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Report to the Scientific Director

PROJECT 4.1 - ADDENDUM REPORT

**Nature and Extent of Internal Radioactive
Contamination of Human Beings, Plants,
and Animals Exposed to Fallout**

By

S. H. Cohn, R. W. Rinehart, J. K. Gong,
J. S. Robertson, W. L. Milne, W. H. Chapman,
and V. P. Bond

Naval Radiological Defense Laboratory
San Francisco, Calif.

Naval Medical Research Institute
Bethesda, Md.

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ABSTRACT

The first instance of exposure of human beings to mixed fission products occurred as a result of the contaminating detonation of 1 March 1954. Beta activity in the urine of these exposed human beings indicated significant internal contamination from the ingestion and inhalation of fallout material. The body burden of the group of human beings with the greatest internal contamination was of the order of the maximum permissible concentrations for the individual radionuclides.

Few of the fission products present in the environment were readily absorbed by the bloodstream from the lungs and the gastrointestinal tract. Most of those radioelements that gained entry into the body had short radiological and biological lives, and thus the levels of activity in the tissues of the body were relatively low. The concentration of radioisotopes at 6 months postdetonation was barely detectable in the urine of most of the exposed individuals.

The human body burden of individual radionuclides was estimated from radiochemical analysis of the human urine and of the tissues and urine of animals from Rongelap. The mean body burdens of the radionuclides in the Ailinginae group were approximately one-half those of the Rongelap group, and the mean body burdens of the American group were about one-fourth those of the Rongelap group.

Radioiodine is probably the most hazardous internal radioemitter at early times after exposure. Of the longer lived fission products Sr^{90} is the most abundant and presents the greatest potential hazard in this particular group.

Oral administration of calcium EDTA, beginning seven weeks postdetonation, to a representative group of individuals from Rongelap increased the rate of excretion of activity. However, the decrease of the body burden was insignificant since the excretion rate was very low at this late time.

High levels of activity were found in water and on the external surfaces of plants. The contamination of the internal portions of fruits and vegetables was small; however, high levels of contamination were found in the coconut tree sap. Of the individual radionuclides, Sr^{90} , because of its high solubility and relatively long radioactive half life, was probably the isotope of greatest potential hazard in the environment.

Gross beta activity of urine and tissue samples of animals indicates significant internal contamination of all the animals. The level of internally deposited radioisotopes in the pigs from Rongelap was ten times the amount in human beings from the same area.

Studies of animals provided direct data on the nature and distribution of the contamination in the tissues of the body. Over 90 per cent of the activity in the body of animals was localized in the skeleton. The pattern of deposition of the fission products in the skeleton seen in autoradiographs was similar to that of alkaline earths. The fish and clam had a much lower concentration of the alkaline and rare earths and a body burden considerably higher than that of the land animals. Since fish form a large staple item in the diet of the island populations, this high level of contamination is of considerable importance.

Studies made on egg production of contaminated hens gave no evidence of any radiation effect. The production rate for eggs was normal, and the eggs produced were also normal.

The extraordinary ability of fowl to mobilize calcium in shell formation resulted in the presence of very high activity in the shells of the first few eggs. The activity was associated with the fission products of the alkaline earth group.

Fertility of the hens and hatchability of the eggs produced by the mating of contaminated roosters and hens showed no radiation effects. The baby chicks hatched from these eggs are growing normally, and the amount of radioactivity in their tissues is barely detectable.

In the 6-month postdetonation period, neither significant gross changes nor pathological changes, which could definitely be ascribed to radiation, were detected in any of the animals from Rongelap.

Analysis of the internal contamination in human beings indicates that the dose to the tissues of the body was near but did not exceed the maximum permissible dose levels. The activity fixed in the body decreased rapidly as a function of time. The contribution of the effects of internal contamination to the total radiation response observed appears to be small on the basis of the estimated body burden of the radioelements. In view of the short half life of the most abundant fission products in this situation, the possibility that chronic irradiation effects will occur is quite small.

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CHAPTER 1

INTRODUCTION

1.1 OBJECTIVES

The objectives of the study of internal radioactive contamination, Project 4.1, are as follows:

1. To determine the nature and extent of the internal radiation hazard to human beings exposed to the fallout from the 1 March 1954 detonation of Operation CASTLE.
2. To evaluate the contribution of the internal contamination to the acute and long-term radiation syndrome.
3. To determine the feasibility of an internal decontamination therapy program.
4. To determine the amount and type of contamination sustained by exposed animals, food plants, soil, and water of the contaminated atolls.

The nuclear detonation of 1 March was the first instance in which a large group of people received a significant internal contamination from fission products with an accompanying external dose of less than lethal magnitude.

A detailed study of the internal contamination in the exposed human population and in animals was made to determine the kind and degree of internal deposition. There were three general problems to be investigated: (1) The first was to determine the contribution of the internal contamination to the acute radiation syndrome observed in order to evaluate the importance of internal contamination when combined external and internal irradiation occurs. (2) The second problem concerned the possibility of long-term effects. (3) The third question was directly answered by the study made, that is, what was the qualitative and quantitative nature of the internal contamination produced by exposure of individuals to mixed fission products. There has been no previous situation in which human beings have been exposed to an environment contaminated with mixed fission products.

Evaluation of the internal contamination of the human beings was made by a study of the radioelements excreted. Since very little information is presently available concerning the ratio of excreted radioelements to the amount deposited in the body, it was necessary to base the evaluation on data obtained from animals that had been contaminated in the same event. Detailed studies of animal tissues and animal excreta then provided data on which estimates of the human body burden were based.

1.2 GENERAL NATURE OF INTERNAL RADIATION TOXICITY

The nature of the radiation hazard from internally deposited fission products can best be understood in terms of the biophysical behavior of the radionuclides.

Fission products entering the body through inhalation or ingestion concentrate in various tissues and act as sources of internal radiation. The ability of a radionuclide to enter the

blood stream is determined by its solubility, chemical properties, and physical state. The radioelements formed in fission are predominantly oxides which have a limited solubility in body fluids. On this basis only a few of the radioelements can become available to the body. However, the amount that can produce injurious effects when deposited within the body is minute because of the close proximity of the isotope to the tissues it irradiates and because the isotope continues to irradiate these tissues until it is removed by biological turnover or is rendered harmless by radioactive decay. The effects of radiation from internally deposited emitters are the same as those from external radiation. The distinguishing feature of internal radiation, however, is its long continuing nature.

TABLE 1.1—Biologically Hazardous Internally Deposited Fission Products

Radioelement	Type* of radiation	Fission* abundance (%)	Half life		Fraction reaching critical organ†, ‡	
			Radiological* (days)	Biological†, ‡ (days)	By ingestion	By inhalation
Sr ⁸⁹	β	4.6	53	3.9 × 10 ³	0.25	0.22
Y ⁹¹	β	5.9	57	> 500	2.8 × 10 ⁻⁴	0.14
Zr ⁹⁵	β, γ	6.4	65	> 100	0.35	
Ru ¹⁰³	β, γ	3.7	42	20	0.04	
Ru ¹⁰⁶	β	0.5	365	20	0.04	
I ¹³¹	β, γ	2.8	8	180	0.2	0.15
Ba ¹⁴⁰	β, γ	6.0	12.8	~200	0.07	0.20
La ¹⁴⁰	β, γ	6.0	1.7	35	1.2 × 10 ⁻³	0.1
Ce ¹⁴¹	β, γ	5.7	28	> 100	0.25	
Pr ¹⁴³	β	5.4	13.8	50	1.3 × 10 ⁻³	0.063
Ce ¹⁴⁴	β, γ	5.3	275	500	2 × 10 ⁻⁴	0.10

*From G. T. Seaborg and I. Perlman, Rev. Mod. Phys., 20: 585 (1948).

†From J. G. Hamilton, Rev. Mod. Phys., 20: 718 (1948).

‡From National Bureau of Standards Handbook 52, U. S. Government Printing Office, Washington 25, D. C., 1953.

Radioactive isotopes follow the same metabolic processes in the body as the naturally occurring inactive isotopes of the same element and of chemically similar elements. Thus strontium and barium, which are analogous chemically to calcium, are deposited in the calcifying tissue of the bone. Although nearly 200 radioisotopes are produced in the fission process, only a few are potential chronic internal radiation hazards. These fission products, listed in Table 1.1, constitute a high percentage of the fission yield and localize chiefly in bone. The "bone seekers" have, in general, long radiological and biological half lives and produce high-energy beta particles. Thus they cause greater damage to bone and to the radio-sensitive bone marrow than to other tissues. The damage to the blood-forming tissue results in a reduction of blood cells and thus affects the entire body.

Information on the biological effects of internally deposited isotopes is derived from the limited studies of accidental radioisotopic poisoning in humans or from animal experimentation. The best documented data on the effects of small amounts of internally deposited emitters in human beings are obtained from studies of radium poisoning. As a result of radium deposition, terminal anemia, bone necrosis, and osteogenic sarcoma appeared after a number of years. The residual activity in the body associated with these effects was 1 to 2 μg of radium. Radium is a particularly hazardous element when deposited internally because of its very long radiological half life.

Very few data are available on the long-term biological effects in human beings of the shorter lived isotopes such as Sr⁸⁹, I¹³¹, P³², and Na²⁴. The metabolism, excretion, and bio-

logical effects of a number of fission products have been studied in animals by Hamilton,⁷ Abrams,¹ Bloom,² and others. However, most of these studies do not cover the problem of the long-term effects produced by small amounts of internally deposited isotopes in animals.

Few data are available concerning the effects of internal contamination with mixed fission products from nuclear detonations. Contamination is not produced by every detonation of a nuclear device. For example, no internal contamination was detected in any of the individuals exposed to the airbursts at Nagasaki and Hiroshima.

In field tests of the contaminating type of atomic detonation, animals that inhaled fission products during short periods of exposure were found to have insignificant amounts of internal contamination.

The long-term effects (primarily malignant changes) resulting from radium deposition have been used to set the limits for maximum permissible body concentrations of various radionuclides.¹³ The maximum permissible level of a radioisotope is the maximum amount which may be deposited in the body without the production of detectable damage. The value for both internal and external irradiation to any tissue is 0.3 rem/week.

CHAPTER 2

INTERNAL CONTAMINATION IN HUMAN BEINGS

The internal contamination study was begun 15 days postdetonation with the collection of pooled 24-hr urine samples from the Marshallese and American groups. Maximum activity in the urine occurred during the first few days after internal contamination. By one week an approximate equilibrium state was reached in which the contaminants remaining in the body were firmly fixed, chiefly in the skeletal tissues. The activity in the urine then was derived from radioelements which were replaced in the natural process of biological turnover. Thus the study made is essentially that of an equilibrium condition.

All the urine samples were sent back to laboratories in the United States for analysis since the high background encountered in the field masked the relatively low levels of activity in the aliquot samples used. A field laboratory is most desirable for a rapid survey and was shown to be feasible if adequate facilities are provided for the counting of the samples.

The first urine samples, mentioned above, were collected by the Los Alamos Scientific Laboratory (LASL). Similar samples collected 44 days postdetonation were also sent to LASL. On the 23d, 24th, and 47th days postdetonation, 24-hr urine collections from each individual in the Rongelap and Ailinginae groups were sent to the AEC New York Operations Office (NYOO-AEC) for analysis. In addition, samples from representative individuals in these groups were collected 2½, 3, and 6 months postdetonation and sent to NYOO-AEC.

The Naval Radiological Defense Laboratory (NRDL) collected samples from each member of the American, Rongelap, and Ailinginae groups at 43 to 45 days postdetonation. Samples from representatives of these groups were also collected at 2½, 3, and 6 months by NRDL. In addition, samples from a representative group of 6 Americans and 20 Marshallese were collected for 6 consecutive days beginning 33 days postdetonation.

2.1 METHODS

Since a complete radiochemical analysis of all the urine samples was not feasible, samples were analyzed only for Sr⁸⁹, Ba¹⁴⁰, the rare-earth group, and fissile material. These analyses are the most useful for evaluating the concentration and identity of all the potentially hazardous internally deposited radioactive isotopes. Measurement was also made of the gross beta activity of all the samples.

A scanning method for beta measurement, consisting of a basic oxalate precipitation with a lanthanum carrier, was employed on an aliquot of the 24-hr urine samples in order to facilitate the processing of the large number of urine samples being sent from the field. This method rapidly concentrates the radioactive elements into a small volume and eliminates the normally present K⁴⁰ background. A carbonate precipitation of the entire 24-hr sample increased the sensitivity of measurement sufficiently for analysis of samples collected later than 2½ months postdetonation.

The beta activity was counted with a thin end-window Geiger-Müller counter. The counter was calibrated with a U_2O_3 standard, and an appropriate correction for self absorption was made using a Sr^{89} standard.

2.2 RESULTS AND DISCUSSION

2.2.1 Beta Activity of the Urine

Internal deposition of radioactive elements was evidenced by the presence of significant amounts of beta activity in the urine. This activity decreased rapidly as a function of time, since it was derived chiefly from short-lived radioisotopes. For example, at 3 months postdetonation, the mean activity of the urine of the Rongelap adults was 28 per cent of the value measured 45 days postdetonation, and at 6 months the activity in the urine was barely detectable in most of the individuals.

TABLE 2.1—Summary of Human Urine Analysis, Gross Beta Activity*

Time (postdetonation)	Rongelap (age)			Ailinginae (age)			American
	<5	5-16	>16	<5	5-16	>16	
1½ months							
Case No.	7	11	31	1	2	10	25
24-hr vol. (ml)	165	439	581	150	275	722	1158
Dis/min/24 hr	404	758	1208	217	126	553	309
2½ months							
Case No.			10				
24-hr vol. (ml)			824				
Dis/min/24 hr			705				
3 months							
Case No.			10				
24-hr vol. (ml)			379				
Dis/min/24 hr			339				
6 months							
Case No.	8	12	33		3	12	
24-hr vol. (ml)	360	510	625		400	655	
Dis/min/24 hr	12	5	0		0	0	

*All values corrected for decay.

Comparison of the means of the urine samples for the adults of the Rongelap and Ailinginae groups and the American group indicates that the highest activity was in the Rongelap group at 45 days postdetonation (Table 2.1). The Ailinginae group had less than one-half the activity present in the Rongelap group, and the American group had about one-fourth the activity of the Rongelap group.

The mean gross beta activity of the urine of the Rongelap, Ailinginae, and American groups was roughly proportional to the external dose each group received. However, a comparison of the mean beta activity of the urine of the Ailinginae and the American groups indicated that the latter had a somewhat lower amount of internal contamination although both groups received about the same external dose. This may be accounted for by the fact that the Marshallese drank contaminated water from open containers and ate contaminated food up to the time of evacuation at 55 hr postdetonation. The American personnel ingested much less contaminated food and water, since both were largely stored in closed containers. Indoctri-

TABLE 2.2—Gross Beta Activity in Urine of the Rongelap Group*
(46 days postdetonation)

Age and case No.	Total 24-hr volume (ml)	Beta Activity (dis/min/24 hr)	Age and case No.	Total 24-hr volume (ml)	Beta Activity (dis/min/24 hr)
<5 yr			>16 yr		
2	120	712	4	455	634
3	150	894	7	810	1700
5	155	313	9	365	201
23	40	223	10	980	549
33	260	0	11	450	1583
54	80	385	13	340	1677
69	455	301	14	780	2460
Mean	165	404	18	455	1670
			22	47	77
6-15 yr			30	960	438
20	265	1900	34	750	570
24	550	0	37	480	792
26	650	1032	40	550	1450
35	255	0	46	330	495
36	190	236	49	425	0
39	280	1100	52	780	0
47	650	1705	55	320	1080
67	450	674	56	700	3220
72	110	507	57	550	1095
75	440	0	58	750	2170
76	380	1180	60	810	580
Mean	439	758	62	980	1985
			63	635	2260
			66	855	1715
			68	300	2010
			71	290	1450
			73	230	0
			78	965	52
			79	465	2038
			80	540	1353
			82	670	2140
			Mean	581	1208

*Values corrected for decay.

nation of the American group concerning radiation hazards probably was also a factor in reducing the amount of contamination which they received.

The variation of gross activity among the individuals in any of the three groups is quite large (Tables 2.2 and 2.3). This is chiefly the result of variations in the quantity of water and both the kind and quantity of food ingested. The degree of exposure of the individual to airborne activity is also a factor in determining the individual degree of contamination. While there were large variations among individuals, the day-to-day levels of activity for each individual were fairly consistent.

Further information on the source of individual variations was obtained by grouping the individuals from Rongelap and Ailinginae according to age (Tables 2.2, 2.3). Although the activity excreted per unit volume of urine is about the same for both children and adults, the mean activity of the urine excreted in 24 hr by children under 15 years of age was significantly lower than that excreted by adults. The available data do not indicate definitely whether the lower total excretion indicates a smaller total body burden in the children resulting from lower inhalation and ingestion or whether it represents a higher degree of fixation of the radioelements by growing bone.

TABLE 2.3—Gross Beta Activity in Urine of Ailinginae and American Groups*

Ailinginae (46 days postdetonation)			Americans (44 days postdetonation)		
Age and case No.	Total 24-hr volume (ml)	Beta Activity (dis/min/24 hr)	Case No.	Total 24-hr volume (ml)	Beta Activity (dis/min/24 hr)
<5 yr					
6			401	1970	0
8			2	650	0
44	150	217	3	1224	820
Mean	150	217	4	440	78
			5	735	0
6-15 yr			6	900	248
48	180	164	7	1340	0
53			8	1410	1260
81	370	88	9		
Mean	275	126	10		
			11	1580	385
>16 yr			12	1460	0
1	900	765	13	1810	965
16	880	827	14	720	438
28	680	1202	15	1380	830
29	780	0	16	1930	0
31	260	846	17	945	
41	920	62	18	1520	0
43	610	754	19	1300	466
45	850	680	20	1070	0
51	410	400	21	550	353
70	440	0	22		
Mean	722	553	23	1180	0
			24	1160	750
			25	1380	187
			26	510	323
			27	565	
			28	1220	0
			Mean	1158	309

*Values corrected for decay.

No correlation was found between body weight and the total activity per 24 hr excreted in the urine by members of the Rongelap group.

Gross beta activity measurements were also made by NYOO-AEC on the samples sent to them. Their results (Harley⁸) essentially corroborate the findings by NRDL, particularly the ratio of the activities among the three groups studied. The absolute values of the activity determined by NYOO-AEC, however, were lower than the NRDL values by a constant factor.

2.2.2 Radiochemical Analysis of the Urine: Estimate of Body Burden

Radiochemical analysis of the Rongelap urine samples indicates that the alkaline-earth and rare-earth groups together contributed 75 per cent of the beta activity at 45 days postdetonation (Table 2.4). The predominant radionuclide is Sr⁸⁹, which contributes 42 per cent of the total beta activity at this time.

Assays of fissile material made on pooled samples of urine were all negative within experimental limits.

The early urine samples analyzed by the LASL⁹ (collected 15 days postdetonation) contained large amounts of radioiodine in addition to the alkaline and rare earths.

On the basis of the radiochemical analysis of the urine, the body burden (the radioisotopic deposition in the tissues) was estimated. The ratio between the activity of the urine and the amount of isotope fixed in the body is required for this calculation. However, few ratios are available for the deposition of the various radioelements in humans, so that it was necessary to utilize ratios obtained from animal studies. Of the animals collected from the islands, the pig was selected as the closest to the human in size and metabolism. A detailed study was therefore made on the excretion of these animals and on the radioactive content of various tissues. Details of the animal study are presented in Chap. 3.

TABLE 2.4—Radiochemical Analysis of Rongelap Urine
(45 days postdetonation)

Sample No.	Beta Activity (dis/min/24 hr)			
	Gross beta activity	Sr ⁸⁹	Ba ¹⁴⁰	Rare-earth activity
1	1370	490	120	197
2	1260	510	130	244
3	1020	480	120	324
4	1210	626	150	284
5	1460	328	110	474
6	1200	727	170	353
Av.	1253	526	134	312
Total beta activity, %	100	42	10.7	25.5

TABLE 2.5—Mean Body Burden of Rongelap Group

Radioisotope	Activity at 82 days (μ c) (NRDL)	Activity at 1 day (μ c) (NRDL)	Activity at 1 day (μ c) (LASL)
Sr ⁸⁹	0.19	1.6	2.2
Ba ¹⁴⁰	0.021	2.7	0.34
Rare-earth group	0.03	1.2	
I ¹³¹ (in thyroid)	0	6.4	11.2
Ru ¹⁰³			0.013
Ca ⁴⁵	0	0	0.019
Fissile material	0	0	0.016 (μ g)

The estimate of the mean body burden of the Rongelap group at 82 days postdetonation is presented in Table 2.5. The body burden at 1 day postdetonation was calculated in the following manner. A formula was obtained from urinary excretion data (reported by Cowan, Farabee, and Love⁵) in a case of accidental inhalation of Sr⁹⁰. The excretion curve was best represented by four exponentials. [Very similar results were obtained by approximating the biological decay of strontium with a power function, based on human excretion of the metabolically similar element, radium (Norris, Speckman, and Gustafson¹⁴ and Looney¹¹).]

Estimates were made of other radioelements present in significant amounts at 1 day, as shown in Table 2.5. These estimates were made on the basis of the level of Sr⁸⁹ at 1 day

together with data on the activity of the various fission products at this same time¹⁰ and animal isotope absorption and retention data.^{7,13}

LASL has also estimated the body burden at 1 day on the basis of radiochemical analysis of pooled urine samples from a representative number of Rongelap and American individuals (Harris⁹). These calculations were based on the analysis of I¹³¹ in the early samples of urine (15 days postdetonation), as well as the above-mentioned physical and biological data on fission products (Hamilton,⁷ National Bureau of Standards Handbook 52,¹³ and Hunter and Ballou¹⁰). Their findings are presented in Table 2.5.

On the basis of an assumed uptake of 20 per cent per 24 hr, the integrated dose to the thyroid from I¹³¹ and other shorter lived iodine isotopes was calculated by NRDL to be about 100 rep. LASL has estimated (Harris⁹) that this dose was about 150 rep for the Rongelap group and 50 rep for the Americans.

The differing approaches used by NRDL and LASL for estimating the body burden gave results which, except for Ba¹⁴⁰, are very close.

The mean body burdens of the individual nuclides presented in Table 2.5 were calculated for the Rongelap group. Values for the Ailinginae group were approximately one-half those of the Rongelap group, and values for the Americans were about one-fourth those of the Rongelap group.

The total amount of radioactive material present in the gastrointestinal tract at 1 day postdetonation in members of the Rongelap group was estimated as approximately 3 mc. This activity was contributed chiefly by isotopes of short radiological and biological half life and limited solubility. Thus the levels of activity in the tissues of the body were relatively low. The concentration of radioisotopes at 6 months postdetonation was barely detectable in the urine of most exposed individuals.

Iodine, which is quite soluble, is probably the most hazardous internal radioemitter in the early period following exposure (Harris⁹). The dose to the thyroid was appreciable, but it was low compared to the partially or totally ablating doses of I¹³¹ used in therapy of hyperthyroidism or carcinoma. At 1 day postdetonation Sr⁸⁹ was calculated to be near the maximum permissible level¹³ for this nuclide. At later times following exposure of this group, this longer lived fission product presents the greatest potential internal hazard.

The present study confirms the observation made in animal experiments, that most of the radioactive elements formed in fission as well as the fissile material itself, are not readily absorbed by the bloodstream from the lungs and the gastrointestinal tract. Only iodine, strontium, barium, and a few of the rare-earth elements were absorbed to any significant degree.

An attempt to measure bone-fixed radioactive emitters by means of sensitive film badges taped below the knee, over the epiphysis of the tibia, on a number of persons yielded no positive results.

No correlation could be obtained between the degree of internal contamination and the clinical and hematological findings. In view of the short half life of the most abundant fission products deposited internally in this situation, the possibility that chronic irradiation effects will occur is quite small. Thus, an evaluation of the data on the internal contamination, including that of Sr⁸⁹, leads to the conclusion that the internal hazard to the contaminated inhabitants of the Marshall Islands is minimal both from the acute and the long-range point of view.

2.3 ROUTE OF ENTRY OF INTERNAL CONTAMINATION

Internal deposition of fission products resulted from inhalation and ingestion of the fallout material. Ingestion appears to be the more important of the two routes of entry into the body. The activity in the air settles out fairly rapidly, but contaminated food, water, and utensils retain their activity for long periods of time.

The particles with which the activity was associated were considerably larger than the optimum size for deposition in the alveolar tissue of the lung (see Chap. 4). Thus the probability of the retention of inhaled airborne contamination was not appreciable during the exposure period.

The hypothesis that ingestion was the chief source of internal contamination is supported by the finding that the gastrointestinal tract, its contents, and the liver of autopsied chickens and pigs sacrificed at early intervals following detonation were more active than the alveolar tissue.

The importance of ingestion as a continuing source of contamination is evidenced by the level of internal contamination of the Rongelap pigs. These animals had about ten times the body burden of the human population. Since the airborne activity had already dropped to a low value at the time of evacuation of the humans, the contamination of the pigs during their prolonged stay on the island necessarily derived from ingestion of radioactive food and water.

2.4 INTERNAL RADIOACTIVE DECONTAMINATION THERAPY

Since there is no method of counteracting the effects of radiation from internally deposited emitters, treatment consists of removing the nuclides from the body as rapidly as possible.

TABLE 2.6—Internal Radioactive Decontamination Calcium EDTA
Treatment of Rongelap Group
[Beta Activity (dis/min/24 hr)*]

Patient No.	7	14	39	52	63	66	71	Daily av.
Pretreatment (Day 35-41)	1750	810	612	2190	2291	3610	3255	2074
Post-treatment (Day 55-59)	6206	3922	5885	3583	6356	4284	3190	4775

*All values corrected for decay to day 35.

The ability of ethylene-diamine-tetra-acetic acid (EDTA) to mobilize certain of the fission products from the skeleton and to increase the rate of their excretion has previously been demonstrated (Foreman and Hamilton,⁶ Cohn et al.³). It is most effective with the rare-earth group, but it has no effect on strontium (Cohn and Gong⁴). These studies have shown that most of the biologically hazardous material remaining in the body is firmly fixed in bone within a short time, so that effective systemic decontamination by chemical agents can occur only in a short period following exposure. Nevertheless, an attempt to effect internal decontamination was made at 7 weeks postdetonation, since it was felt that whereas optimum decontamination could not be accomplished at this time, any procedure which enhanced the elimination of some of the radioelements from the body was valuable in reducing the ultimate hazard to the contaminated individual.

A representative group of seven individuals from Rongelap were selected for this study. During a control period of 5 days, 24-hr urine samples were collected daily for radioanalysis in order to establish a basal excretion rate. During the next 3 days, calcium EDTA was administered daily (1 g per 25 lb of body weight) by oral administration. (Although the slow-drip intravenous administration of sodium EDTA in 5 per cent dextrose is the method of choice, only the oral administration of calcium EDTA was feasible in this situation.)

Twenty-four-hour urine samples were collected daily during the treatment period and for 5 days following treatment to determine the effectiveness of EDTA in accelerating the excretion rate of the radioelements.

No side effects from the use of EDTA were observed. Blood counts and blood pressure remained unchanged throughout the treatment.

The mean activity of the urine during the EDTA treatment period was 2.5 times the pretreatment activity (Table 2.6). The probability that the differences observed are due to chance is less than 0.01. Thus the oral administration of EDTA for a period of 3 days beginning 52

days postdetonation increased the excretion rate of internally deposited fission products, but the over-all effect on decreasing the body burden was insignificant, as the excretion rates were very low at this time.

2.5 SUMMARY AND CONCLUSIONS

The first instance of internal deposition of mixed fission products in humans occurred as a result of the contaminating event of 1 March. This internal contamination resulted from both inhalation and ingestion of contaminated material, the latter being the more important.

Few of the fission products present in the environment were readily absorbed from the lungs and the gastrointestinal tract. Radiochemical analysis of the urine samples from the Rongelap group indicates that strontium, barium, and the rare-earth group together constituted 75 per cent of the total beta activity of the urine at 45 days postdetonation. Strontium-89 was the predominant radionuclide at this time, contributing 42 per cent of the total beta activity. Assays for fissile material in the pooled urine samples were all negative.

The human body burden of individual radionuclides was estimated from radiochemical analysis of the human urine and of the tissues and urine of animals from Rongelap. The mean body burdens of the radionuclides in the Ailinginae group were approximately one-half those of the Rongelap group, and the mean body burdens of the American group were about one-fourth those of the Rongelap group. While the activity excreted per unit volume of urine was the same for adults and children of the Rongelap group, the total activity excreted in the urine in 24 hr by children under 15 years of age was significantly lower than that excreted by the adults.

The total amount of radioactive material in the gastrointestinal tract at 1 day postdetonation was estimated to be 3 mc in members of the Rongelap group. This 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 relatively low. The concentration of radioisotopes at 6 months postdetonation was barely detectable in the urine of most of the exposed individuals.

The estimated dose to the thyroid from I^{131} and other short-lived iodine isotopes was 100 to 150 rep for the Rongelap group. Iodine is probably the most hazardous internal radioemitter at early times after exposure. The dose to the thyroid, while appreciable, was low compared to the partially or totally ablating doses of I^{131} used in the treatment of hyperthyroidism or carcinoma.

At 1 day postdetonation, the concentration of Sr^{89} was calculated to be near the maximum permissible level for this nuclide. At later times following exposure, this longer lived fission product presents the greatest potential internal hazard.

Oral administration of calcium EDTA beginning 7 weeks postdetonation to a representative group of individuals from Rongelap increased the rate of excretion of activity 2.5 times. However, the decrease of the body burden was insignificant, as the excretion rate was very low at this time.

Analysis of the internal contamination indicates that the dose to the tissues of the body was near but did not exceed the maximum permissible dose levels. The activity fixed in the body decreased rapidly as a function of time. The contribution of the effects of internal contamination to the total radiation response observed appears to be small on the basis of the estimated body burden of the radioelements. In view of the short half life of the most abundant fission products in the situation, the possibility that chronic irradiation effects will occur is quite small.

CHAPTER 3

INTERNAL CONTAMINATION IN ANIMALS

The internal contamination of a number of animals collected on the Marshall Islands was studied. The activity in their urinary excretion was studied, and radiochemical analyses were made of various tissues. These data provided the bases for estimating the body burden of the radioisotopes in human beings. In addition, hematological and pathological studies were made, and autoradiographs of selected tissues were prepared. A number of the animals are also being studied for the appearance of possible long-term effects of radiation.

A special study was carried out to determine the effect of the radiation on the fertility of chickens and the hatchability of their eggs.

The animals collected on Rongelap and Utirik Islands included 41 chickens, 9 baby chicks, 11 swine, 4 ducks, and 1 cat. These were all shipped alive to NRDL. Three fish and one large clam were taken from the Rongelap lagoon. Collection dates and mortality data for these animals are presented in Table 3.1. In addition, a boar, a cat, and two chickens were autopsied in the field, and representative tissues were collected.

3.1 METHODS

Tissue samples were taken from all animals that died spontaneously or were sacrificed. Specimens were obtained from the lung, liver, gastrointestinal tract, and the skeleton. The samples were ashed at 550°C in a muffle oven, and the ash made up to volume with 2N HCl. An aliquot was then dried for beta measurement. The beta activity was determined by means of a thin end-window Geiger-Müller counter. Strontium-89 was used as the basis for the mass absorption correction for the samples, since it was the major radioelement deposited. The correction calculated is an approximation, since mass absorption is a function of the average energy of the sample. Beta activity was measured in total disintegrations per minute, and this value was converted to microcuries, "Sr⁸⁹ equivalent."

The gamma activity of the tissue samples was measured in a well-type sodium iodide scintillation counter, which has an efficiency of about 40 per cent for a Co⁶⁰ standard. The gamma activity was obtained in total disintegrations per minute, and this value was converted to microcuries, "Co⁶⁰ equivalent."

Samples were analyzed radiochemically for Sr⁸⁹, Ba¹⁴⁰, the rare-earth group, I¹³¹, and fissile material.

For excretion studies the animals were caged individually, and their excreta were collected at 24-hr intervals. The feces and urine were ashed together for the chickens, whereas they were collected separately for the pigs. Beginning 5 weeks postdetonation, the excreta of a representative group of chickens were collected at weekly intervals for a period of 2½ months. Collection of pig excreta was begun at 6 weeks postdetonation, and the collection was made at weekly inter-

TABLE 3.1 — Mortality and External Radiation Dose of Animals from the Living Areas of Rongelap and Utirik

Series	A			B			C			D			Total		
	280 r (day 8)			330 r (day 25)			340 r (day 33)			360 r (day 51 to 53)			Total received	Dead	Sacrificed
Animals	Total received	Dead	Sacrificed	Total received	Dead	Sacrificed	Total received	Dead	Sacrificed	Total received	Dead	Sacrificed			
Hens	6	1 (day 23)	1 (day 23)				20	2 (days 42 and 43)	2 (day 44)	11	5†		37	8	3
Roosters	1						2	1 (day 49)		1			4	1	
Chicks							9	9					9	9	
Ducks							4		1 (day 56)				4		1
Pigs	1		1 (day 45)	7		4‡				3§			11		5
Cat	1												1		
													66	18	9

* Day (postdetonation) of collection is given in parentheses.

† Day 67, No. 36; day 74, No. 39; day 92, No. 35; day 99, No. 7; day 130, No. 24.

‡ Day 38, sow; day 57, No. 6; day 82, Nos. 24 and 25.

§ Animals from Utirik; all others from Rongelap (Utirik animals received 32 r external dose).

vals for a 6-week period. Radioanalysis of the excreta was performed in the same manner as that of the tissue samples described above.

3.2 RESULTS AND DISCUSSION

3.2.1 Gross Observations

The animals had been roaming free on the islands. Although malnourished, they showed no evidence of disease. Autopsy of two chickens that died during shipment revealed no pathological findings that could be associated with radiation.

On the basis of an assumed 1-hr effective fallout time, the Rongelap animals received an integrated external dose of 280 to 360 r, depending on the date of their collection (see Table 3.1). The Utirik pigs had received a calculated dose of 32 r at the time of their evacuation. The animals all showed extensive external contamination, ranging from 0.5 to 5 mr/hr at 30 days postdetonation. This activity was reduced about 75 per cent by a washing with water alone.

3.2.2 Radioactivity of Tissues and Excreta

The gross beta activity of the pig at 82 days postdetonation was about 4 μ c. The distribution of activity in the individual tissues is shown in Table 3.2. Over 90 per cent of the beta activity was localized in the skeleton. The highest activity in a soft tissue was found in the liver, which had, however, less than 0.5 per cent of the total body burden. The colon contents had the second highest activity for the soft tissues, about 0.24 per cent of the total. The alveolar tissue of the lung had an activity less than 0.02 per cent of the total activity in the body.

Gross beta and gamma activity of the chickens at 74 days postdetonation was approximately 0.2 μ c. The gross activity per body weight of the chicken is approximately the same as that of the pig. The distribution of activity in the tissues of the chicken (Table 3.3) was very similar to that in the pig. Most respiratory activity was localized in the turbinates, as a result of entrapment of the large particles that could not penetrate to the alveolar tissue.

The beta activity in the skeleton of chickens at 160 days dropped to 4 per cent of the value at 24 days postdetonation, whereas in the same period the gamma activity dropped to 0.2 per cent of the 24-day value. These data indicate that most of the activity is associated with short-lived isotopes. The initial drop in activity is very rapid, and after 45 days the decay curve is essentially that of Sr^{89} , the most abundant of the longer lived elements deposited.

The residual total beta activity found in the two larger fish at 4 months postdetonation averaged 2.5 μ c (Table 3.4). There was, at the same time, about twice as much gamma activity. The fish were collected 56 days postdetonation, and the drop in activity between that time and the analysis at 4 months represents only radiological decay. Thus the results are not directly comparable to those obtained from animals that were returned alive and in which biological turnover, as well as radiological decay, was operating.

The largest fraction of the gross beta activity in the fish was contributed by the concentration of radioactive material in the viscera. In two of the fish in which bones and muscle were separated and analyzed, equal amounts of activity were found in each fraction. However, the storage of these fish in formaldehyde for 3 months may have permitted the diffusion of the radioelements from bone to muscle to take place. Further studies on fresh fish will clarify this point.

The contamination of the fish was considerably greater than that of the land animals studied. Since fish form a large staple item in the diet of the island populations, the high level of contamination is very important to consider.

At the end of a 2½-month experimental period, the excretion by the chickens of beta and gamma activity per 24 hr was 5 per cent of the value measured at the start at 37 days postdetonation (Fig. 3.1).

Analysis of pig excreta indicates a similar decrease of activity with time. In a 6-week period the gamma activity excreted per 24 hr decreased to about 2.5 per cent of the activity excreted at 44 days postdetonation (Fig. 3.1).

TABLE 3.2—Radiochemical Analysis of Tissues and Urine of Pigs from Rongelap*
(82 days postdetonation)

Sample	Beta activity (dis/min/total sample)			
	Gross activity ($\times 10^{-3}$)	Sr ⁸⁹ ($\times 10^{-3}$)	Ba ¹⁴⁰ ($\times 10^{-3}$)	Total rare earth ($\times 10^{-3}$)
Pig No. 24 (25.8 kg)				
Skeleton (total)	8890	5660	660	1010
Liver	31	0.40	0.33	6.4
Colon and contents	12	5.0	2.4	3.2
Lung (alveolar)	1.5	0.22	0.20	0.8
Stomach	1.2	0.22	1.1	1.3
Intestine (small)	2.3	0.62	0.50	0.51
Kidney	3.3	0.21	0.42	0.74
Remaining tissues	690			
Total	9630	5667	665	1020
Urine sample, 24 hr	13	8.7	1.2	1.6
Pig No. 25 (22.7 kg)				
Skeleton (total)	8600	5100	530	690
Liver	27	0.53	0.20	5.5
Colon and contents	16	5.0	3.2	4.9
Lung (alveolar)	1.1	0.26	0.23	0.33
Stomach	2.0	0.29	0.13	0.30
Intestine (small)	2.6	0.83	0.88	0.88
Kidney	3.1	0.14	0.19	0.52
Remaining tissues	220			
Total	8870	5107	534	702
Urine sample, 24 hr	6.2	4.4	0.40	0.54
Summary				
Gross beta activity	Skeleton	Total body	Urine (24 hr)	
Sr 89	62.0	58.0	69.0	
Ba 140	6.8	6.5	7.9	
Rare earth	9.7	9.0	10.5	
Total	78.5	73.5	87.4	

* All values corrected for decay.

The excreta of the Utirik pigs contained less than 10 per cent of the gross beta activity found in the excreta of the Rongelap pigs at the same time. This ratio of 10 was approximately the same ratio found between the activity of the Rongelap and Utirik food, water, and soil samples.

3.2.3 Radiochemical Analysis of Tissues and Excreta

Radiochemical analysis of pig tissues indicated that 62 per cent of the skeletal beta activity was derived from Sr⁸⁹, 7 per cent from Ba¹⁴⁰, and 10 per cent from the rare-earth group at 82 days postdetonation (Table 3.2). The radioisotopic composition of the urine at this time was very similar to that of the skeleton. This distribution of activity in the body of the pig probably represents the distribution in human beings. The absolute amount of human internal contamination in the Rongelap group was, however, only $\frac{1}{10}$ of that found in the animals.

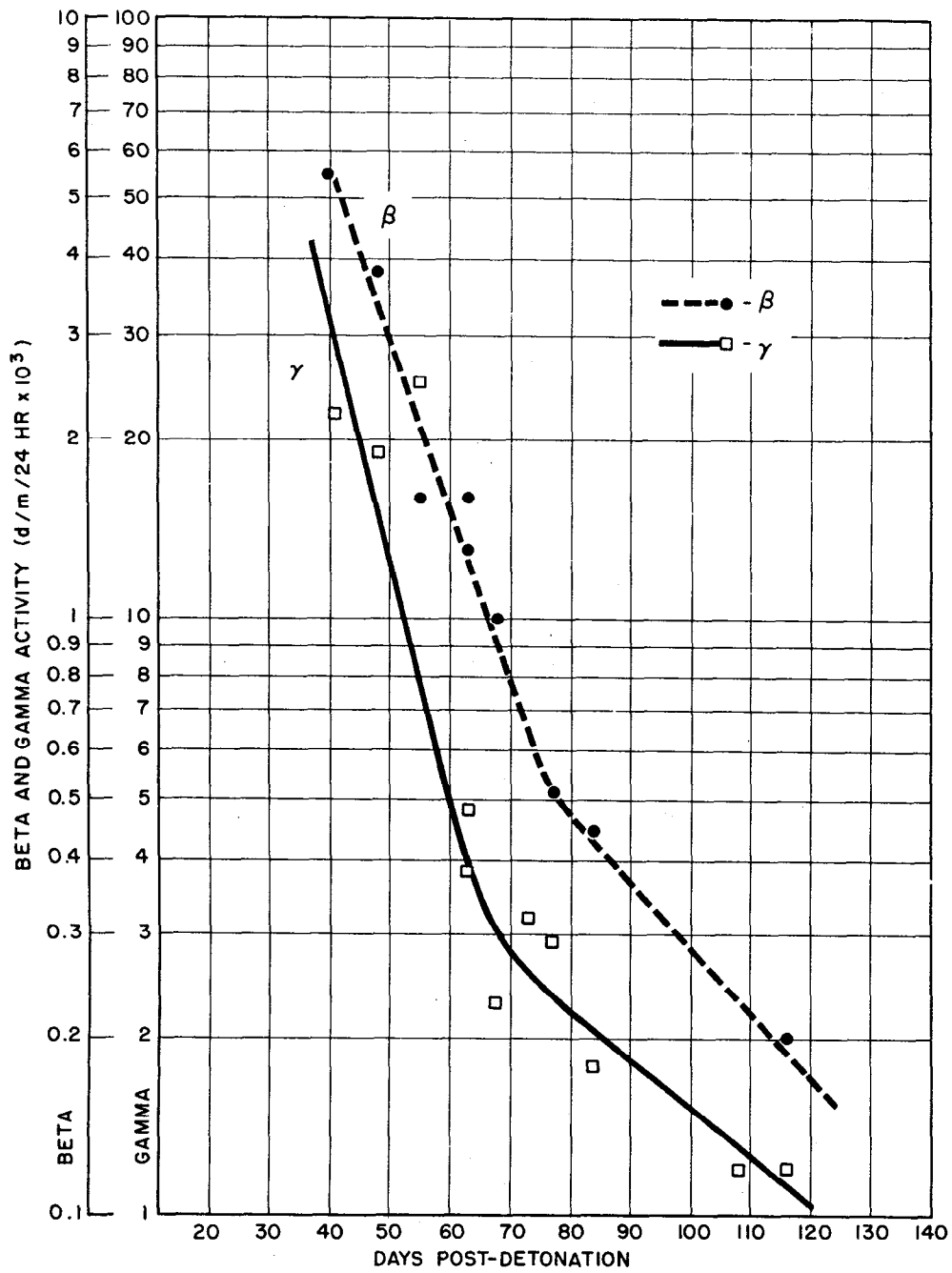


Fig. 3.1—Beta and Gamma Activity in Chicken Excreta.

TABLE 3.3—Beta and Gamma Activity of Chickens from Rongelap ($\mu\text{c} \times 10^4$)

(Beta, Sr^{89} equivalent; Gamma, Co^{60} equivalent.)

	Hen No. 1		Hen No. 2		Hen No. 39		Hen No. 36		Hen No. 35		Hen No. 7		Hen No. 24	
Day of death*	Day 23		Day 23		Day 74		Day 97		Day 121		Day 138		Day 159	
Day analyzed*	Day 24		Day 24		Day 79		Day 107		Day 122		Day 140		Day 159	
Tissue	Activity		Activity		Activity		Activity		Activity		Activity		Activity	
	Beta	Gamma	Beta	Gamma	Beta	Gamma	Beta	Gamma	Beta	Gamma	Beta	Gamma	Beta	Gamma
Tibia	7600	3850	8180	4610	133	695	253		215.5	59	41.3	31.3	33.2	8.1
Skeleton	11030	55800	11900	66900	1930	8600	3670†		3120	850†	600	454†	437	117.5†
Liver	119	21	352	271	12	72	34		32	33	17.7	13.5	10.7	1.8
Gizzard					4.1	17	7.0		8.5	7.6	10.3	7.9	3.6	0.6
Gizzard (content)					0.93				1.4		7.5	1.2	0	0.3
Crop					0.43	5.0	2.0		7.9		12.2	9.3	4.5	0
Intestine (large) and contents					0.63	10.0	3.0		6.3		14.0	10.7	8.9	0.29
Intestine (small) and contents					1.6	4.0	3.0				8.4	6.4		
Pancreas					0.16									
Spleen								1.0						0.75
Kidney	198	46			1.17	9.0	9.0		14.2	10.0	14.9	12.4	0.26	0
Lungs (alveoli)	17	28	0	26	0.57	4.0	2.0		1.4	4.5	5.6	4.3	0.79	0.23
Trachea					0.24	2.0	1.0		10.7	3.7	0.9	0.2	16.8	0.83
Turbinates					3.87	19	22		15.3	7.6				

* Day postdetonation.

† Calculated using ratio of gamma activity skeleton/tibia.

TABLE 3.4—Beta and Gamma Activity of Fish from Rongelap Lagoon*
(3 months postdetonation)

Fish No. 1 (802 g)						
	Gross activity (μc)			Radiochemical analysis in barium, strontium, and rare-earth fraction (%)		
	Beta	Gamma	Barium, strontium, and rare earth; Total activity (%)	Sr ⁸⁹	Ba ¹⁴⁰	Rare earth
Head	0.568	1.26	9.9	38.3	9.6	52.1
Scales, fins and tail	0.500	0.58	9.5	17.4	9.9	72.7
Viscera	0.900	2.36	48.0	1.4	0.6	98.0
Gills	0.160	0.43	7.8	13.9	6.7	79.4
Remainder of body	0.596	1.78	8.3	45.2	11.2	43.6
Total	2.724	6.41				
Fish No. 2 (507 g)			Fish No. 3 (168 g)			
	Gross activity (μc)		Gross activity (μc)			
	Beta	Gamma	Beta	Gamma	Beta	Gamma
Head	0.101	0.23	0.045	0.017		
Scales, fins, and tail	0.067	0.23	0.058	0.084		
Viscera	1.620	2.14	0.115	0.205		
Gills	0.043	0.09	0.023	0.011		
Skeleton	0.197	0.35	0.030	0.070		
Muscle	0.151	0.53	0.038	0.074		
Total	2.179	3.58	0.301	0.461		
Clam No. 1						
Total beta activity— 6.4×10^6 dis/min						
Radiochemical analysis						
Radioelement			Total activity (%)			
Zr ⁹⁵			21.4			
Ru ^{103,106}			32.4			
Other			11.4			
Sr ⁸⁹			0.7			
Ba ¹⁴⁰			0.7			
Rare earths			33.4			

* Samples collected two months postdetonation.

At 4 months postdetonation the alkaline earths comprised less than 2 per cent of the total activity in the clam (Table 3.4). The rare-earth group constituted 33 per cent of the total beta activity. The balance of the activity was contributed chiefly by Zr⁹⁵ (21 per cent) and Ru^{103,106} (32 per cent). About 50 per cent of the material found in the viscera of the fish was of the rare-earth group. Very small amounts of strontium and barium were found. In the tissues of the fish, strontium, barium, and the rare earths contributed only about 10 per cent of the total activity.

3.3 AUTORADIOGRAPHS

A number of autoradiographs of the tibiae and femurs of 1 chick, 4 pigs, 1 rooster, and 2 chickens were prepared both at NRDL and at the Argonne National Laboratory (ANL) to deter-

mine the pattern of deposition of fission products. Contact printing on X-ray no-screen film was found to be the most satisfactory method of preparing the autoradiographs. The discussion and conclusions presented below summarize the findings reported by Norris et al.¹⁵

The autoradiograph of a tibia from a chicken sacrificed at 45 days postdetonation (Fig. 3.2) indicated a relatively uniform distribution of the activity throughout most of the bone, with the highest concentration of activity in the area adjacent to the epiphysis. This area of high activity corresponds to an area of dense trabecular bone.

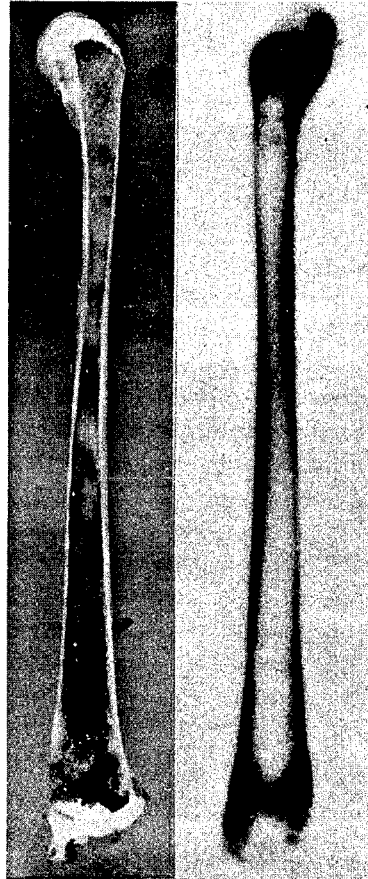


Fig. 3.2—Autoradiograph of Tibia of Chicken Sacrificed 45 Days Postdetonation (ANL).

The tibia and femur of a baby chick, which died spontaneously 47 days postdetonation, showed the heaviest concentration of radioactive material in the diaphysis (Fig. 3.3). The end regions of the bone, which were laid down after the animals were removed from the contaminated environment, were relatively lacking in activity. The region of greatest activity was in the diaphysis, which appeared to be abnormally constricted, possibly because of a decreased rate of endosteal resorption.

The tibiae from pig 1 sacrificed 45 days postdetonation and from pig 6 sacrificed 58 days postdetonation each have an area under the growing epiphysis free of activity (Figs. 3.4 and 3.5). As in the chick described above, this area corresponds to the growth that took place after the animals were removed from the area of contamination. The marrow cavity in these tibiae contained dense trabecular bone along their entire length, a formation not normally found in mammalian bones. The centers of the diaphysis were abnormally thick, possibly because of a failure of the normal resorptive process. In pig 1 there were also two distinct areas of increased density in the trabecular region, which appear as two lines of radioactivity in the autoradiograph.

The pattern of radioactive deposit in the bones of two adult animals is illustrated in a sow sacrificed 38 days postexposure (Fig. 3.6) and in a boar sacrificed 26 days postdetonation (Fig. 3.7). Faint deposits of activity in the trabecular bone were noted, separate from the higher level in the epiphysis, which is characteristic of uptake of the alkaline earths by adult bone.

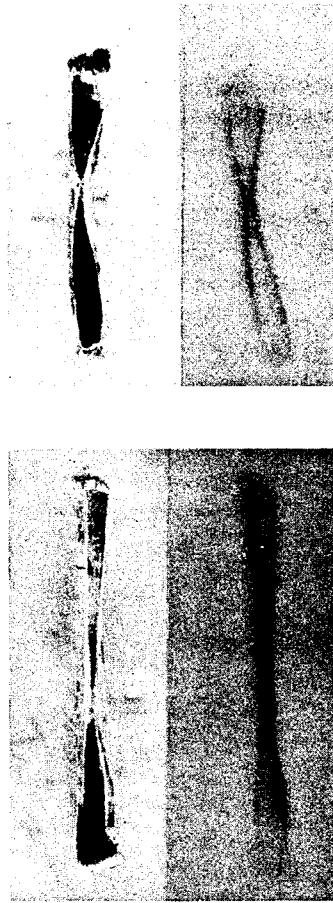


Fig. 3.3—Autoradiograph of Tibia and Femur of Baby Chick Sacrificed 46 Days Postdetonation (ANL).

Bloom² has shown that atypical osseous tissue in trabecular space is a characteristic histopathological finding following radioactive deposition. For example, clinical studies have shown that following radium deposition in bone, atypical osseous tissue is formed in cancellous bone. These formations appear as areas of increased density in roentgenograms (Looney¹¹).

It is difficult to interpret the anomaly in the pig, described above, and the dense trabecular bone in both the pig and chicken. No normal controls are available for comparison with these animals, and the history of the animals from the time of exposure to the time of collection is not known. Severe dietary changes and disease also produce changes in the pattern of deposition of osseous tissue, and such changes are often indistinguishable from changes produced by exposure to radiation.

3.4 PATHOLOGY

Sections of lung, liver, and tibia, as well as thyroid and other endocrine organs of most of the fowl and pigs dying spontaneously or sacrificed, were examined by Reed at NRDL. A few pathological changes were found, including an aplastic marrow in one of the bones of a duck.

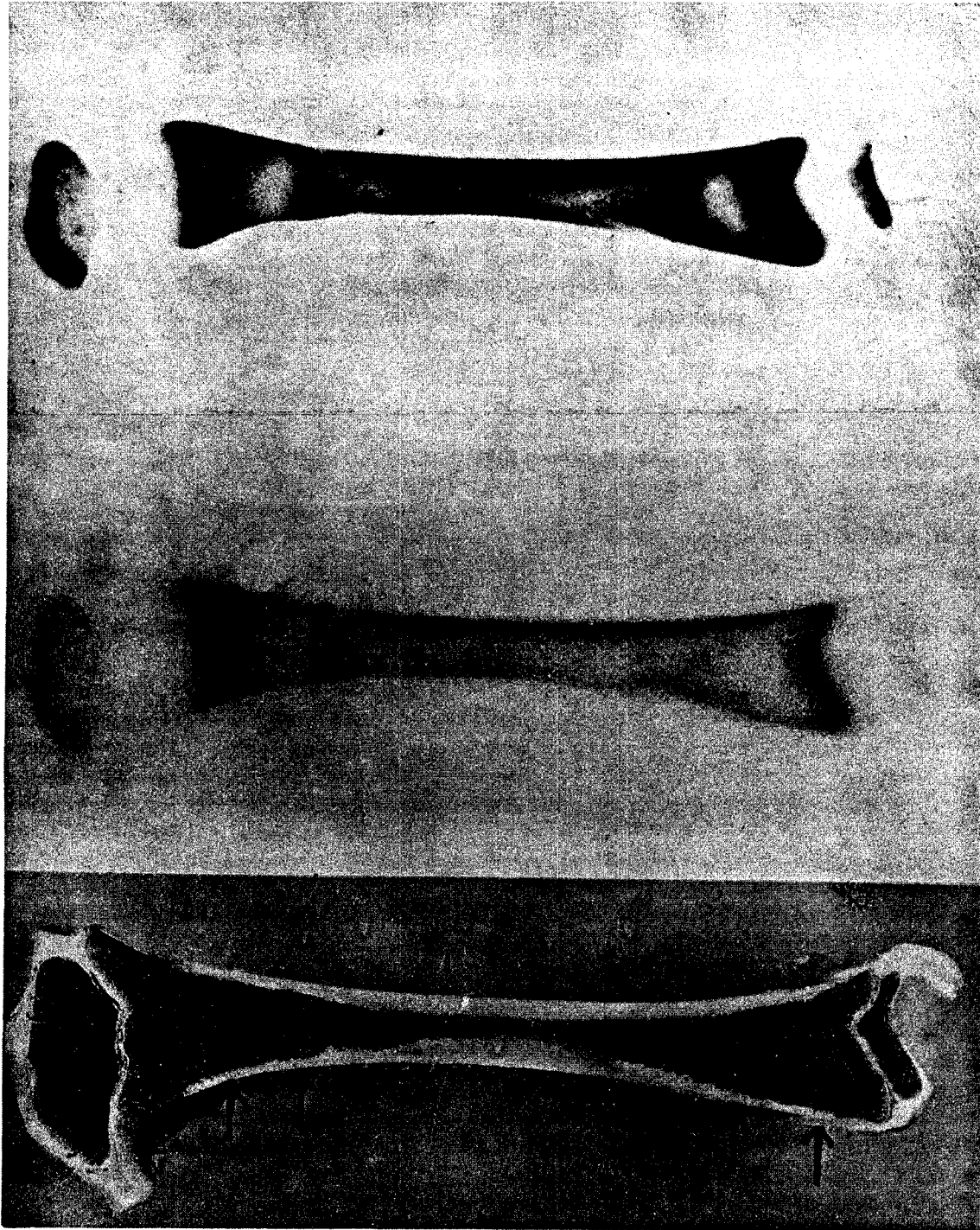


Fig. 3.4—Autoradiograph of Tibia of Pig Sacrificed 45 Days Postdetonation (ANL).

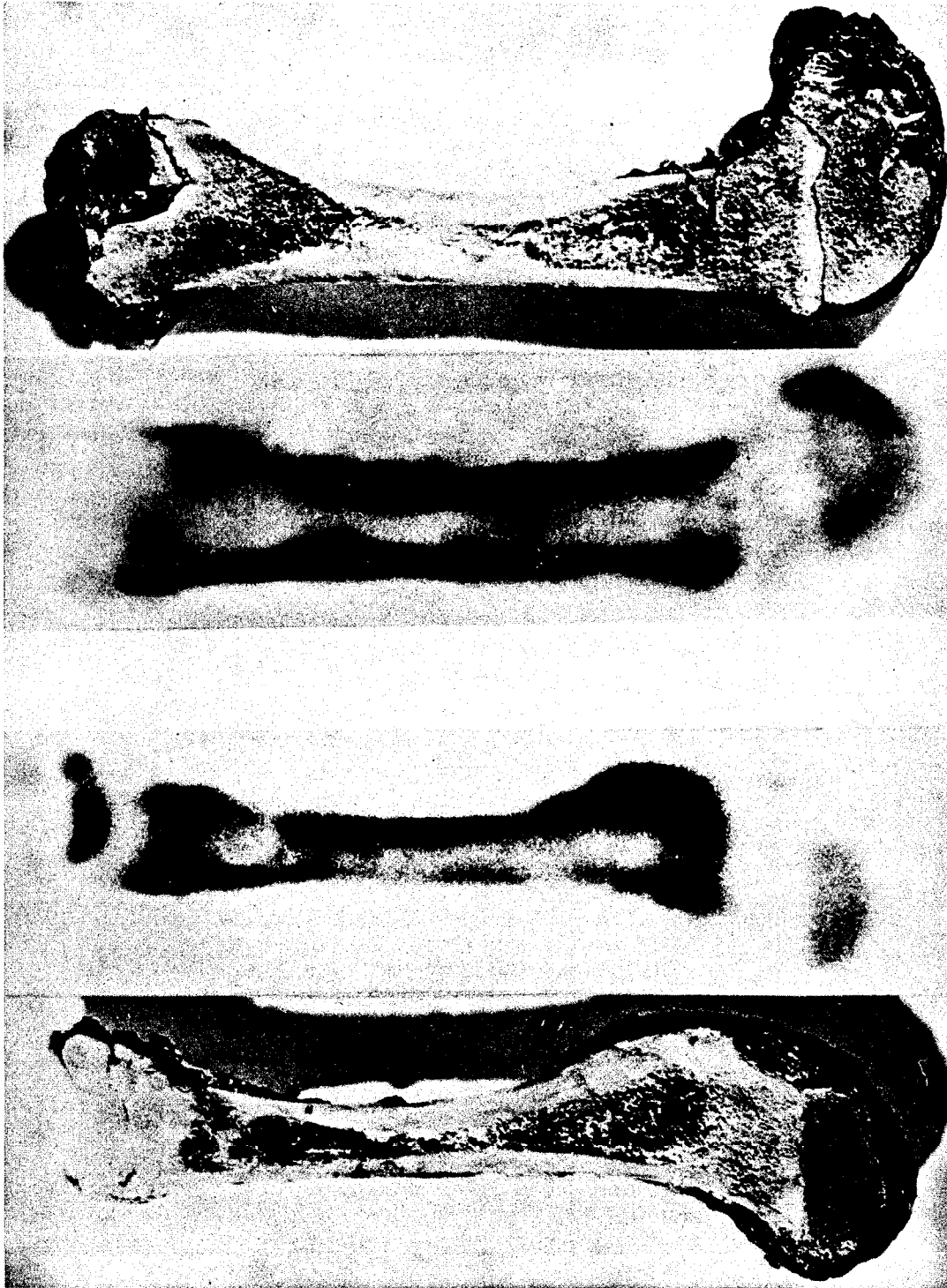


Fig. 3.5—Autoradiograph of Tibia and Femur of Fig 6 Sacrificed 57 Days Postdetonation.



Fig. 3.6—Autoradiograph of Tibia of Adult Sow Sacrificed 38 Days Postdetonation.

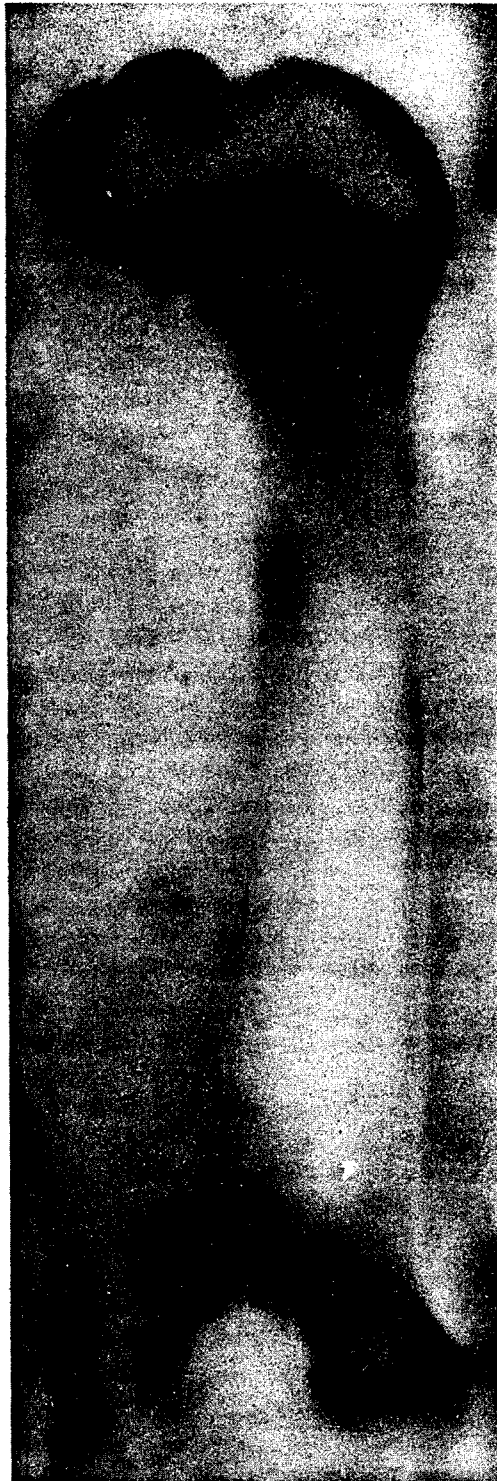


Fig. 3.7—Autoradiograph of Femur of Boar Sacrificed 26 Days Postdetonation.

However, none of the changes could definitely be ascribed to radiation. Sections of bone examined by Lisco at ANL indicated no detectable pathological changes.

3.5 EGG PRODUCTION IN CHICKENS

In birds extraordinary demands are made on the calcium metabolism in the production of egg shells. It was, therefore, of particular interest to observe, during the process of egg production, the metabolism of those internally deposited radioelements that are metabolically similar to calcium.

Forty-four days after detonation, a group of hens from Rongelap began laying eggs for the first time since their collection. During the next month and a half, 319 eggs were laid by 13 hens. All the eggs were normal except for two eggs from one hen which were laid without shells. The shells were complete, smooth and of normal shape. The weights of the eggs ranged from 30 to 64 g, but those from a given hen were of uniform weight. In 14 eggs studied, the shell and membranes weighed an average of 13.6 per cent of the whole egg weight, and the ashed shell weighed 6.8 per cent of the whole egg weight. These values are within the normal range for eggs of domestic hens.

The gross beta and gamma activities of the shell, albumen, and yolk were measured in the first 50 eggs obtained, and the gamma activity of the shell was measured in the remainder of the eggs. An increasing amount of gamma activity appeared in the shells of the first few eggs laid by each hen. The maximum gamma activity was usually noted in about the eighth egg laid. After the activity reached a maximum value, the subsequent eggs in the series showed a general decline in activity. An example of this phenomenon is illustrated in Fig. 3.8.

The highest gamma activity found in a single egg shell was 66,300 counts/min, measured at 60 days postdetonation (Fig. 3.8). For a gamma energy of approximately 1 Mev, this figure corresponds to 0.07 μ c. The yolks and albumens had much less activity than the shells, as was anticipated. The average distribution of gamma activity in the eggs is given in Table 3.5.

The results of the radiochemical analysis of two eggs are presented in Table 3.6.

The alkaline earths are the principal fission products deposited in the shell. In the albumen and yolk the beta activity contributed by the alkaline earths was only a little greater than that associated with the rare earths.

The pattern of deposition of the radioactivity within the egg was also studied by means of autoradiographs. A series of 50 eggs was hard boiled and sectioned, and autoradiographs were prepared of the cut surfaces. Only four of the yolks of these 50 eggs were sufficiently radioactive to produce autoradiographs (see Fig. 3.9). There is a correlation between the rings of radioactivity in the yolk and those of pigment.

The amount of activity removed from the body of the chicken through egg laying is very much greater than the amount excreted in the urine and feces during the period of this study. Egg production in the chicken represents a unique form of natural decontamination.

3.6 FERTILITY AND HATCHABILITY STUDIES IN CHICKENS

Fertility studies on the contaminated chickens were begun three and one half months postdetonation, with the mating of hens and roosters and the incubation of the eggs obtained. In the first clutch of 20 eggs, four were hatched. One of the chicks had the crippling slipped-tendon condition, congenital perosis, which is not uncommon. Radioanalysis of the chick tissues indicated that only a barely detectable amount of radioactive material was transferred to the chick, although the mother hen had at this time an appreciable contamination.

In another hatch six months postdetonation, 65 eggs were incubated. Of these, 28 were infertile, 3 fertile ones were opened prematurely, 11 developed complete embryos but failed to hatch, and 23 live chicks were hatched, one of which had congenital perosis. The latter chick and six normal ones were sacrificed and their tissues radioanalyzed. Again, only barely detectable amounts of internally deposited activity were found. The remaining baby chicks are being raised and observed for possible long-term effects. At the present time all the chicks

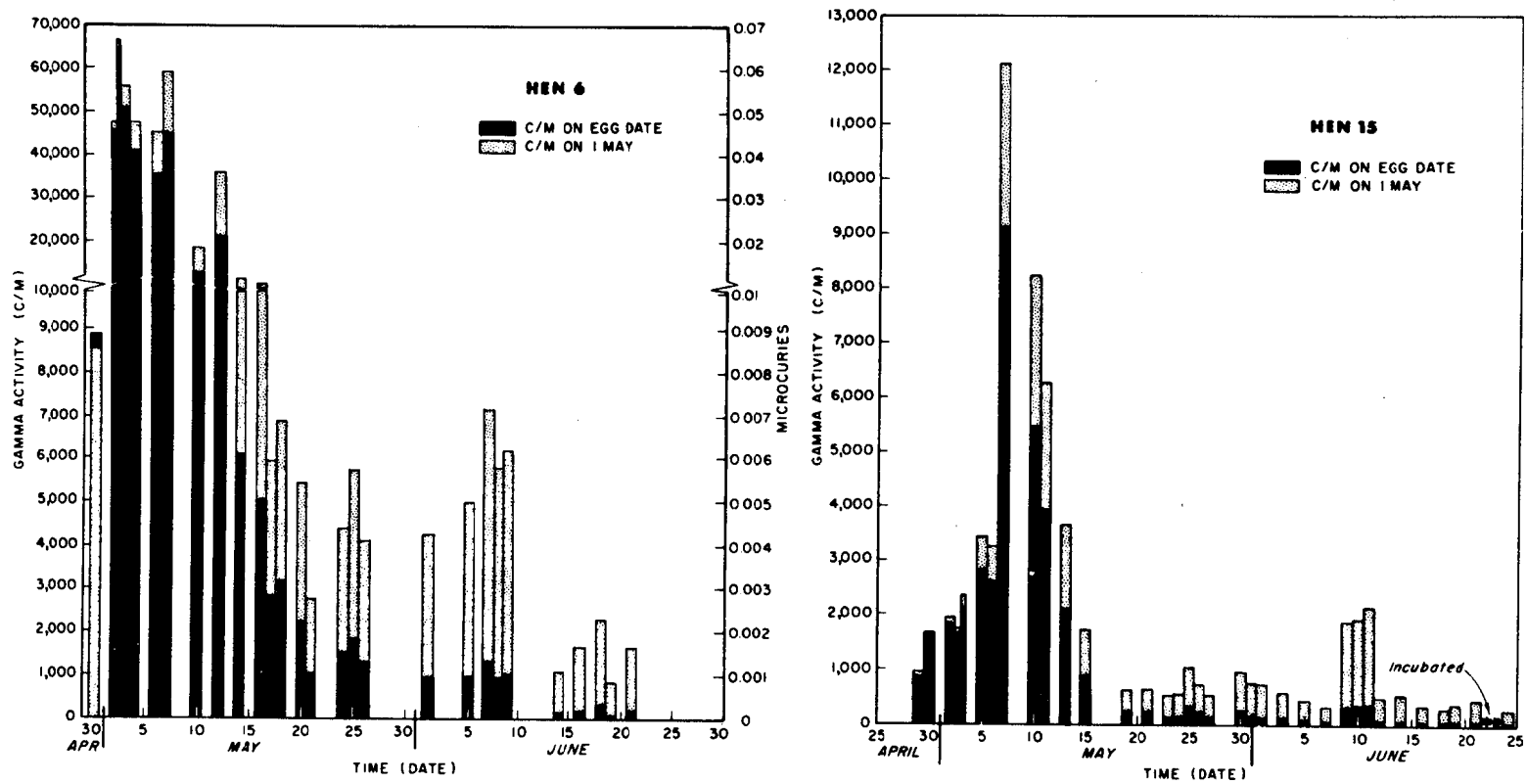


Fig. 3.8—Appearance of Gamma Activity in Egg Shell as a Function of Time. (Maximum gamma activity was noted in eighth egg laid as typically illustrated in hen 15. The gamma activity in hen 6 represents the highest concentration of activity seen in any of the shells.)

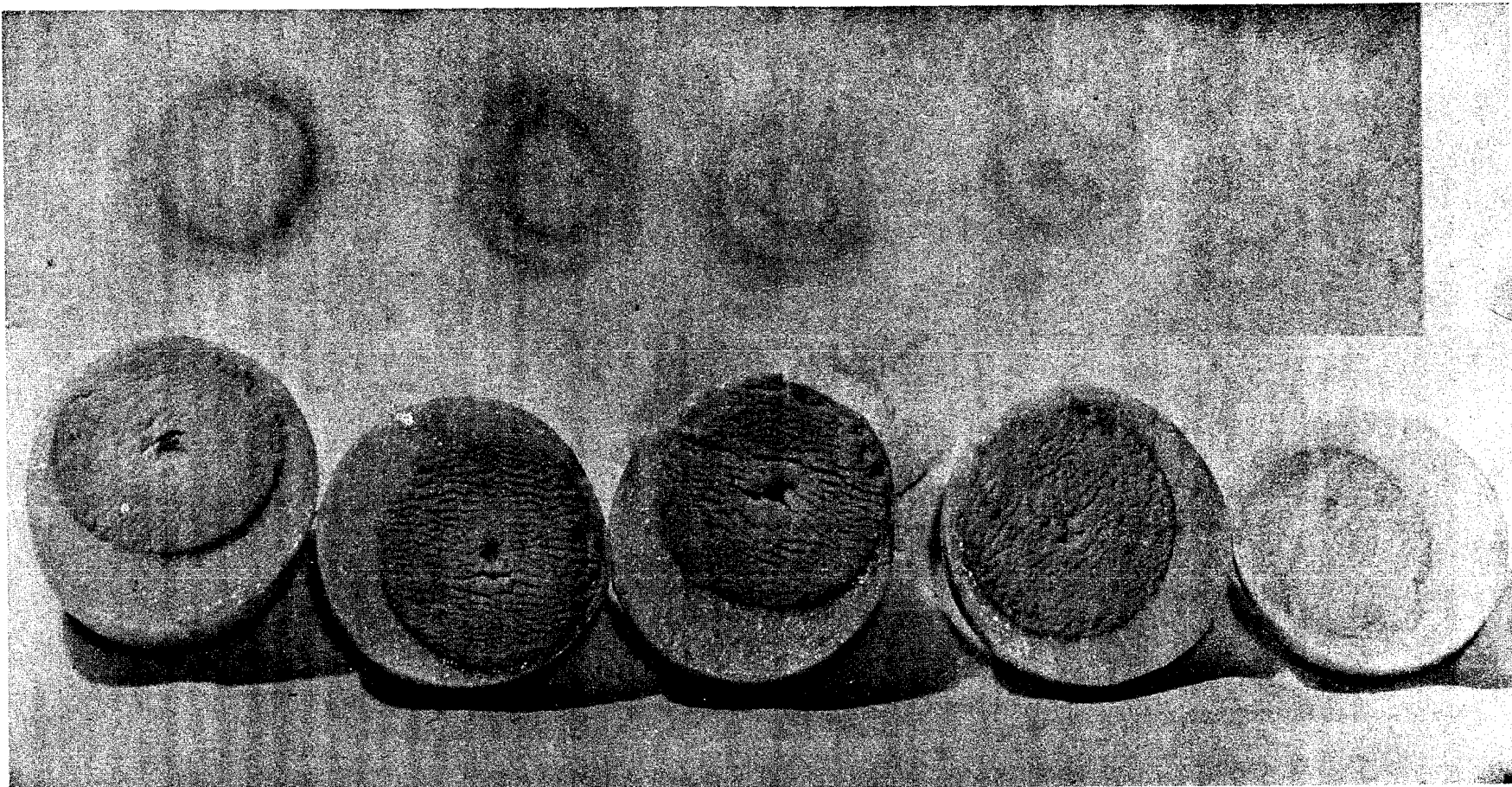


Fig. 3.9—Autoradiograph of Chicken Eggs, Showing Pattern of Deposition of Fission Products in Yolk.

are growing normally and are in good health. Comparison of the fertility and hatchability data from the Rongelap hens with those from domestic hens does not demonstrate any effect of radiation on these phenomena.

TABLE 3.5—Distribution of Radioactivity in Chicken Eggs

	Total gamma activity (%)	Total beta activity (%)
Shells	81	68
Yolks	15	23
Albumen	4	8

TABLE 3.6—Radiochemical Analysis of Chicken Eggs
(Beta activity given in disintegrations per minute per total tissue at 4 months postdetonation)

Sample	Sr ⁹⁰	Ba ¹⁴⁰	Rare earths	Gross beta activity
Egg No. 27				
Yolk	355	546	663	1,560
Albumen	52	92	90	260
Shell	18,080	3520	6060	30,000
Egg No. 29				
Yolk	315	825	997	2,178
Albumen	45	132	132	316
Shell	22,300	4900	7830	38,000

3.7 INTERNAL RADIOACTIVE DECONTAMINATION STUDIES IN CHICKENS

A study was undertaken to determine the ability of both sodium EDTA and zirconium citrate (Schubert and White¹⁶) to increase the excretion rate of internally deposited fission products in the Rongelap chickens. On the basis of previous experience, it was not expected that any appreciable decontamination could be effected at the time of this experiment (four months following internal radioactive deposition).

The excretion rates of eight chickens with large body burdens of internal contaminants were determined for a period of four days as the base line for the study. Following this, two chickens were injected daily intraperitoneally with 75 mg sodium EDTA for four days; two received injections of 70 mg of zirconium citrate (Schubert and White¹⁶); and two were injected with both zirconium citrate and sodium EDTA. Two chickens were kept as controls. The mean beta and gamma activity excreted by these chickens was determined individually for each of the treatment days and for one day following cessation of treatment. Neither the zirconium citrate nor the sodium EDTA alone was effective in increasing the excretion rate as reflected by the beta-activity measurements made. The combined administration of zirconium citrate and sodium EDTA, however, doubled the excretion rate of the beta activity. No detectable change in the rate of excretion of gamma activity was noted. The excretion rate of fission products at this long period postcontamination was less than 0.1 per cent per 24 hr. Thus the enhancement of the excretion rate by the combination of zirconium citrate and sodium EDTA did not significantly decrease the total body burden.

3.8 SUMMARY AND CONCLUSIONS

Studies of animals provided data on the nature and distribution of the radioisotopes in the tissues and the excreta. Over 90 per cent of the activity in the body of animals was localized in the skeleton. The pattern of deposition of the fission products in the skeleton seen in autoradiographs resembles that of the alkaline earths. Morphological changes which were observed in some of the bones may be the result of the exposure of the animal to radiation, although the effects of severe dietary changes and disease cannot be ruled out.

The alkaline earths, Sr⁸⁹ and Ba¹⁴⁰, and the rare-earth group together constituted 75 per cent of the gross beta activity in the pig at 82 days postdetonation. The fish and clam had a much lower concentration of the alkaline and rare earths, and a body burden considerably higher than that of the land animals.

The internal distribution of fission products in the pig is probably representative of the distribution in human beings. An estimate of the human body burden was derived from the data on pigs.

Studies made on egg production of contaminated hens gave no evidence of any effect of radiation. The rate of production of the eggs was normal, and the eggs produced were also normal. The extraordinary ability of fowl to mobilize calcium in shell formation resulted in the presence of very high activity in the shells of the first few eggs. The activity was associated with the fission products of the alkaline-earth group. A significant amount of activity was found in the yolk and lesser amounts in the albumen. The removal of activity from the body of chickens by egg production provides an effective natural decontamination process.

Fertility of the hens and hatchability of the eggs produced by the mating of contaminated roosters and hens showed no effect of radiation. The baby chicks hatched from these eggs are growing normally, and the amount of radioactivity in their tissues is barely detectable.

Although the administration of the combination of zirconium citrate and sodium EDTA to chickens doubled the excretion rate of fission products, the rate at this long time after exposure was so low that the body burden was little affected.

In the six-month period postdetonation neither significant gross changes nor pathological changes that could be definitely ascribed to radiation were detected in any of the animals from Rongelap. Gross beta activity of urine and tissue samples indicated that all the animals had significant internal contamination. The level of internally deposited radioisotopes in the pigs from Rongelap was 10 times the amount in human beings from this area. The difference in the amount of internal contamination of the animals and the human beings was the result of the prolonged stay of the animals in the contaminated area. The chickens were found to have the same concentration of radioisotopic material per unit of body weight as the pigs.

All the animals remaining will be observed throughout their lifetime for the possible appearance of any long-term biological effects resulting from their exposure to external and internal radiation.

CHAPTER 4

ENVIRONMENTAL STUDIES

4.1 CHEMICAL AND PHYSICAL CHARACTERISTICS OF THE FALLOUT MATERIAL

The fallout material consisted largely of calcium carbonate and calcium oxide particles. The fission products were adsorbed mainly on the smaller particles. The fallout material was found to be 10 per cent water soluble and almost completely soluble in acid. An extensive physical and radiochemical analysis of the fallout material, and determination of its gamma and beta spectrum, was performed by the Chemical Technology and Nucleonics Division of the NRDL and will be issued as separate reports.

4.2 RADIOCHEMICAL ANALYSIS OF FOODSTUFFS, WATER, AND SOIL

4.2.1 Methods

Radiochemical analyses of the food, water, and soil samples reported here were performed by the Chemical Technology Division of the NRDL. Additional analyses were made by NYOO-AEC and the NMRI. Since a complete radiochemical analysis of all biological samples was not feasible, only Sr⁸⁹, Ba¹⁴⁰, fission material, and the rare-earth groups were measured in addition to the total beta activity. These measurements were found to be the most useful for determining the nature of the contamination.

The initial external activity of the samples, as detected with a rapid survey instrument (1M-57/PDR-27 Radiac meter), is listed in Table 4.1. The values are given in milliroentgens per hour at surface contact unless otherwise stated. For the food plants with large surface areas, a range of activities is given.

A more accurate estimate of the activity of environmental samples was obtained by using a 4π gamma ionization chamber. A number of samples were first evaporated to dryness and wet ashed with fuming nitric acid. The residues were made up to volume in HCl, and an aliquot evaporated to dryness for beta-activity determination. The samples were then counted with a proportional counter, with appropriate corrections made for mass absorption, backscatter, and geometry of the counter. The disintegration rate determined was plotted against millivolt reading of the ionization chamber for the samples. This ratio was then used to convert the gamma ionization chamber readings into disintegrations per minute.

The values obtained in this manner are listed in Tables 4.2, 4.3, and 4.4, discussed below. An empirically derived ratio of alpha activity to ionization chamber reading was also obtained on a number of samples. An estimate of the alpha activity of the remaining samples was then obtained by applying this ratio.

TABLE 4.1—Survey of Initial Activity of Samples Collected from Rongelap

Samples	Initial activity at surface (mr/hr)	
	Rongelap	Utirik
Day 32 Postdetonation		
I. Soil (containing considerable organic material)	2.5*	
Soil	4.0*	
Grass	5.0	
Pandanus (long stem)		
1. Fruit	0.5 to 5.0	
2. Stem	5.0	
3. Leaves	0.5 to 5.0	
Day 52 Postdetonation		
II. Grass	2.0	0.35
Water		
1. Well	0.1	0
2. Cistern	0.1	0
Pandanus (2)	1.5-5.0	1.5
Coconut tree sap		
1	0.1	0
2	0.1	0
3	0	0
Coconuts (2)	0.4	0
Arrowroot	0.25	0.1
Papaya	0	0
Soil		
1. Surface sample	2.0	0.2
2. Sample (6 in. deep)	0.2	
Breadfruit		
Squash	0.2	
Eggs (gull)	2.0	
3 fish, 1 clam (in formalin)	0.4	
Day 54 Postdetonation		
III. Hair	2.0	
Grass (from village)	5.0 at 2 in.	
Thatch	5.0 at 12 in.	
Coconut (3)	0.1-5.0	
Pandanus (3)	0.3-5.0	
Papaya (2)	1.5+3.5	
Day 25 Postdetonation		
Dirt (about 1 liter)	4.5*	
Water (about 1 liter)	0.5*	
Day 32 Postdetonation		
Water (about 1 liter)	0.5	

* Measured through 0.3 cm of glass.

4.2.2 Results and Discussion

(a) **Food.** A summary of the alpha and beta activity on the surface of representative food samples from Rongelap at 43 days postdetonation is presented in Table 4.2. The beta activity was relatively high, the alpha activity fairly low. The surface contamination found on Utirik samples was considerably lower than that found on samples from Rongelap.

After the surface contamination of the foodstuffs was measured, decontamination measures were studied. Coconuts, papayas, and pandanus were decontaminated by scrubbing with

TABLE 4.2—External Contamination on Representative Rongelap Food Samples
(Day 42 postdetonation)

Food sample	Alpha activity of fission material ($\mu\text{g} \times 10^4$)	Beta activity (Sr^{89} equivalent) (μc)
Coconuts (3)	2.2–3.3	0.9–1.3
Papayas (2)	0.6–1.0	0.2–0.4
Pandanus (surface)	22–56	8.9–22.7
Pandanus (peel)	3.5	0.76
Pandanus (leaves)	8	1.71
Pandanus (stem)	4	0.40

tap water and detergent. Approximately 90 per cent of the activity was removed from the papayas and the coconuts (Table 4.3). Decontamination of the pandanus was more difficult than that of the other foodstuffs because of their rough surface. Only 50 per cent of the contamination of the pandanus could be removed. In general, effectiveness of decontamination was inversely proportional to the roughness of the surface of the fruit.

After the surface was washed, the edible portions of the food were surveyed with an ionization chamber. Beta activity of individual fruits ranges from 0.001 μc for a papaya to 0.2 μc for a pandanus (Table 4.4). The relatively high levels of activity detected in the pandanus were probably the result of contamination from the rough external surface of the fruit. It is possible that active material was transported through the root system of the plant and deposited in the fruit during the two-month period that the fruits were growing in the contaminated area. The arrowroot may very likely have been contaminated in this manner since it is a tuber growing underground.

The likelihood of the incorporation of radioactive contaminants into plants is supported by the very high levels of activity found in the Rongelap coconut tree sap at two months (1.4 μc /liter) and in the Utirik specimens, which showed about $\frac{1}{10}$ of this activity (Table 4.4). The presence of this activity in the tree sap appears to be the result of uptake of contaminated water by the root system since both the water and the tree sap had approximately the same isotopic composition.

Larsen¹² has shown that small amounts of fission products are immediately taken up by plants growing on soils contaminated from fallout material. This early uptake of fission products seems to be a function of the solubility of the material. The predominant fission product incorporated into the plants in Larsen's studies as well as in the present studies is Sr^{89} . This high strontium concentration is probably a function of the relatively high solubility of the element in water. Thus Sr^{89} , because of its abundance and long half life, is the radionuclide of greatest potential hazard associated with ingestion of the contaminated food.

The radiochemical analysis for alkaline-earth and rare-earth activity of the material found on pandanus, grass, and thatch, as well as in water and coconut tree sap, is listed in Table 4.5. The highest levels of beta activity found on thatch, grass, and pandanus at this time appeared in the rare-earth fraction. The alkaline earths, strontium and barium, contributed a small fraction of the total beta activity. There was a marked variation in the ratio of the strontium,

TABLE 4.3—Removal of Radioactivity from Rongelap Foodstuffs by Washing Procedures

Samples	Unwashed food		Water wash		Water plus detergent		By both washes	
	Beta (Sr ⁹⁰ equiv.) (μc)	Alpha of fissile material ($\mu\text{g} \times 10^3$)	Beta (Sr ⁹⁰ equiv.) (μc)	Alpha of fissile material ($\mu\text{g} \times 10^3$)	Beta (Sr ⁹⁰ equiv.) (μc)	Alpha of fissile material ($\mu\text{g} \times 10^3$)	Beta (Sr ⁹⁰ equiv.) (μc)	Alpha of fissile material ($\mu\text{g} \times 10^3$)
Coconuts								
1	1.21	0.30	0.33	0.08	0.76	0.19	1.09	0.27
2	0.93	0.22	0.42	0.10	0.42	0.10	0.84	0.20
3	1.31	0.33	0.76	0.19	0.42	0.11	1.18	0.30
Pandanus								
1 (small)	8.90	2.20	2.55	0.63	1.90	0.47	4.45	1.10
2 (large)	22.72	5.62	0.63	1.26	6.27	1.55	6.90	2.81
Papaya								
1 (short)	0.39	0.10	0.19	0.05	0.18	0.04	0.37	0.09
2 (long)	0.24	0.06	0.12	0.03	0.10	0.02	0.22	0.05

TABLE 4.4—Radioactive Contamination Associated with Edible Portions of Food Plants of Rongelap and Utirik (Day 69 postdetonation)

Food plant	Sample	Estimated μc beta activity*	
		Rongelap	Utirik
Pandanus	Entire edible portion	0.02–0.2 (3)	0.036
Coconut	Entire milk	0.004	0.008
Papaya	Entire edible portion	0.025	0.001
Arrowroot	Entire washed edible portion	0.004 (2)	
	Entire unwashed plant	0.152 (3)	
	Entire unwashed plant		0.160
Squash	Entire edible portion	0.200	
Coconut tree sap	1 liter	1.41 (3)	0.11 (3)

* Number of individual samples analyzed are given in parentheses.

TABLE 4.5—Radiochemical Analysis of Various Rongelap Samples

Sample	Total beta activity	Strontium activity	Barium activity	Rare-earth activity		
				Sepr. time	Time t	t (hr)
Disintegrations per Minute per Total Sample, Day 72 Postdetonation						
Thatch (7.2 g)	1.85×10^7	1.06×10^5	9.15×10^5	6.88×10^6	5.84×10^6	66
Grass (9.8 g)	2.21×10^6	1.03×10^5	3.77×10^3	8.28×10^5	7.35×10^5	52
Pandanus (entire core)	2.70×10^5	5.21×10^4	1.96×10^4	1.92×10^5	1.82×10^5	52
Water						
Filtered	1.24×10^5	5.12×10^4	1.07×10^4	2.11×10^4	1.24×10^4	66
Solids	2.85×10^5	6.70×10^3	1.61×10^5	1.55×10^5	1.46×10^5	52
Wash from hen No. 1		1.05×10^4	9.8×10^3	2.38×10^2		
Disintegrations per Minute per Total Sample, Day 93 Postdetonation						
Rongelap coconut tree sap (300 cc)		1.22×10^5	2.72×10^4	4.57×10^4		
Rongelap cistern						
Water liquid		4.29×10^5	5.6×10^4	9.0×10^4		
Solid (800 cc)		2.26×10^3	7.45×10^3	11.2×10^3		
Total		4.31×10^5	6.4×10^4	10.1×10^4		

barium, and rare-earth components between the samples of thatch, pandanus, and water, probably as a function of the different solubilities of the radioelements.

(b) Water. One of the most important sources of contamination for the Marshall Islanders was their water supply. Water on the island of Rongelap was obtained from one of two systems, open cisterns or open wells. Rongelap and Utirik cistern water showed higher levels of contamination than the corresponding well-water samples, as might be expected. At 34 days postdetonation a representative sample of water taken from an open well had a beta activity of approximately $0.88 \mu\text{c}/\text{liter}$. Another sample of water taken from a cistern had an activity of $2.5 \mu\text{c}/\text{liter}$ at 34 days postdetonation.

The distribution of activity between particulate and dissolved material in the well water is presented in Table 4.6. Much of the activity was associated with particles that could be filtered out of the water. Individual isotope analysis (Table 4.5), as well as the beta to gamma ratio of Rongelap water samples (Table 4.7), indicates that the ratio of strontium to rare earth, at 2 months postdetonation, is much higher in the liquid phase than in the filtered solid particles. This is largely the result of the higher solubility of strontium salts compared to the other radioelements and its longer radiological half life. Sr⁸⁹ is potentially the most hazardous

TABLE 4.6—Distribution of Activity Between Particulate and Dissolved Material in Rongelap Well Water (43 Days Postdetonation)

Sample	Beta activity μc (Sr ⁸⁹ equivalent)	Total beta activity, %
A. Gross assay		
H ₂ O + suspended matter	0.54	60.6
HNO ₃ wash of bottle	0.35	39.4
Total	0.89	100.0
B. Centrifugation of an aliquot		
Supernatant	0.018	35.3
Precipitate	0.033	64.7
Total	0.051	100.0
C. Filtration		
1. Through fine frittered glass		
Supernatant	0.010	20.0
Material filtered out	0.040	80.0
Total	0.050	100.0
2. Through #42 Whatman paper and fine frittered glass bed		
Supernatant	Background	Less than 10
Material filtered out	0.045	90-100
Total	0.045	100

fission product associated with ingestion of water or with food in this particular situation. This would not be true for situations in which exposure to fallout occurred at later times than that which occurred here; nor would it be correct when long periods of exposure are to be considered, as compared with the short exposure of the Marshallese.

There was less than 7×10^{-6} $\mu\text{g/liter}$ of fissile material found in the cistern water examined.

(c) Soil, Thatch, and Grass. The most representative samples of fallout material on the contaminated islands were obtained from soil and roof thatch. A sample of soil taken at 25 days in the center of the village on Rongelap showed an activity of approximately 0.13 $\mu\text{c/g}$. The contamination was associated with very fine particulate matter and was uniformly distributed throughout the individual samples. Autoradiographs of the soil samples indicated that the activity was associated chiefly with fallout particles ranging from 60 to 200 μ in diameter. A sample of earth was separated in a multiple sieve and the specific activity of the various fractions determined in an ionization chamber. The bulk of the activity was associated with the smaller earth-particle sizes (less than 400 μ) in diameter, and the highest specific activity was observed on the smallest earth-particle size (less than 200 μ), Table 4.8.

TABLE 4.7—Beta-Gamma Data on Water from Rongelap and Utirik
(66 Days Postdetonation)

Sample	Amount of sample (cc)	Ion chamber Rdgs (mv)	Beta activity (Sr ⁸⁹ equivalent) (μ c)	Beta to gamma ratio (μ c/mv)
Rongelap Water				
Well solids	800	0.042	0.09	2.1
Well filtrate		0.011	0.04	3.6
		0.053	0.13	
Cistern solids	800	0.067	0.10	1.5
Cistern filtrate		0.180	0.62	3.4
		0.247	0.72	
Utirik Water				
Well solids	600	0.002	0.002	1.0
Well filtrate		0.005	0.006	1.2
		0.007	0.008	
Cistern solids	800	0.029	0.02	0.7
Cistern filtrate		0.041	0.15	3.7
		0.070	0.17	

TABLE 4.8—Distribution of Beta Activity Among Various Size Rongelap Earth Particles

Diameter range (μ)	Weight of fraction (g)	Total weight (%)	Beta activity (μ c/fraction)	Beta activity (percentage of total activity)
>1650	148.9	44.6	0.89	3.3
833-1650	15.3	4.5	0.33	1.2
416-833	42.6	12.7	1.20	4.4
208-416	121.2	36.3	18.10	66.7
<208	5.8	1.7	6.46	23.9

Absorption measurements made on the earth samples gave the results listed below.

Particle-size range	T _{1/2}	Percentage
800-1600 μ	104 mg Al/cm ²	37
	38 mg Al/cm ²	43
	7 mg Al/cm ²	20
400-800 μ	92 mg Al/cm ²	54
	26 mg Al/cm ²	35

(Remaining fraction very soft)

The activity associated with earth samples had about equal amounts of a hard and a medium component, in addition to a very soft component.

After the initial gross activity was surveyed, samples of human hair, grass, thatch roofing, and soil were analyzed for their beta and alpha activity (Table 4.9). The presence of fissile material on the thatch was of the order of 10^{-3} $\mu\text{g/g}$ and on grass about one-half that on the thatch, at 35 days postdetonation. The fissile material activity in human hair was quite low.

TABLE 4.9—Gross Beta and Alpha Activity of Soil, Grass, and Thatch from Rongelap

Sample	Days post-detonation	Beta activity (Sr ⁸⁹ equivalent) $\mu\text{c/g}$	Fissile material ($\mu\text{g/g} \times 10^3$)
Thatch			
T1-5	35	2.7	1.3
T6	56	1.6	
Grass			
G1-3	35	0.5	0.3
G4	41	0.3	0.1
G5	67	0.09	
G6 (Utirik)	67	0.03	
Soil			
S1 (organic)	41	0.10	0.47
S2 (sandy)	41	0.06	0.03
S3 (surface)	67	0.06	3.0
S4 (6 in. deep)	67	0.003	1.9
S5 (Utirik)	67	0.006	0.8

4.3 SUMMARY AND CONCLUSIONS

The fallout material was found to consist largely of calcium oxide and calcium carbonate. The fission products were adsorbed mainly on particles of 60 to 200 μ . This material was 10 per cent soluble in water and completely soluble in acid.

Radioanalysis of soil and water samples from Rongelap indicated high levels of radioactive contamination from the fallout at early times following detonation. Significant amounts of beta activity as well as smaller amounts of fissile material were present on the external surfaces of plants 42 days after detonation. Only small amounts of beta activity and no alpha activity were detected in the edible portion of foods. It appears that during the first month a limited amount of fission products is available to plants growing on contaminated soil. However, high levels of activity in the coconut tree sap were detected which had an isotopic composition very similar to that of the water analyzed.

Radiochemical analysis of thatch and water samples indicated a marked variation in the ratio of Sr⁸⁹, Ba¹⁴⁰, and the rare-earth group probably as a function of the different solubilities of the radioelements. The activity of food, water, and soil samples from Utirik was approximately $\frac{1}{10}$ of those of Rongelap.

It appears from this study that the ingestion of contaminated water was one of the principal sources of contamination for the Marshallese. Of the individual radionuclides, Sr⁸⁹, because of its high solubility and relatively long half life, was probably the isotope of greatest potential hazard in the environment.

CHAPTER 5

RECOMMENDATIONS

5.1 FIELD

1. In any emergency situation arising from a contaminating type detonation, the early collection of 24-hr urine samples would be highly desirable.
2. EDTA decontamination procedures should also be applied early following contamination. EDTA is of potential benefit only when administered at early intervals following exposure.
3. Information concerning the nature of the internal-radiation hazard should be widely disseminated to minimize the inhalation and ingestion hazard.
4. Simple procedures for decontamination of food and water should be developed for use in emergency situations.

5.2 LABORATORY

1. More data on the ratio of body burden of individual fission products to the amounts excreted in the urine as a function of time should be obtained. These data would serve as the basis for an estimate of the internal hazard to human beings exposed to fission products.
2. The combined effects of internal and external radiation should be studied in detail.
3. Radiochemical techniques for the isolation and measurement of individual radionuclides in urine and in the tissues should be improved.
4. The long-term effects of internally deposited radioactive isotopes, particularly the effects in bone, should be studied.

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