.

B. LONG-TERM STUDY

In evaluating the long-term effects resulting from both an acute exposure and chronic exposure to low-level residual contamination, Sr⁹⁰ is the critical element to measure. The determination of its level in the urine of the Marshallese has been made yearly to estimate the body burden.



Fig. 1

Autoradiograph of tibia of pig at one month following exposure to 1 March 1954 nuclear detonation
Left: Section of bone. Right: Autoradiograph of bone.
The black areas in autoradiograph are produced by radioactivity and mark the sites of deposition of fission products in the bone.

In addition, Cs^{137} , which is of minor significance as an internal radiation hazard, serves as a useful tracer for studying the movement of fall-out through the terrestrial environment to man. Like Sr^{90} , Cs^{137} has a gaseous precursor with a sufficiently long half-life to avoid early condensation in the fireball. Cs^{137} thus follows Sr^{90} into the stratosphere and is present in fall-out in like quantities. While there are significant dissimilarities in the ecological cycles and metabolic behaviour for the two isotopes, nevertheless there are some interesting parallels in their behaviour that make Cs^{137} useful in tracing the not-so-readily measurable Sr^{90} .

Zn^{65} , a neutron-induced radionuclide, is easily measured and has been followed since its identification in the Marshallese in 1957. The levels of Zn^{65} are also too low to be considered an internal radiation hazard. Interest in the movement of zinc through the environment centres on the fact that it is almost entirely transported through marine life and enters man through the seafood chain. Co^{60} , another neutron-induced radionuclide,

was detected and measured for the first time in the Marshallese in 1961. Its level is very low and was masked previously by the higher levels of the other radionuclides. As mentioned above, many other fission products and some other neutron-induced activities are present in the marine and terrestrial environmental samples, but their levels in man were too low to detect in this situation.

C. ESTIMATION OF BODY BURDENS

The body burdens of fission products in the Marshallese have been assessed in three ways: by whole-body γ -spectrometry, by radiochemical analysis of the urine or bone, and by estimation of body burden from dietary intake. The method of choice is the direct *in vivo* measurement with the whole-body spectrometer [7]. This method is, of course, restricted to the analysis of γ -emitters. A "portable" (20 t) whole-body counter was designed at the Brookhaven National Laboratory and transported aboard a ship to the Marshall Islands in 1958, 1959 and 1961 to measure the levels of internally-deposited γ -emitters in the radioactively contaminated population [8]. Details of the experimental procedures and the automatic data-handling techniques for analysing the complex multi-component spectra have been presented [8]. A typical γ -spectrum for an adult Marshallese male, as obtained with the whole-body counter in 1961, is shown in Fig. 2.

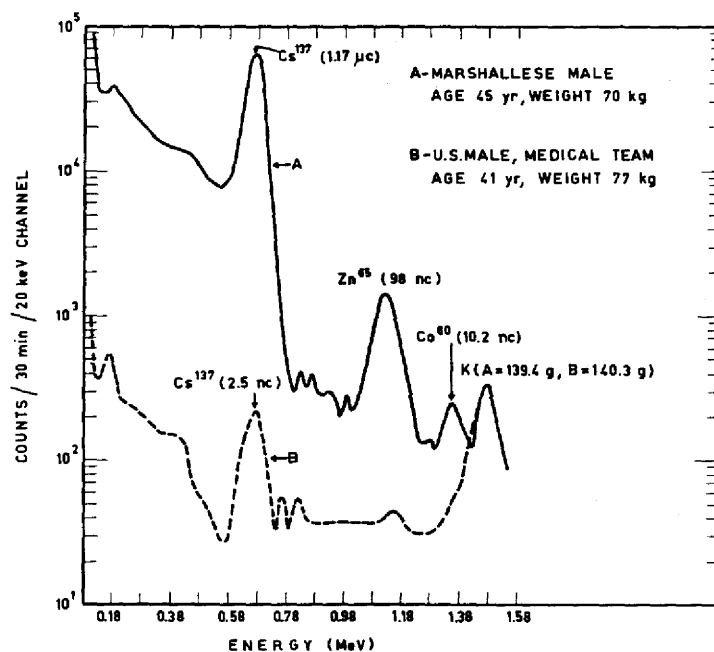


Fig. 2

Spectrum of typical Marshallese adult male compared with that of member of the US Medical team

The principal γ -photopeaks are those of K^{40} , Zn^{65} , Co^{60} and Cs^{137} . The spectrum of a member of the medical team of approximately the same age and body weight obtained at the same time is also shown for comparison.

For the β -emitter Sr^{90} , radiochemical analysis of the urine has provided data for estimating the body burden. In 1958 an estimate was made of the equilibrium body burden from the Sr^{90}/Ca level of the diet.

Strontium-90

The urinary excretion of Sr^{90} by the exposed Marshallese immediately after the acute exposure in 1954 is shown in Fig. 3. The Sr^{90} level in the urine fell rapidly. The excretion curve over the first three years could be described by the sum of two exponentials with half-lives of 40 and 500 d, respectively [9]. This latter value for the main component corresponds with a value of 850 d obtained in a clinical tracer study of parenterally-administered $Sr^{85}Cl_2$ [10].

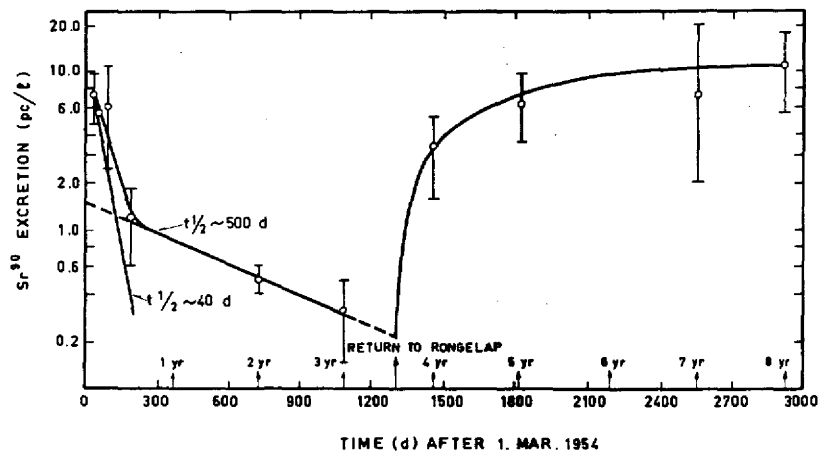


Fig. 3

Sr^{90} excretion in urine of exposed Rongelap people
The vertical bars represent estimated standard deviation.

The mean body burdens of Sr^{90} for the Rongelap population from 1954 to 1962 are shown in Fig. 4. On the return of the Marshallese to their home island, Rongelap, in June 1957, with its persistent low level of contamination, the level of Sr^{90} in the urine increased, reflecting an increased body burden. In this situation it is clear that ingestion was the primary route of entry of the contaminant. The estimated mean body burden, as obtained from the urinalysis data in March 1958 (nine months after their return), was 2 ± 1.02 nc of Sr^{90} [9]. This level checks with the values obtained from the bone samples of vertebra and ilium obtained at this time from a deceased 35-year-old adult male Marshallese. The Sr^{90} level in these bones was 3.9 pc/g calcium which, when normalized [11] to the whole

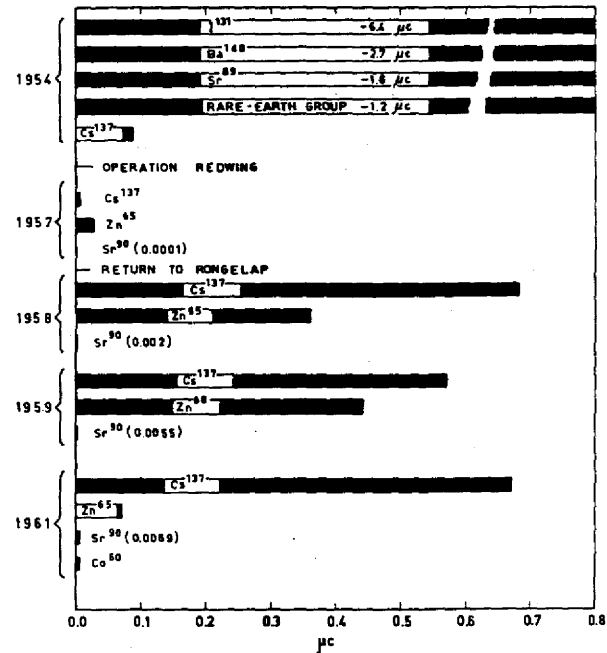


Fig. 4

Estimated body burdens of isotopes in Rongelap people, 1954-1962

skeleton, gives an average value of approximately 2 pc/g calcium or 2 nc for the body burden. This was approximately tenfold higher than the average (0.19 pc/g Ca) reported in January 1958 for adult bone in the world [12]. In March 1962 another check by direct measurement of a bone sample was possible. An autopsy samples of rib and vertebra of a 79-year-old Marshallese woman gave Sr⁹⁰ values of 13.7 ± 0.5 and 16.3 ± 0.4 pc/g. These bone values are equivalent to a skeletal body burden of 9.1 - 13.7 nc when normalized [11].

The average Sr⁹⁰ excreted by the adult group in 1962 was 11.45 ± 1.30 pc/l, or 114 ± 14 pc Sr⁹⁰/g Ca. The Sr⁹⁰ body burden estimated from these urine data was 12.0 nc, which compares favourably with the value obtained from the bone samples above. The variation in urinary excretion values of Sr⁹⁰ is quite large in the population (Table III). This variation is probably a result of analytical errors, physiological fluctuations in urinary excretion and the variation in the Sr⁹⁰ level in the individual diets. With the small sample size and the wide spread in any group, it is not possible to detect any significant difference in urinary Sr⁹⁰/Ca between the group of Marshallese exposed in 1954 and the unexposed control group. There are also wide differences in Sr⁹⁰/Ca levels as a function of age and sex. For example, in 1962 the Rongelap children (5-15 yr) had a mean Sr⁹⁰/Ca ratio in urine more than twice that of the adult group. In a world-wide study, children (5-15 yr) were found to have three times higher skeletal Sr⁹⁰/Ca ratio than adults [12]. The Sr⁹⁰/Ca ratio was found to be independent of

TABLE III
STRONTIUM-90 IN MARSHALLESE 24-h URINE
APRIL 1962

Group	Age (yr)	No. of subjects	Sr ⁹⁰ pc/l	Ca mg/l	Sr ⁹⁰ /Ca pc/g
<u>EXPOSED</u>					
Male	5-15	3	9.33±1.70*	52±14	189±18
	> 15	7	9.06±1.11	175±26	53±5
Female	5-15	2	9.60±2.30	25±11	426±96
	> 15	8	15.89±2.50	104±12	165±26
Total	5-15	5	9.44±1.18	41±11	284±66
	> 15	15	12.70±1.66	137±16	113±20
<u>NON-EXPOSED</u>					
Male	5-15	1	28.60±0	140±0	204±0
	> 15	4	8.88±2.46	129±11	66±14
Female	5-15	-	-	-	-
	> 15	7	10.24±3.07	100±49	143±28
Total	5-15	1	28.60±0	140±0	204±0
	> 15	11	9.75±2.07	110±31	115±21
<u>TOTAL</u>					
All	5-15	6	12.63±3.34	42±9	270±55
All	> 15	26	11.45±1.30	126±16	114±14

* Standard error of estimate

age after the age of 20 [12]. Rongelap females invariably had over twice as high urinary Sr⁹⁰/Ca ratio as did males, but the basis for the difference is not readily apparent.

The third method for estimating internally-deposited Sr⁹⁰ employs the Sr⁹⁰/Ca ratio of the diet and the discrimination of the body for calcium against strontium from diet to bone. From an analysis of the diets of 14 male adult Marshallese, an average daily intake of 67.5 pc Sr⁹⁰/g Ca was estimated [5]. With the presently accepted discrimination factor of four, an equilibrium value for body burden of 17 pc Sr⁹⁰/g Ca would be expected. This calculation requires a number of assumptions, for example, that the dietary intake of strontium remains essentially constant. While this figure is a rough estimate, it does agree well with the value calculated from the urinary excretion data. The daily intake of Sr⁹⁰ was estimated to be 15 pc, assuming a daily calcium intake of 1 g [9]. From the curve of Sr⁹⁰ urinary

excretion, it can be seen that the body burden does appear to be approaching an equilibrium level at a value below the 17 pc/g calcium estimated. This value also agrees with the equilibrium value of 23 nc (23 pc/g Ca) calculated from the urinary excretion data [9].

Caesium-137

The Cs^{137} urinary excretion level for the six months immediately following exposure can be expressed as a single exponential function with a biological half-life of 110 d (Fig. 5). This value is in agreement with the mean value obtained in a study of four adult males reported by RICHMOND [13].

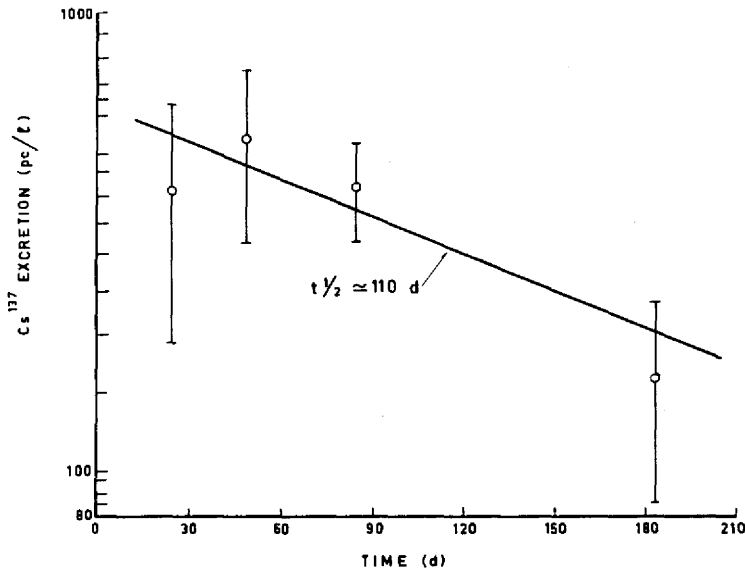


Fig. 5

Cs^{137} excretion in urine of exposed Marshallese people for 6 months following exposure. The vertical bars indicate estimated standard deviation.

The mean body burden of Cs^{137} calculated in 1957 from urine data (before the return of the Marshallese to their home island) was 7 nc, or comparable to the mean level measured in the population of the United States at that time. Four Rongelap inhabitants measured in 1957 in the whole-body counter at Argonne National Laboratory also had approximately the same mean Cs^{137} body burden, 11 nc [14]. This value for Cs^{137} at this time indicates that the residual activity from the original acute exposure in 1954 was very low three years later. Actually the original Cs^{137} level in 1954 (88 nc) decreased to essentially zero by virtue of the effective half-life of Cs^{137} being 110 d. Thus the body burden of 7-11 nc in 1957 indicates continued exposure to world-wide fall-out of Cs^{137} during the 3-yr absence of the Marshallese from Rongelap.

The mean body burden of Cs^{137} increased rapidly after the return of the Marshallese to Rongelap, reaching an equilibrium value varying between

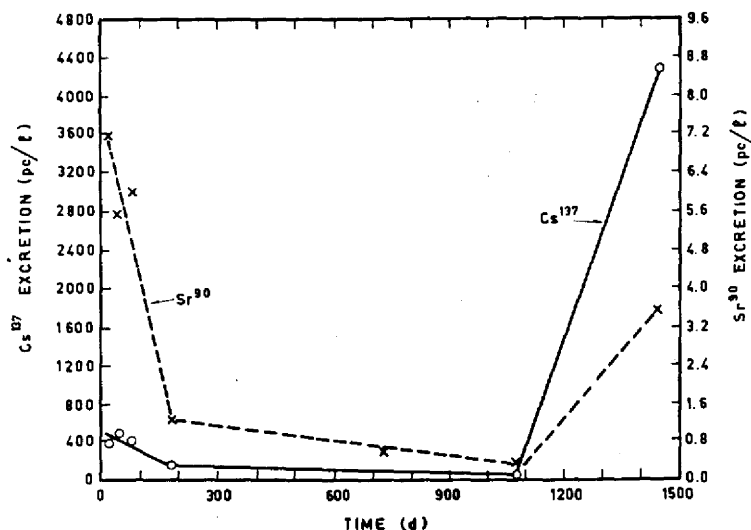


Fig. 6

Excretion levels of urinary Cs¹³⁷ and Sr⁹⁰, 1954-1958

0.58 and 0.68 μc . The rise in urinary excretion parallels that of Sr⁹⁰, but the rate is much higher (Fig. 6). The mean Cs¹³⁷ body burden remained about the same between 1958 and 1962, indicating an equilibrium with the environment (Fig. 4). These levels result from the intake of Cs¹³⁷ in the Rongelap diet as well as from world-wide fall-out. The Cs¹³⁷, with its shorter biological half-life, comes into equilibrium with the dietary level more rapidly than Sr⁹⁰. Sr⁹⁰ levels continue to approach an equilibrium in the Rongelap population. Cs¹³⁷ is thus a better indicator of recent fall-out than is Sr⁹⁰.

The mean Cs¹³⁷ body burden in 1961 for the Marshallese adult male is about 14.7 nc/kg body weight, about 300 times the mean for the medical team counted at the same time (48 pc/kg). The distribution of Cs¹³⁷ in 1961 as a function of age, sex and previous exposure is shown in Fig. 7. Although the mean values for the exposed group are slightly higher than those of the non-exposed group, the variation in each group is large and thus the mean values do not differ significantly from each other. In terms of Cs¹³⁷/kg body weight, there is no significant difference between the two age groups. Females do have a lower mean value of Cs¹³⁷/kg body weight than males.

In a study involving 6000 subjects, it was found that Cs¹³⁷/kg body weight is age-dependent as well as sex-dependent [14]. The authors report that the body burden increases up to the age of 17-22 yr, followed by a gradual fall in the ratio of Cs¹³⁷/kg over the life span. Females were found to have lower Cs¹³⁷ concentration per unit body weight than males by a factor of 1.145 [14]. In both males and females the Cs¹³⁷/g K ratio increased to the ages of 22 and 17, respectively, and then remained constant over the remaining life span [14].

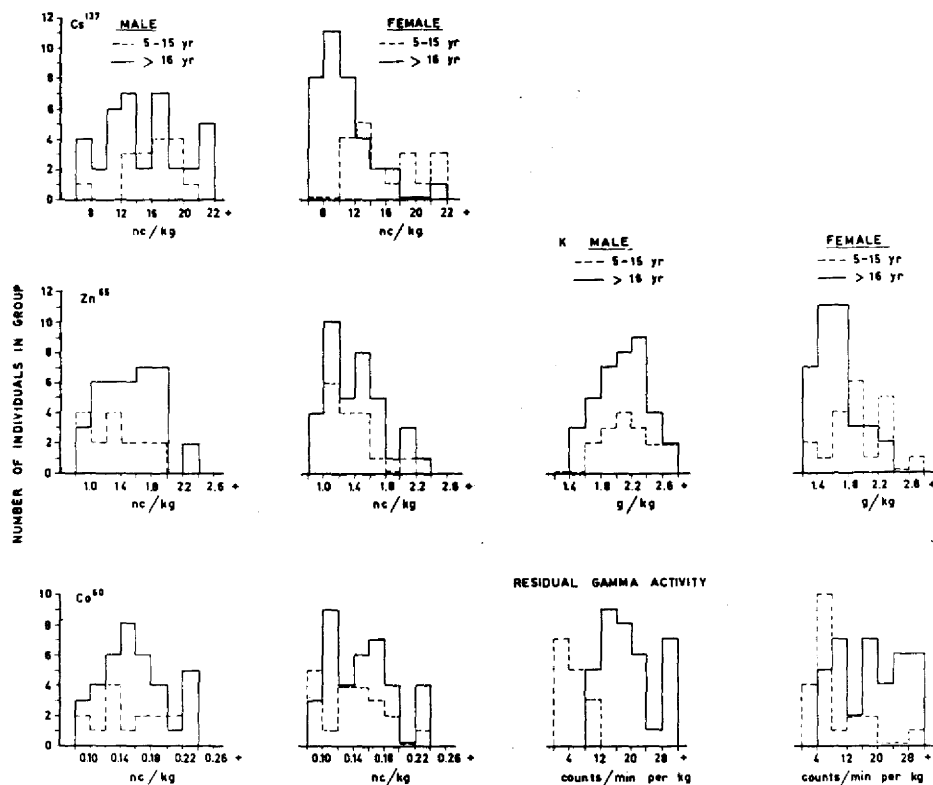


Fig. 7

Frequency distribution of levels of radionuclides in various Marshallese groups in 1961

Zinc-65

Zn^{65} was detected in high quantities in lagoon fish in 1954-55 [4]. The first measurement of the body burdens of a few people from Rongelap was made by whole-body counting in 1957 and averaged 44 nc [15]. In the 1958 medical survey, Zn^{65} body burdens increased eightfold to 360 nc, as measured by whole-body counting in a large group of people at Rongelap (Fig. 4). The Zn^{65} level increased further in 1959 and then fell precipitously in 1961 to a value of 1.5 nc/kg. The decrease in Zn^{65} level is presumably a result of the decreased Zn^{65} intake in the diet. If the Zn^{65} level in the diet were drastically reduced during the period 1959-1961, the observed fall in Zn^{65} body burden could be accounted for by the normal loss of Zn^{65} by biological and radioactive decay. The effective half-life of Zn^{65} in man is approximately 140 d [16]. Although Zn^{65} is found everywhere in the soil (10 mc/mile²), only in the vicinity of the Hanford works is it found in people in measurable amounts [17]. Zn^{65} has also been reported in groups of reactor workers [18] and cyclotron workers [19].

The Zn^{65} body burden in the Marshallese in 1961 was 1.5 nc/kg, about equal to that found in a few reactor workers at BNL [18], but considerably higher than the mean value for the medical team (15 pc/kg). This latter value is of doubtful significance, since it is not much higher than the precision of the instrument. The Zn^{65} concentration in terms of body weight in the Marshallese does not differ significantly between the exposed and the unexposed groups, nor does it appear to differ on the basis of sex or age (Fig. 7).

Potassium-40

The adult Marshallese male has an average of 2.12 g/kg body weight as compared to the mean of 1.72 for the medical team. This higher value for the Marshallese probably reflects their well-developed musculature and low body fat, as the K is proportional to the lean body mass. The K values for Marshallese females are generally lower than those for the males, 1.52 - 1.71 g/kg body weight. The K values like those for Cs^{137} are age-dependent and, as previously mentioned, the ratio of Cs^{137}/K^{40} is a constant after approximately 20 years of age in both males and females [14].

Cobalt-60

A new and interesting finding of this study was the detection of the presence of Co^{60} in the Marshallese population. The mean value is fairly uniform, about 11% of the Zn^{65} level, but the spread in values in any one group is again quite large (Fig. 7). There is a fairly good correlation between Zn^{65} and Co^{60} values. This induced activity had not been previously detected in this population because its low level was masked by the relatively large peaks of the other radionuclides in the relatively short counting times employed.

Residual γ -Activity

The residual γ -activity in the spectra remaining after the subtraction of the major components indicates that there are some residual radionuclides not present in sufficient quantity to reveal themselves via their photopeaks (Fig. 7). The adult groups had approximately the same level of residual activity, while the juveniles had less than half of the adult mean level. This difference may be the result of the error in geometry calibration of the counter for children at the low energy end of the spectra. Undoubtedly part of this residual activity in the low energy region results from bremsstrahlung from internally-deposited Sr^{90} .

D. TREATMENT

An effort was made at 52 d after the contaminating event in 1954 to increase the excretion rate of the internally-deposited fission products in seven Marshallese subjects. During a control period of 5 d, 24-h urine samples were collected and analysed to establish a mean base-line excretion

rate. During the next 3 d, Ca-EDTA was administered daily, 1 g/25 lb body weight administered orally in the form of pills. Complete urine samples were collected daily during this treatment period and for 5 d thereafter.

Although there were large fluctuations in the daily urinary excretion, the mean gross β -activity in the urine following EDTA treatment was 2.5 times the pre-treatment level. Only gross β -activity was measured; the individual radionuclides present were not identified. The oral administration of the EDTA did significantly increase the fission product excretion rate. The overall effect on decreasing the body burden in this situation, however, was small as the excretion rate was very low at this time. From previous animal studies, it is known that EDTA is most effective in chelating the rare-earth elements and not effective at all with bone-deposited alkaline earths, such as strontium. It is therefore probable that the rare-earth body burden was affected by this treatment.

SUMMARY

The medical study of the Marshallese accidentally exposed to local fall-out in 1954 is unique in that, along with the study of the Japanese fishermen, it provides the only data existing on the metabolism of mixed fission products in a human population. Early diagnosis of the internal radioactive contamination was made by radiochemical analysis of the excreta of the exposed people and by radiochemical analysis of the tissues and excreta of animals simultaneously exposed. Initially, Sr⁸⁹, Ba¹⁴⁰, I¹³¹ and its shorter-lived daughters and a number of rare-earth elements contributed the major portion of the internal radiation dose.

After a year, the principal radioisotopes were Sr⁹⁰, Cs¹³⁷ and Zn⁶⁵. Subsequently these radionuclides and, more recently, Co⁶⁰ as well, have been measured periodically. Since 1958 the γ -spectra of a number of Marshallese have been obtained with a "portable" whole-body counter.

The report discusses the findings of these studies for the past eight years. The results of an early attempt to alter the rate of removal of the mixed fission products in the Marshallese with calcium-disodium EDTA are presented. The metabolism of the radionuclides and their relationship to levels present in the environment is also discussed.

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