

Chapter V

Internal Deposition of Radionuclides
In Human Beings and Animals

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5.1 Introduction

FOLLOWING A NUCLEAR detonation in the spring of 1954, a large group of people were contaminated with fission products. In addition to a sublethal external gamma radiation exposure and beta irradiation of the skin, delectable amounts of radionuclides were deposited internally. It has been assumed that in all situations resulting from a contaminating event, the ratio of external to internal dose would be exceedingly high. However, 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. Three general problems were investigated: (1) The determination of the contribution of the internal contamination to the acute radiation syndrome observed; (2) The possibility of long term effects, and (3) The qualitative and quantitative nature of the internal contamination produced by exposure of individuals to mixed fission products. There was no previous situation in which human beings were exposed to an environment contaminated with mixed fission products. Concurrent studies were undertaken by the Japanese, however, on radioactive materials to which a small group of Japanese fishermen, near Rongelap at the time of the detonation, were exposed. The report of the extensive investigations undertaken on the ashes by the Japanese have been published (4).

Evaluation of the internal contamination of the human beings was made by a study of the radioelements excreted. As 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 which 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.

5.2 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 which 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.

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 two hundred radioisotopes are produced in the fission process, only a few are potential chronic internal radiation hazards. These fission products, which are listed in Table 5.1, constitute a high percentage of the fission yield, and localize chiefly in bone. The "bone-seekers" have, in

Table 5.1.—Biologically Hazardous Internally Deposited Fission Products

RADIO-ELEMENT	TYPE OF RADIATION	FISSION ¹ ABUNDANCE PERCENT	HALF-LIFE		FRACTION REACHING CRITICAL ORGAN ^{2,3}	
			RADIOL. ¹ DAYS	BIOL. ^{2,3} DAYS	BY INGESTION	BY INHALATION
Sr ⁹⁰	β	4.6	53	3.9×10^3	.25	0.22
Y ⁹¹	β	5.9	57	>500	2.8×10^{-4}	0.14
Zr ⁹⁵	β, γ	6.4	65	>100	.35	
Ru ¹⁰³	β, γ	3.7	42	20	.04	
Ru ¹⁰⁶	β	0.5	365	20	.04	
I ¹³¹	β, γ	2.8	8	180	0.2	0.15
Ba ¹⁴⁰	β, γ	6.0	12.8	~200	.07	.20
La ¹⁴⁰	β, γ	6.0	1.7	35	1.2×10^{-3}	0.1
Ce ¹⁴¹	β, γ	5.7	28	>100	.25	
Pr ¹⁴³	β	5.4	13.8	50	1.3×10^{-3}	.063
Ce ¹⁴⁴	β, γ	5.3	275	500	2×10^{-4}	0.10

From: ¹ Seaborg and Perlman, Rev. Mod. Physics, 20:585, 1948.

² Hamilton, J. G. Rev. Mod. Physics, 20:718, 1948.

³ Handbook 52, U. S. Dept. of Commerce, National Bureau of Standards.

general, long radiological and biological half-lives and produce high-energy beta particles. Thus, they cause greater damage to bone and to the radiosensitive 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 is 1 to 2 micrograms of radium. Radium is a particularly hazardous element when deposited internally because of its long biological and radiological half-life.

Very few data are available on the long term biological effects in human beings of the shorter lived isotopes such as Sr⁹⁰, I¹³¹, P³² and Na²⁴. The metabolism, excretion and biological effects of a number of fission products have been

studied in animals by Hamilton (1), Abrams (2), Bloom (3). However, most of these studies do not cover the problem of the long term effects in animals produced by small amounts of internally deposited isotopes.

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 individuals exposed to the air burst 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 a few bone seeking radioisotopes in the body (5). Maximum permissible body content of other radioisotopes are estimated on quantities resulting in a dose of 0.3 rem per week to the tissue of highest concentration.

5.3 Internal Contamination in Human Beings

THE INTERNAL CONTAMINATION study was begun 15 days post-detonation with the collection of pooled 24 hour urine samples from the Marshallese and American groups. Maximum activity in the urine occurs during the first few days after internal contamination. By 1 week an approximate equilibrium state is reached in which the contaminants remaining in the body are firmly fixed, chiefly in the skeletal tissues. The activity in the urine then derives from radioelements which have been replaced in the natural process of biological turnover. Thus, the study made is essentially that of an equilibrium condition.

The urine samples were sent 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 for the Los Alamos Scientific Laboratory (LASL). Similar samples collected 44 days post detonation were also sent there. On the 23rd, 24th and 47th days post detonation, 24-hour urine collections from each individual from Rongelap and Ailinginae were sent to the New York Operations Office, Atomic Energy Commission (NYOO-AEC) for analysis. In addition, samples from representative individuals in these groups were collected 2½, 3 and 6 months post detonation and sent to NYOO-AEC.

The USNRDL collected samples from each member of the exposed groups at 43 and 46 days post detonation. Samples from representatives of these groups were also collected at 2½, 3 and 6 months by the USNRDL. In addition, samples from a representative group of 6 Americans and 20 Marshallese were collected for 6 consecutive days beginning 33 days post detonation.

5.31 Methods

As a complete radiochemical analysis of all the urine samples was not feasible, samples were analyzed for only Sr⁹⁰, 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.

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

The beta activity was counted with a thin end window Geiger-Muller counter. The counter was calibrated with a U₂O₃ standard, and an appropriate correction for self-absorption was made using a Sr⁹⁰ standard.

5.32 Findings and Interpretations

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, as it was derived chiefly from short-lived radioisotopes. For example, at 3 months post detonation, the mean activity of the urine of adults from Rongelap was 28 percent of the value measured 45 days post detonation, and at 6 months, the activity in the urine was barely detectable in most of the individuals.

Comparison of the means of the urine samples for the adults from Rongelap and Ailinginae and from Americans from Rongerik indicated that at 45 days post detonation the

Table 5.2.—Summary of Human Urine Analysis, Gross Beta Activity

TIME POST DETONATION	1½ MONTHS			2½ MONTHS			3 MONTHS			4 MONTHS		
	No.	VOLUME (24 HRS) ML	D/M 24 HRS	No.	VOLUME (24 HRS) ML	D/M 24 HRS	No.	VOLUME (24 HRS) ML	D/M 24 HRS	No.	VOLUME (24 HRS) ML	D/M 24 HRS
Rongelap												
Age in years												
A (<5)	7	165	404							8	360	12
B (5-16)	11	439	758							12	510	5
C (>16)	31	581	1208	10	824	705	10	379	339	33	625	0
Ailinginae												
Age in years												
A (<5)	1	150	217									
B (5-16)	2	275	126							3	400	0
C (>16)	10	722	553							12	655	0
American	25	1158	309									

All values corrected for decay.

highest activity was in the Rongelap group (Table 5.2). The Ailinginae group had less than half that of the Rongelap group, and the Americans had about one-quarter the activity of the Rongelap group.

The mean gross beta activity of the urine of the three groups above was roughly proportional to the external dose each group received. However, a comparison of the mean beta activity of the urine of Ailinginae and American groups indicated that the latter had a somewhat lower amount of internal contamination, even though both groups received about the same external dose. This may be accounted for by the fact that the Ailinginae group drank contaminated water from open containers and ate contaminated food up to the time of evacuation, whereas the Americans ingested much less contaminated food and water, since both were largely stored in closed containers. Indoctrination of the Americans 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 5.3 and 5.4). 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 air-borne 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 the Rongelap and Ailinginae groups according to age (Tables 5.3 and 5.4). While 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 hours by children under 15 years was significantly lower than that excreted by adults. The data available 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 radio-elements by growing bone.

Table 5.3.—Gross Beta Activity in Urine of Rongelap People on 46th Day Post Detonation

CASE NO.	TOTAL VOLUME 24 HRS (ML)	BETA ACTIVITY D/M/24 HRS	CASE NO.	TOTAL VOLUME 24 HRS (ML)	BETA ACTIVITY D/M/24 HRS
Age < 5 yrs			Age > 16 yrs		
2	120	712	4	455	634
3	150	894	7	810	1,700
5	155	313	9	355	201
23	40	223	10	980	549
33	260	0	11	450	1,583
54	80	385	13	340	1,677
69	455	301	14	780	2,460
Mean	165	404	18	455	1,670
			22	47	77
			30	960	438
			34	750	570
			37	480	792
			40	550	1,450
			46	330	495
			49	425	0
			52	780	0
			55	320	1,080
			56	700	3,220
			57	550	1,095
			58	750	2,170
			60	810	580
			62	980	1,985
			63	635	2,260
			66	855	1,715
			68	300	2,010
			71	290	1,450
			73	230	0
			78	965	52
			79	465	2,038
			80	540	1,353
			82	670	2,140
			Mean	581	1,208
Age 6-15 yrs					
20	265	1,900			
24	550	0			
26	650	1,032			
35	255	0			
36	190	236			
39	280	1,100			
47	650	1,705			
67	450	674			
72	110	507			
75	440	0			
76	980	1,180			
Mean	439	758			

Values corrected for decay.

No correlation was found between body weight of the people from Rongelap and the total activity per 24 hours excreted in their urine.

Gross beta activity measurements were also made on the samples sent to NYOO, AEC.* Their results essentially corroborate the find-

*Personal communication from Dr. J. Harley, NYOO, AEC.

ings by the USNRDL, 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 USNRDL values by a constant factor.

2. Radiochemical Analysis of the Urine: Estimate of Body Burden. Radiochemical analysis of the Rongelap urine samples indicated that the alkaline earth and rare earth groups together contributed 75 percent of the

Table 5.4.—Gross Beta Activity in Urine of People From Ailinginae and the Americans

AILINGINAE DAY 46 POST DETONATION			AMERICANS DAY 41 POST DETONATION		
CASE No.	TOTAL VOLUME 24 HRS (ML)	BETA ACTIVITY D/M/24 HRS	CASE No.	TOTAL VOLUME 24 HRS (ML)	BETA ACTIVITY D/M/24 HRS
Age < 5 yrs			401	1,970	0
6			2	650	0
8			3	1,224	820
44	150	217	4	440	78
Mean	150	217	5	735	0
Age 6-15 yrs			6	900	248
48	180	164	7	1,340	0
53			8	1,410	1,260
81	370	88	9		
Mean	275	126	10		
Age > 16 yrs			11	1,580	355
1	900	765	12	1,460	0
16	880	827	13	1,810	965
28	680	1202	14	720	438
29	780	0	15	1,380	830
31	260	846	16	1,930	0
41	920	62	17	945	
43	610	754	18	1,520	0
45	850	680	19	1,300	466
51	410	400	20	1,070	0
70	440	0	21	550	353
Mean	722	553	22		
			23	1,180	0
			24	1,160	750
			25	1,380	187
			26	510	323
			27	565	
			28	1,220	0
			Mean	1,158	300

Values corrected for decay.

beta activity at 45 days post detonation (Table 5.5). The predominant radionuclide is Sr⁹⁰, which contributes 42 percent 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 LASL (collected 15 days post detonation) contained fair 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 on Rongelap, 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

Table 5.5.—Radiochemical Analysis of Urine From the Rongelap People (45 days post detonation)

SAMPLE NO.	BETA ACTIVITY—D/M/24 HOUR			
	GROSS BETA ACTIVITY	Sr ⁹⁰	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	325	110	474
6.....	1200	727	170	353
Average.....	1253	526	134	312
Percent of total Beta activity.....	100	42	10.7	25.5

on the radioactive content of various tissues. Details of the animal study are presented in a subsequent section.

The estimate of the mean body burden of the Rongelap group at 82 days post detonation is presented in Table 5.6. The body burden at one day was calculated in the following manner. A formula was obtained from urinary excretion data reported by Cowan, Farabee and Love (6) in a case of accidental inhalation of Sr⁹⁰. The excretion curve was best represented by four exponential terms. (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) (6, 7, 8).

Estimates were made of other radioelements

present in significant amounts at one day, as shown in Table 5.6. These estimates were made on the basis of the level of Sr⁹⁰ at one day, together with the data on the activity of the various fission products at this same time (9) and animal isotope absorption and retention data (1, 5).

The LASL has also estimated the body burden at one day, on the basis of radiochemical analysis of pooled urine samples from a representative number of the Rongelap and American groups (10). These calculations were based on the analysis of I¹³¹ in the early samples of urine (15 days post detonation) as well as the above mentioned physical and biological data on fission products (1, 5, 9). Their findings are presented in Table 5.6.

Table 5.6.—Mean Body Burden of the Rongelap Group

RADIOISOTOPE	ACTIVITY AT 82 DAYS (USNRDL) μC	ACTIVITY AT 1 DAY (USNRDL) μC	ACTIVITY AT 1 DAY (LASL) μC
Sr ⁹⁰	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 (μgm)

On the basis of an assumed uptake of 20 percent per 24 hours, the integrated dose to the thyroid from I^{131} and other shorter-lived iodine isotopes was calculated by the USNRDL to be about 100 rep. The LASL has estimated that this dose was about 150 rep for Rongelap group and 50 rep for the Americans.

The differing approaches used by the USNRDL and the LASL for estimating the body burden gave results which, except for Ba^{140} , are very close.

The mean body burdens of the individual nuclides presented in Table 5.6 were calculated for the Rongelap group. Values for the Ailinginae group were approximately half those of the Rongelap group, and values for Americans, about one-fourth those of the Rongelap group.

The total amount of radioactive material present in the G. I. tract at one day post detonation in the members of Group I 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 post detonation 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 (10). The dose to the thyroid was appreciable, but low compared to the partially or totally ablating doses of I^{131} used in therapy of hyperthyroidism or carcinoma. At one day post detonation Sr^{90} was calculated to be near the maximum permissible level (5) for this nuclide. At later times following exposure, 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 from the lungs and the G. I. tract. Only I, Sr, Ba 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 Sr^{90} , 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.

5.33 Source of Internal Contamination

The fallout material consisted largely of calcium oxide and calcium carbonate. The fission products were adsorbed mainly on fairly large particles. The material was 10 percent soluble in water, and completely soluble in acid.

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 amount of fission products reaching the bloodstream through the respiratory tract is a function of particle size and solubility of the airborne contaminants. The particles with which the activity was associated were considerably larger than the optimum size for deposition in the alveolar tissue of the lung. 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 gastro-intestinal 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 pigs from Rongelap. These animals had about ten times the body burden of the human population in the same locality. As the air-borne 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.

Radioanalysis of water and soil samples from Rongelap indicated high levels of contamination from the fallout at early times following detonation.

It appears that during the first month a limited amount of fission products was available to plants growing on the contaminated soil. Significant amounts of beta activity as well as small amounts of alpha activity were present on the external surface of plants at 42 days post detonation. Only very small amounts of beta activity and no alpha activity were detected in the edible portions of fruits such as pandanus, papayas and coconuts. However, high levels of activity were found in the coconut tree sap, and the isotopic concentration was very similar to that of water.

High levels of activity were found in fish taken from Rongelap lagoon. It appears that the ingestion of contaminated water and fish were the principal sources of internal contamination of human beings. Of the individual radionuclides, Sr⁹⁰, because of its high solubility and relatively long radioactive half-life was probably the isotope of greatest potential hazard in the environment.

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. 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 (11-13). It is most effective with the rare earth group, but has no effect on strontium (13). 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 7 weeks post detonation, since it would mobilize and make detection of isotopes easier, even though it was realized that the procedure would have limited value at this time.

A representative group of seven individuals from Rongelap were selected for this study. During a control period of 5 days, 24-hour urine samples were collected daily for radioanalysis in order to establish a basal excretion rate. During the next 3 days, calcium EDTA was administered orally, 1 gm per 25 lbs of body weight daily instead of the preferable intravenous drip because parenteral therapy was not practical under the circumstances.

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 pre-treatment activity. 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 post detonation increased the excretion rate of internally deposited fission products, but the over-all effect on decreasing the body burden was slight, as the excretion rates were very low at this time.

Summary. The first instance of internal deposition of mixed fission products in humans occurred as a result of fallout following a ther-

monuclear explosion. This internal contamination resulted from both inhalation and ingestion of fallout material.

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. Of the individual radionuclides, Sr⁹⁰, because of its high solubility and relatively long radioactive half-life was probably the isotope of greatest potential hazard in the environment.

Few of the fission products present in the environment were readily absorbed from the lungs and the G. I. tract. Radiochemical analysis of the urine samples from the Rongelap people indicates that Sr, Ba and the rare earth group together constituted 75 percent of the total beta activity of the urine at 45 days post detonation. Sr⁹⁰ was the predominant radionuclide at this time, contributing 42 percent of the total beta activity. Assays for fissile material in the pooled urine samples were 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 Rongelap, and the mean body burdens of the Americans about one-fourth of the Rongelap group. While the activity excreted per unit volume of urine was the same for adults and children from Rongelap, the total activity excreted in the urine in 24 hours by children under 15 years of age was significantly lower than that excreted by the adults.

The total amount of radioactive material in the G. I. tract at one day post detonation was estimated to be 3 mc in people from Rongelap. 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 post detonation was barely detectable in the urine of most of the exposed individuals.

The estimated dose to the thyroid from I¹³¹ and other short-lived iodine isotopes was 100

to 150 rep for Rongelap. Iodine is probably the most hazardous internal radioemitter at early times after exposure. The dose to the thyroid, although greater than tolerance, was low compared to the partially or totally ablating doses of I¹³¹ used in the treatment of hyperthyroidism or carcinoma.

At one day post detonation, the concentration of Sr⁹⁰ 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 post detonation 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 slight, as the excretion rate was very low at this time.

Analysis of the internal contamination indicates that the dose to the tissue of the body was near, but, with exception of the dose to the thyroid, 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 small.

5.4 Internal Contamination of Animals

THE INTERNAL CONTAMINATION of a number of animals collected on Rongelap was studied. The activity in their urine was studied, and radiochemical analyses were made of various tissues. These data provided the basis 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 from Rongelap and Utirik included 41 chickens, 9 baby chicks, 11 swine, 4 ducks and 1 cat. These were shipped alive to the USNRDL. Three fish and one large clam were taken from the Rongelap lagoon. Collection dates and mortality data for these animals are presented in Table 5.7. In addition, a boar, a cat and two chickens were autopsied in the field, and representative tissues were collected.

5.41 Methods

Tissue samples were taken from all animals which died spontaneously or were sacrificed.

Specimens were obtained from the lung, liver, G. I. tract and the skeleton. The samples were ashed at 550° C. in a muffle oven, and the ash made up to volume with 2 N HCl. An aliquot was then dried for beta measurement. The beta activity was determined by means of a thin end-window Geiger-Muller counter. Sr⁹⁰ was used as the basis for the mass absorption correction for the samples, as it was the major radioelement deposited. The correction calculated is an approximation, as mass absorption is a function of the average energy of the sample. Beta activity was measured in total d/m, and this value was converted to μc . "Sr⁹⁰ 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 percent for a Co⁶⁰ standard. The gamma

Table 5.7.—Mortality and External Radiation Dose of Animals From the Living Areas of Rongelap and Utirik

EXTERNAL DOSE (**DAY OF COLLECTION) ANIMALS	SERIES A			SERIES B			SERIES C			SERIES D			TOTAL		
	280 r (DAY 8)			350 r (DAY 25)			340 r (DAY 33)			360 r (DAY 51-53)			TOTAL REC'D	DEAD	SAC'D
	TOTAL REC'D	DEAD	SAC'D	TOTAL REC'D	DEAD	SAC'D	TOTAL REC'D	DEAD	SAC'D	TOTAL REC'D	DEAD	SAC'D			
Hens.....	6	1	1				20	2	2	11	5		37	8	3
		Day 23	Day 23					Day 42	Day 44		Day 51-53				
								Day 43			67 #30				
											74 #39				
											92 #35				
											99 #7				
											130 #24				
Roosters....	1						2	1		1			4	1	
								Day 49							
Chicks.....							9	9					9	9	
Ducks.....							4		1				4		1
									Day 56						
Pigs.....	1		1	7		4				3*			11		5
			Day 45			Day 51-53									
						35 Sow									
						57 #6									
						82 #24									
						82 #25									
Cat.....	1												1		
													66	18	9

*Animals from Utirik; all others from Rongelap (Group IV area animals rec'd 32 r external dose).

**Day Post Detonation.

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activity was obtained in total d/m, and was converted to μC " Co^{60} equivalent."

Samples were analyzed radiochemically for Sr^{90} , Ba^{140} , the rare earth group, I^{131} and fissile material.

For excretion studies, the animals were caged individually, and their excreta collected at 24-hour intervals. The feces and urine of chick-

ens were collected and ashed combined, but were collected and ashed separately for the pigs. Beginning 5 weeks post detonation, the excreta of a representative group of chickens was collected at weekly intervals for a period of 2½ months. Collection of pig excreta was begun at 6 weeks post detonation, and the collection was made at weekly intervals for a 6-week pe-

Table 5.8.—Radiochemical Analysis of Tissues and Urine of Pigs From Rongelap on 82nd Day Post-Detonation

BETA ACTIVITY—D/M/TOTAL SAMPLE				
SAMPLE	GROSS ACTIVITY $\times 10^{-3}$	Sr^{90} $\times 10^{-3}$	Ba^{140} $\times 10^{-3}$	TOTAL RARE EARTH $\times 10^{-3}$
Pig #24 (25.8 kgm)				
Skeleton (total).....	8890	5660	660	1010
Liver.....	31	0.40	0.33	6.4
Colon & 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 #25 (22.7 kgm)				
Skeleton (total).....	8600	5100	530	690
Liver.....	27	0.53	0.20	5.5
Colon & 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 hrs.....	6.2	4.4	0.40	0.54
SUMMARY				
GROSS BETA ACTIVITY		SKELETON	TOTAL BODY	URINE (24 HRN)
Sr^{90}		62.0	58.0	69.0
Ba^{140}		6.8	6.5	7.9
Rare Earth.....		9.7	9.0	10.5
		78.5	73.5	87.4

All values corrected for decay.

riod. Radioanalysis of the excreta was performed in the same manner as that of the tissue samples, described above.

5.42 Findings and Interpretation

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

On the basis of an assumed 12-hour effective fallout time, the animals from Rongelap received an integrated external dose of 280 to 360 r, depending on the date of their collection (see Table 5.7). The pigs from Utirik 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 per hour at 30 days post detonation. This activity was reduced about 75 percent by a washing with water alone.

Radioactivity of Tissues and Excreta. The gross beta activity of the pigs at 82 days post

detonation was about 4 uc. The distribution of activity in the individual tissues is shown in Table 5.8. Over 90 percent 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 percent of the total body burden. The colon contents had the second highest activity for the soft tissues, about 0.24 percent of the total. The alveolar tissue of the lung had an activity less than 0.02 percent of the total activity in the body.

Gross beta and gamma activity of the chickens at 74 days post detonation 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 5.9) was very similar to that in the pig. Most respiratory radio activity was localized in the turbinates, as a result of entrapment of the large particles, which could not penetrate to the alveolar tissue.

The beta activity in the skeleton of chickens at 160 days dropped to 4 percent of the value at 24 days post detonation, while in the same period the gamma activity dropped to 0.2 percent

Table 5.9.—Beta and Gamma Activity of Chickens From Rongelap (μ c \times 10³)

	HEN #1		HEN #2		HEN #39		HEN #36		HEN #35		HEN #7		HEN #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	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA
Tibia.....	7600	3650	8150	4610	133	695	253	215.5	59	41.3	31.3	33.2	8.1	
Skeleton.....	11030	55600	11900	66900	1930	8600	3670*	3120	850*	660	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 (L) and contents.....					0.63	10.0	3.0	6.3	—	14.0	10.7	8.9	0.29	
Intestine (S) and contents.....					1.6	4.0	3.0	—	—	8.4	6.4	—	—	
Pancreas.....					0.16	—	—	—	—	—	—	—	0.75	0
Spleen.....					—	—	1.0	—	—	—	—	—	0.26	—
Kidney.....	198	46			1.17	9.0	9.0	14.2	10.0	14.9	12.4	0.79	0.23	
Lungs (Alveoli).....	17	28	0	26	0.57	4.0	2.0	1.4	4.5	5.6	4.3	16.5	0.63	
Trachea.....					0.24	2.0	1.0	10.7	3.7	0.9	0.2	—	—	
Turbinates.....					3.87	19	22	15.3	7.6	—	—	—	—	

*Calculated using ratio of gamma activity skeleton/tibia.

**Day post detonation.

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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^{90} , the most abundant of the longer-lived elements deposited.

The residual total beta activity found in the two larger fish at 4 months post detonation averaged $2.5 \mu\text{c}$ (Table 5.10). There was, at the same time, about twice as much gamma activity. The fish were collected 56 days post detonation, and the drop in activity between that time

Table 5.10.—Beta and Gamma Activity of Fish From Rongelap Three Months Post Detonation

FISH #1 (802 GM)						
	GROSS ACTIVITY, μc		Ba, Sr AND RARE EARTH TOTAL ACTIVITY (PERCENT)	RADIOCHEMICAL ANALYSIS (PERCENT) IN Ba, Sr AND RARE EARTH FRACTION		
	BETA	GAMMA		Sr ⁹⁰	Ba ¹⁴⁰	RARE EARTH
Head.....	0.568	1.26	9.9	38.3	9.6	52.1
Scales + Fins + 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 #2 (507 GM)		FISH #3 (166 GM)	
	GROSS ACTIVITY, μc		GROSS ACTIVITY, μc		GROSS ACTIVITY, μc	
	BETA	GAMMA	BETA	GAMMA	BETA	GAMMA
Head.....	0.101	0.23	0.045	0.017		
Scales + Fins + 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 #1						
TOTAL BETA ACTIVITY— 6.4×10^4 D/M						
RADIOCHEMICAL ANALYSIS						
	RADIOELEMENT		PERCENT OF TOTAL ACTIVITY			
	Zr ⁹⁵		21.4			
	Ru ^{103, 106}		32.4			
	Other.....		11.4			
	Sr ⁹⁰		0.7			
	Ba ¹⁴⁰		0.7			
	Rare Earths.....		33.4			

Samples collected two months post detonation.

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and the analysis at 4 months represents only radiological decay. Thus, the results are not directly comparable to those obtained from animals which were returned alive, and in which biological turnover as well as radiological decay were 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 analysed, 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 in the lagoon was considerably greater than that of the land animals studied. As fish form a large staple item in the diet of the Marshallese, the high level of contamination is important.

At the end of a 2½-month experimental period, the excretion by the chickens of both beta and gamma activity per 24 hours was 5 percent of the value measured at the start at 37 days post detonation (Fig. 5.1).

Analysis of pig excreta indicated a similar decrease of activity with time. In a 6-week period, the gamma activity excreted per 24 hours decreased to about 2.5 percent of the activity excreted at 44 days post detonation.

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

Radiochemical Analysis of Tissues and Excreta. Radiochemical analysis of pig tissues indicated that 62 percent of the skeletal beta activity was derived from Sr⁹⁰, 7 percent from Ba¹⁴⁰, and 10 percent from the rare earth group at 82 days post detonation (Table 5.8). The radioisotopic composition of the urine at this time was similar to that of the skeleton. The distribution of activity in the body of the pig

may represent the distribution in human beings. The absolute amount of internal contamination in the Rongelap people was, however, only a tenth of that found in the animals.

At 4 months post detonation, the alkaline earths comprised less than 2 percent of the total activity in the clam (Table 5.10). The rare earth group constituted 33 percent of the total beta activity. The balance of the activity was contributed chiefly by Zr⁹⁵ (21 percent) and Ru^{103,106} (32 percent). About 50 percent 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 percent of the total activity.

5.43 Autoradiographs

A number of autoradiographs of the tibiae and femurs of 1 chick, 4 pigs, 1 rooster and 2 chickens were prepared both at the USNRDL and at the Argonne National Laboratory (ANL) to determine 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 (15).

The autoradiograph of a tibia from a chicken sacrificed at 45 days post detonation (Fig. 5.2) 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.

The tibia and femur of a baby chick, which died spontaneously 47 days post detonation, showed the heaviest concentration of radioactive material in the diaphysis (Fig. 5.3). 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 ab-

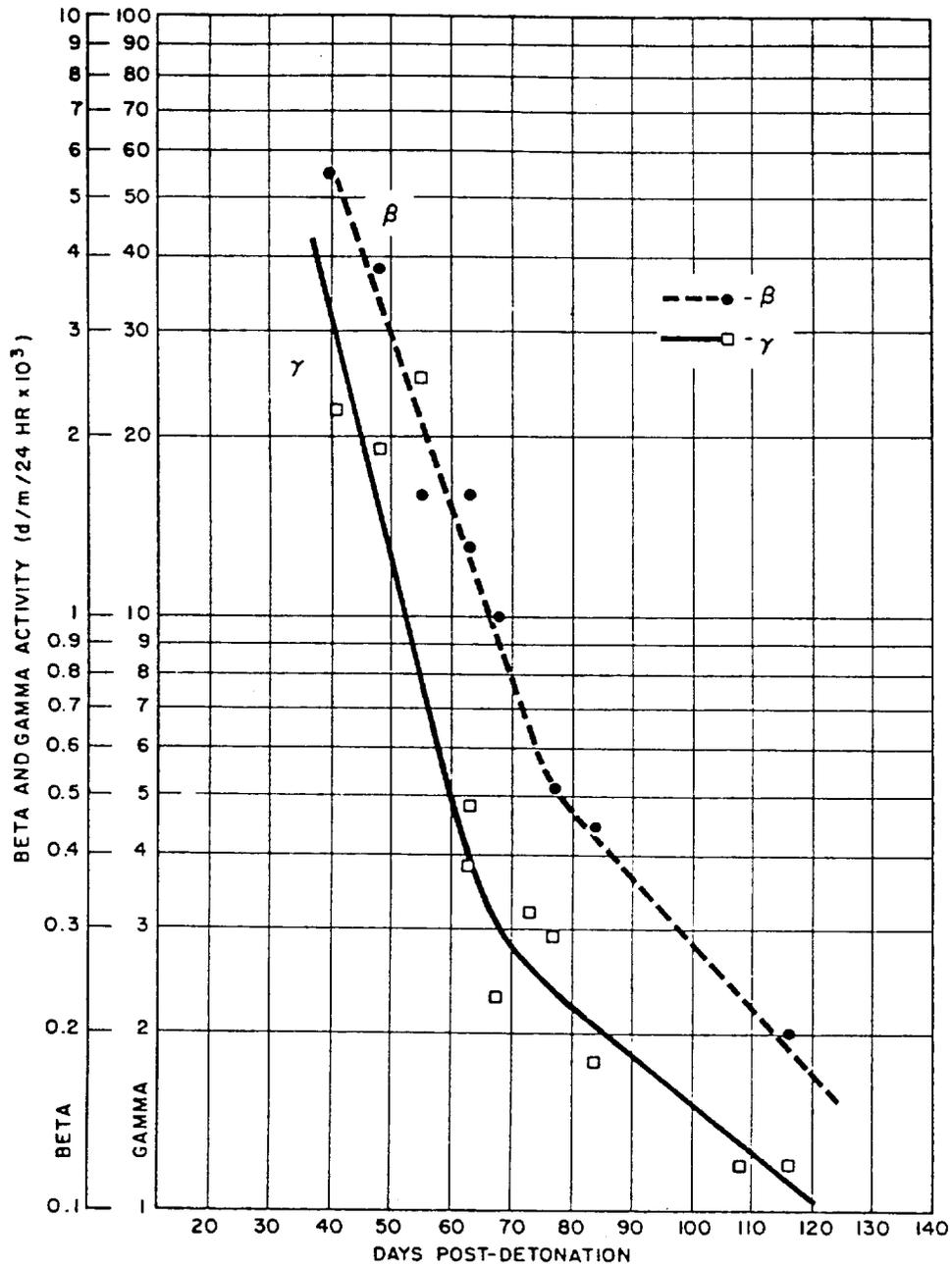


FIGURE 5.1.—Beta and gamma activity in chicken excreta.

normally constricted, possibly because of a decreased rate of endosteal resorption.

A tibia from a pig sacrificed 45 days post detonation had an area under the growing epiphysis

free of activity (Fig. 5.4). As in the chick described above, this area corresponds to the growth which took place after the animal was removed from the area of contamination. The

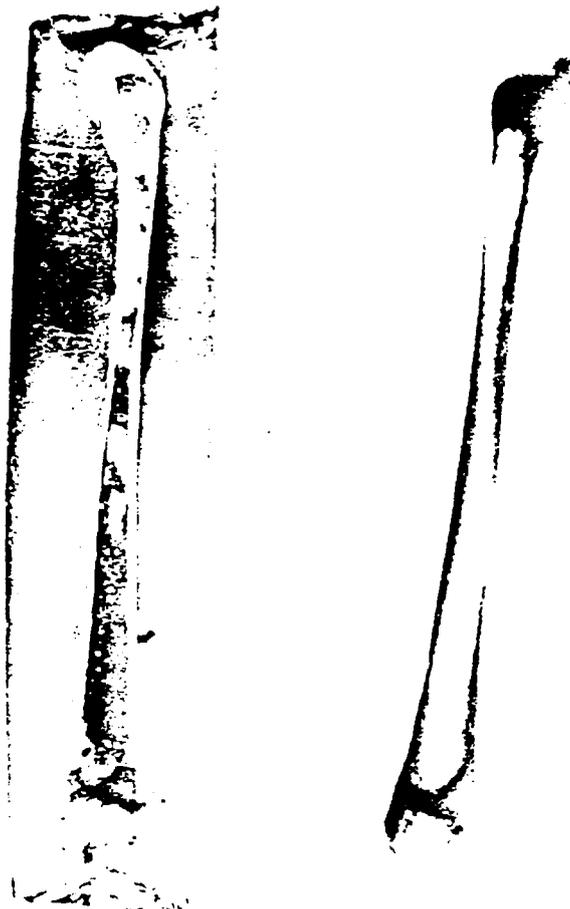


FIGURE 5.2.—Autoradiograph of tibia of chicken sacrificed 45 days post-detonation (ANL).

marrow cavity in this tibia contained dense trabecular bone along its entire length, a formation not normally found in mammalian bones. There are also two distinct areas of increased density in the trabecular region, which appear as two lines of radioactivity in the autoradiograph. The center of the diaphysis was abnormally thick, possibly because of a failure of the normal resorptive process.

No other evidence of a double line of radioactive deposit appeared in the animals studied, except possibly in a sow sacrificed 38 days post exposure (Fig. 5.5). Here a faint deposit of activity in the trabecular bone is noted, separate from the higher level in the epiphysis.

Looney (8) has shown that a typical osseous tissue in trabecular space is a characteristic histopathological finding following radioactive

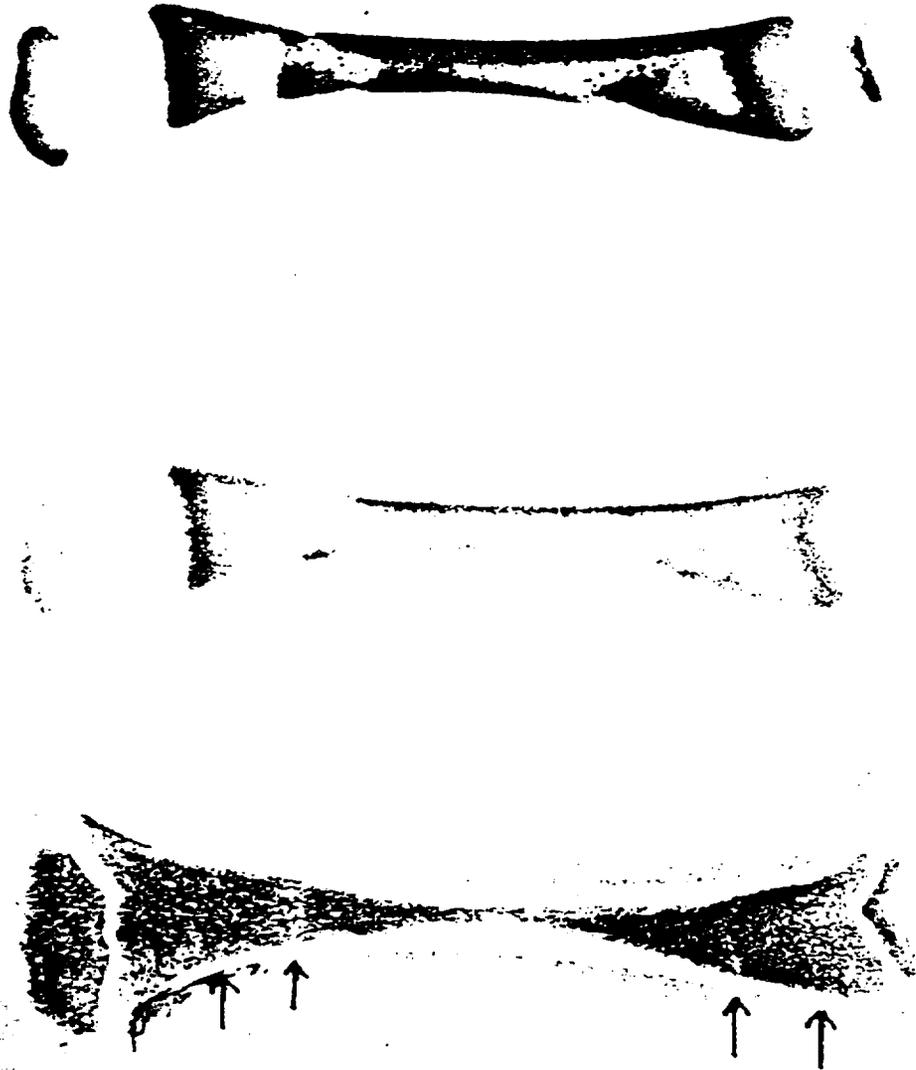


FIGURE 5.4.—Autoradiograph of tibia of pig sacrificed 45 days post-detonation (ANL).

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FIGURE 5.5.—Autoradiograph of tibia of adult sow sacrificed 38 days post-detonation.

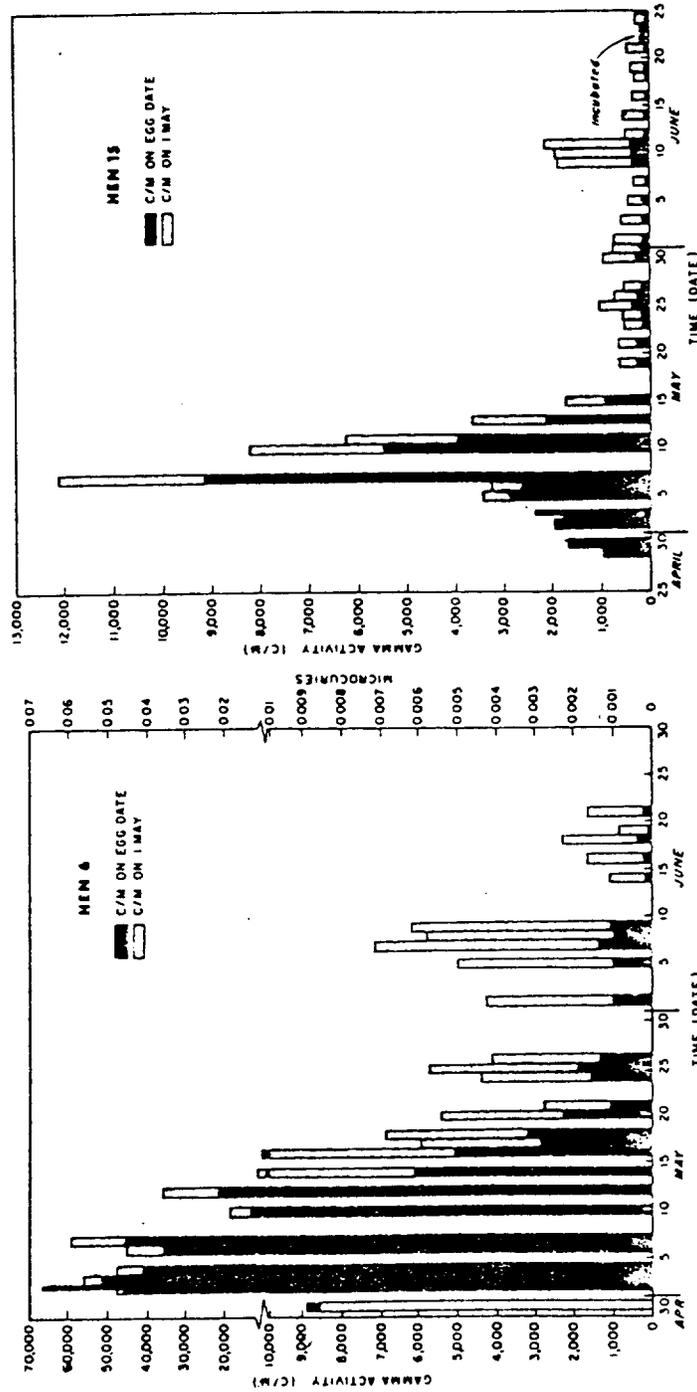


FIGURE 5.6.—Gamma activity in egg shell as a function of time (maximum gamma activity was noted in 8th egg laid as typically illustrated in hen 15. The gamma activity in hen 6 represents the highest concentration of activity seen in any of the shells).

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FIGURE 5.7.—Autoradiograph of chicken eggs showing pattern of deposition of fission products in yolk.

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

5.47 Internal Radioactive Decontamination Studies in Chickens

A study was undertaken to determine the ability of both sodium EDTA and zirconium citrate (15) to increase the excretion rate of internally deposited fission products in the contaminated chickens. On the basis of previous experience, it was not expected that any appreciable decontamination could be effected at the time of this experiment (4 months following internal radioactive deposition).

The excretion rates of 8 chickens with large body burdens of internal contaminants were determined for a period of 4 days as the base line for the study. Following this, two chickens were injected daily I. P. with 75 mg. sodium EDTA for four days; two received injections of 70 mg. of zirconium citrate (15), 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 1 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 post contamination was less than 0.1 percent per 24 hours. Thus, the enhancement of the excretion rate by the combination of zirconium citrate and sodium EDTA did not significantly decrease the total body burden.

5.48 Summary

Studies of animals provided data on the nature and distribution of the radioisotopes in the tissues and the excreta. Over 90 percent 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 external radiation, although the effects of severe dietary changes and other disease cannot be ruled out.

The alkaline earths Sr^{90} and Ba^{140} and the rare earth group together constituted 75 percent of the gross beta activity in the pig at 82 days post detonation. 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 and the eggs produced were both 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.

ENCLOSURE IV

V-D

The total person-years of observation in the 4 study groups (Rongelap exposed, Ailingnae exposed, Utirik exposed, and current unexposed group is 8280.

<u>Rongelap exposed</u>	<u>Ailingnae exposed</u>	<u>Utirik exposed</u>	<u>Unexposed (800-1500 series)</u>
25 years x 60 people = 1500	25 years x 16 people = 400	25 years x 132 people = 3300	22 yrs x 140 people = 3080

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