The Determination of Internally Deposited Radioactive Isotopes in the Marshallese People by Excretion Analysis#

Kent T. Woodward, Ariel G. Schrodt##, James E. Anderson,
Harry A. Claypool, and James B. Hartgering

Division of Nuclear Medicine
Walter Reed Army Institute of Research
Washington 12, D. C.

Work done under the auspices of The Surgeon General, United States Army, and in conjunction with the Division of Biology and Medicine, Atomic Energy Commission

Present Address: Nuclear-Chicago, Chicago, Illinois

INTRODUCTION.

Following the detonation of a thermonuclear device at the Pacific Test Site on 1 March 1954, 239 Marshallese people were exposed to significant levels of gamma radiation from fallout. Estimated total exposures ranged from 175r on Rongelap to 14r on Utirik (1).

These populations were evacuated to Kwajalein for decontamination and care. During the two days of fallout exposure before evacuation was completed, the Marshallese also received some radioactive materials internally by ingestion and inhalation. Estimates of the internal body burden from fallout were obtained from the analysis (1) of urine samples collected soon after exposure.

These data indicated that the acute hazard from internally deposited fission fragments was quite small as compared to the whole body gamma radiation exposure. Although the radioactivity levels in the urine were low, the activity was sufficient to obtain reasonable precision and to warrant additional long term studies of the activity levels and excretion patterns of this rather large and well isolated population.

The people from Alinginae and Utirik were returned to their home islands in June 1954. Radiation intensities on Rongelap, however, precluded an early return to this atoll and the Rongelap people lived on Majuro from June 1954 until July 1957.

Basic data on the food crops of the Marshallese indicated that after resettlement on the contaminated atolls the intake of strontium^{90} would be increased considerably, and that cesium^{137}, zinc^{65}, and cobalt^{60} were dietary constituents of island and ocean foodstuffs, and also would be assimilated (2). The expected increases in the trace amounts of radionuclides in the food supply of a large population would afford an opportunity to investigate the rate of equilibration and the discrimination factors operating between food supply and man. Urinary excretion levels of cesium^{137} and strontium^{90} were measured from March 1954 through March 1958. Zinc^{65} levels were first measured in 1958 samples.

MATERIALS AND METHODS.

One hundred and forty one individual urine samples collected from 24 March 1954 through 7 September 1954 were obtained by the Health and Safety Laboratory, AEC. Urine volumes were small (about 350 ml) and it was necessary to pool samples. This was done according to the age of the subjects and 19 samples of pooled urine were assayed. A 57 liter pooled urine sample from Rongelap was collected and assayed in 1956 (3). Three pooled samples and seven individual samples were assayed in 1957. Thirty individual urine samples were assayed in 1958.

In samples collected in 1954 and 1957 cesium^{137} was scavenged by nickel ferrocyanide (urine made strongly alkaline) and counted in a crystal well counter. A twenty channel gamma-ray spectrum was determined for each sample and the cesium^{137} photo spectrum count rate used. The 1958 samples were assayed directly for Cs^{137}, Zn^{65}, and K^{40} in 2.5 liter plastic containers placed on an 8 x 4 inch (TH activated) sodium iodide crystal. The activity for each radio-isotope was determined by gamma-ray spectral analysis. Sample activities were compared with known radioactive standards (± 5 percent) counted in the same geometry.

Strontium^{90} was precipitated from urine as the carbonate. Yttrium^{90} was separated and identified by its half-life using thin walled gas flow counters.

Urine samples were corrected for radio-active decay to the time of collection.

There is some uncertainty as to the completeness and the duration of time over which samples were collected and therefore twenty-four hour urine volumes are not accurately known. Potassium^{40} excretion, using 360 d/m or 2 gm K/day indicates an average daily volume of about 1180 ml (± 56 percent). It was convenient to use one liter as an average 24 hour urine volume and to express radioassays in micromicrocuries per liter.

RESULTS AND DISCUSSION.

Cesium^{137} Excretion Levels and Body Burden

The urinary excretion levels of cesium^{137} for the years 1954, 1957, and 1958 are shown in Tables 1 - 4. On 24 - 25 March, 1954, the mean excretion level of cesium^{137} for all age groups was 405 uuc per liter. With an excretion rate of 0.46 percent (4) of cesium^{137} body burden per 24 hours, the mean body burden from fallout 24 - 25 days after exposure was $405/4.6 \times 10^{-3} \times 10^{6}$ or 88 muc ($\pm$ 54 percent). This value is about 20 times the average body burden reported by E. C. Anderson, et al (5, 6) for people measured during 1956 - 1957 in the United States. The cesium^{137} urinary excretion levels for the six months following exposure can be expressed as an exponential function, and a best line of fit drawn through the data resulted in a half time for elimination of about 110 days (Fig 1). A biological half time of about 140 days has been observed on volunteers who ingested one microcurie of radio-cesium (4).

From the 1957 Cs^{137} excretion levels (Table 2) the Rongelap group exposed to fallout was estimated to have an average burden of about 7 muc, whereas the Rongelap control group was about 2 muc. Body burden in either group in 1957 is comparable to levels measured in the U. S. population (6). With a half time for elimination of the order of 150 days, the body burden of the exposed Rongelap group should have decreased from the March 1954 level to 7 muc in about 550 days, or late in 1955. A body burden of 7 muc for this group in March 1957 could then indicate a continuing exposure to Cs^{137} during 1956 of the order of 32 micro-microcuries per day from stratospheric-tropospheric fallout while residing on Majuro.

Since the Utirik group was returned to their atoll in 1954, the mean body burden in 1957 was elevated to an estimated 337 muc, some 48 times the Cs^{137} burden of the exposed Rongelap people who resided on Majuro. This long residency time on Utirik atoll after fallout contamination, as compared to the excretion rate of Cs^{137} should have resulted in an equilibrated Cs^{137} burden, with an estimated daily intake of about 1560 uuc of Cs^{137} Unfortunately no systematic survey of foodstuffs grown on these atolls has been reported. Data available, however show that coconut grown on Rongelap contained about 9 uuc Cs^{137} per gram, and arrowroot (Utirik) contained about 8 uuc Cs^{137} per gram. The daily intake of several hundred grams of either staple would be sufficient to account for the 1957 excretion level in the Utirik group.

The Rongelap groups had been resettled for about nine months at the time of the March 1958 medical survey, and urinary excretion levels of Cs^{137} had increased about one hundred fold over 1957 levels. Mean body burden for the two groups at this time was 0.9 uc (± 27 percent) and 1 2 uc (± 47 percent) (Tables 3 and 4) $Cesium^{137}$ body burden may have equilibrated by late 1958 and predicted burdens were about 1.3 and 1.6 uc respectively

$Strontium^{90}$ Excretion Levels and Body Burden

Urinary excretion levels of $strontium^{90}$ are presented in Tables 1, 2, and 3. The $strontium^{90}$ excretion level in 1956 was 0.5 uuc/liter as determined in a pooled sample of 57 liters Figure 2 shows the excretion of Sr^{90} for the three years following fallout exposure. Although there is considerable variation in the data for the various age groups at early times, mean values for all groups plotted suggest that the excretion pattern can be expressed conveniently as the sum of two exponential terms The larger portion of Sr^{90} was excreted with a half time of about 40 days, and a small fraction, 20 percent, was excreted with a half time of about 500 days. This is similar to Cowan's (7) urinary excretion study of an accident case involving inhaled Sr^{90}.

As was noted in the March 1958 Cs^{137} levels, the excretion levels of Sr^{90} were also increased to 3.5-4 0/0.2, or about 20 fold Since Cs^{137} levels increased 4300 - 5?00/34, or about 140 fold, the ratio is about seven in favor of $cesium^{137}$ With the increases in urinary Sr^{90} excretion levels in 1958, it was pertinent to estimate body burden, burden expected at equilibrium, and daily intake of Sr^{90} from these excretion levels.

The metabolic behavior of strontium as outlined in Supplement #6 of the British Journal of Radiology was used to estimate body burden, etc. from urinary excretion levels of strontium^{90} (Appendix). The fraction of strontium absorbed from the gastro-intestinal tract is 0.6 and the biological excretion rate from the total body is 190 days. Of the absorbed fraction, 0.25/0.60, about 42 percent is deposited in bone and the biological half-life is 4000 days. Assuming that the absorbed fraction is excreted entirely in urine, the mean body burden of the exposed Rongelap group in March 1958 was 2 muc (± 52 percent). This is about nine percent of the expected equilibrium value of 23 muc. The estimated burden of strontium^{90} for March 1958 is probably too low and compares with levels measured in stillborn children in the U. S. several years ago (8). The daily intake of strontium^{90} is estimated to be about 15 micromicrocuries or 15 Sunshine Units (assuming a daily calcium intake of one gram).

Dunning (2) reported that the average concentration of strontium^{90} in the Marshallese food supply could be about 360 Sunshine Units, but this would reduce to well under 100 Sunshine Units if the consumption of high Sr^{90} content foods were eliminated. With the elimination of pandanus and land crabs the diet used by Dunning indicated that the intake of strontium^{90} would be 17 Sunshine Units per day. This compares favorably with the estimated intake of about 15 micromicrocuries from excretion analysis.

Zinc^{65} Excretion Levels and Body Burden

In early 1957 Miller (9) detected Zn^{65} in selected residents of Rongelap and Utirik by whole body gamma-ray spectrometry. Body burden ranged from 29.5 to 73.0 muc for the Rongelap residents, and 482 and 229 muc was detected in two subjects from Utirik. The Rongelap subjects were residing on Majuro at this time.

[redacted]

Miller obtained an effective half-time of 110 days for the elimination of Zn^{65}, and for the two subjects from Utirik the urinary to fecal excretion ratio was 1/9.

Assuming the excretion to be entirely exponential and 10 percent of the body burden of Zn^{65} excreted in urine, the March 1958 urinary excretion levels of 174 and 342 micromicrocuries indicate body burden, equilibrium body burden, and daily intake as follows:

RONGELAP

	1954 Exposed Group	Control Group (Unexposed 1954)
Body Burden (March 1958):	280 muc (± 49%)	540 muc (± 90%)
Equilibrated Body Burden:	330 muc	650 muc
Daily Intake:	2100 uuc/day #	4100 uuc/day #
Percent Equilibration:	85.0 percent	83.0 percent

The mean body burden estimated from 1958 excretion analysis for all Rongelap subjects showed a ten-fold increase over the 1957 whole body measurements. This increase correlates with the return of these people to Rongelap atoll from Majuro. Also the 1958 Rongelap Zn^{65} burdens are comparable with the Utirik subjects in 1957, and the Utirik subjects would have been in equilibrium in 1957 (half time of 110 days for the elimination of $zinc^{65}$).

The estimated intake of $zinc^{65}$ (2000 to 4000 uuc per day) can not be accounted for by Zn^{65} activity levels reported in foodstuffs. Although this radio-nuclide reportedly accounts for a large fraction of the total activity in fish, this amounts to only about six uuc per pound of muscle up to 75 uuc per pound of whole fish (2) or at most four percent of the estimated intake.

CONCLUSIONS.

Since resettlement of the Marshallese people on Rongelap atoll in July 1957, the urinary excretion level of $cesium^{137}$ has increased about 140 fold and about

Assuming 100 percent absorption from the GI Tract

20 fold for strontium^{90}. Zinc^{65} was readily detected in samples from the March 1958 medical survey.

The estimated mean body burden at equilibrium for cesium^{137} is about 1.5 microcuries or about 1/6 of the tolerance recommended by the International Commission for Radiological Protection for non-industrial populations. For strontium^{90} the mean body burden of the exposed Rongelap group in March 1958 was estimated to be two millimicrocuries. This is about nine percent of the expected equilibrium value of 23 millimicrocuries. The equilibrated strontium^{90} burden is about 1/5 of tolerance. The estimated mean body burden of zinc^{65} for Rongelap subjects in March 1958 is about 85 percent of the equilibration value of 0.6 microcuries and the equilibration value is 1/70 of tolerance.

BIBLIOGRAPHY

1. Cronkite, E. P., V. P. Bond, C. L. Dunham, etc.
Some Effects of Ionizing Radiation on Human Beings, Atomic Energy Commission, July (1956); WT 923 (1954)

2. Dunning, G. M.
Radioactive Contamination of Certain Areas in the Pacific Ocean from Nuclear Tests, Atomic Energy Commission, August (1957)

3. Conard, R. A., B. Cannon, C. E. Huggins, J. B. Richards, and A. Lowery.
Medical Survey of Marshallese Two Years Post Exposure to Fallout Radiation, Brookhaven National Laboratory, BNL-412 (T-80), September (1956)

4. Woodward, K. T., C. R. Richmond, and W. H. Langham.
Measurements of Retention and Excretion of Radioisotopes of the Alkali Metals by Mice and Rats, Using an Annular Liquid Scintillation Counter, Proc. Health Phys. Soc. 79 (195[illegible])

5. Anderson, E. C., R. L. Schuch, W.R. Fisher, and W. H. Langham.
Radioactivity of People and Foods, Science, 125:1273 (1957)

6. Anderson, E. C.
Radioactivity of People and Milk: 1957, Science, 128:882 (1958)

7. Cowan, F. P., L. D. Farabee, and R. A. Love.
Health Physics and Medical Aspects of a Strontium^{90} Inhalation Accident. Am. J. Roentgenol., Rad. Ther., and Nuc. Med, 67:805 (1952)

8. Libby, W. F.
Current Research Findings on Radioactive Fallout. Proc. Nat. Acad. Sci. (Wash) 42, 945 (1956)

9. Miller, C. E.
Measurements on Some Residents of the Marshall Island, Argonne National Laboratory, Report No. ANL 5755, pg 56 (1957)

TABLE 1

EXCRETION LEVELS OF URINARY CESIUM^{137} AND STRONTIUM^{90}
(uuc per liter) IN THE MARSHALLESE AT VARIOUS TIMES AFTER EXPOSURE IN 1954

DATE OF COLLECTION	AGE GROUPS < 5 yrs		5-16 yrs		16-24 yrs		24-40 yrs		> 40 yrs		MEAN ±S.D.	
	Cs^{137}	Sr^{90}	Cs^{137}	Sr^{90}	Cs^{137}	Sr^{90}	Cs^{137}	Sr^{90}	Cs^{137}	Sr^{90}	Cs^{137}	Sr^{90}
24 March 1954	-	-	889	11.0	294	5.4	372	3.9	258	-	405±218	7.1±2.4
25 March 1954	-	-	-	-	-	-	268	7.7	352	7.7		
17 April 1954	794	16.4	780	-	431	1.7	311	1.2	323	2.3	528±214	5.4±6.5
14 or 31 May 1954	-	-	255	13.4	427	4.2	434	0.9	543	5.5	415±100	6.0±4.6
6 or 7 Sep 1954	-	-	118	2.0	281	1.9	86	0.5	141	0.5	157±73	1.2±0.6

[redacted]

PRIVACY ACT MATERIAL REMOVED

TABLE 2

EXCRETION LEVELS OF URINARY $CESIUM^{137}$ AND $STRONTIUM^{90}$ IN THE MARSHALLESE DURING MARCH 1957

SOURCE	MEAN SAMPLE VOLUME	ACTIVITY (uuc/liter)	
		$CESIUM^{137}$	$STRONTIUM^{90}$
Exposed - Rongelap	4,100 ml	34.	0.2
Controls - Rongelap	3,664 ml	8.	< 0.2
Exposed - Utirik	2,875 ml	1535.	0.2
	TOTAL SAMPLE VOLUME		
#9	5,400 ml	62.	0.5
#20	10,200 ml	168.	0.6
#40	2,700 ml	128.	-
#79	5,400 ml	103.	< 0.2
#82	8,800 ml	120.	< 0.2
#2123	2,700 ml	3,759.	-
#2125	5,400 ml	1,698	< 0.2

PRIVACY ACT MATERIAL REMOVED

[redacted]

PRIVACY ACT MATERIAL REMOVED

TABLE 3

EXCRETION LEVELS OF URINARY CESIUM^{137}, POTASSIUM^{40}, ZINC^{65}, and STRONTIUM^{90} DURING MARCH 1958

1954-EXPOSED GROUP
RONGELAP

SUBJECT	CASE NO	SEX	AGE (1958)	URINE VOL.	ACTIVITY CESIUM^{137}	POTASSIUM^{40}	Cs/K	ZINC^{65}	STRONTIUM^{90}
				(ml)	uuc/l	gm K/l	uuc/gm	uuc/l	uuc/l
	7	M	41	2680	2181	1.0	2203	162	1.6
	9	M	27	5700	1233	0.7	1665	100	3.8
	12	F	23	6745	2924	1.3	2232	264	1.5
	22	F	21	5525	5917	2.5	2357	345	6.0
	26	M	16	5915	4330	1.6	2706	223	2.1
	31	M	36	2580	3393	2.3	1488	238	1.2
	39	F	19	130	13130	--	--	155	NDA
	40	M	34	1740	2275	0.9	2615	148	6.1
	41	M	48	2690	2245	--	--	107	5.3
	66	F	34	2665	2413	1.4	1664	22	3.1
	73	M	22	4125	5584	--	--	147	5.7
	76	M	13	2665	11708	0.3	45031	237	2.8
	79	M	49	1015	3717	2.1	1796	121	2.0

[redacted]

PRIVACY ACT MATERIAL REMOVED

TABLE 4

EXCRETION LEVELS OF URINARY CESIUM^{137}, POTASSIUM^{40}, ZINC^{65}, and STRONTIUM^{90} DURING MARCH 1958

CONTROL GROUP
(UNEXPOSED-1954)
RONGELAP

SUBJECT	CASE NO.	SEX	AGE (1958)	URINE VOL. (ml)	CESIUM^{137} uuc/l	POTASSIUM^{40} gm K/l	Cs/K uuc/gm	ZINC^{65} uuc/l	STRONTIUM^{90} uuc/l
	818	M	7	1880	7674	0.3	24755	99.	6.4
	825	F	16	400	9928	2.8	3546	337.	10.2
	830	M	20	4275	5165	1.9	2662	553.	6.7
	831	M	18	1430	7342	1.4	5063	306	2.7
	836	M	24	585	7023	3.8	1835	881	2.5
	838	M	26	10515	1867	1.1	1652	324.	3.5
	840	M	31	2355	3393	1.4	2456	1262.	4.1
	843	F	33	6490	2068	0.7	3041	75.	1.7
	849	M	39	2640	3880	3.3	1158	948.	5.0
	855	M	60	2655	3176	1.2	2669	120.	2.5
	865	F	25	2125	4624	1.7	2688	319.	4.1
	872	M	14	5275	7736	1.9	3947	195.	1.3
	874	M	10	4650	6141	0.9	6533	131.	3.2
	876	F	20	2155	2909	0.3	9091	163.	3.3
	877	M	20	5245	3311	1.5	2164	233.	2.0
	883	M	46	2615	3366	2.1	1628	271.	2.7
	887	M	13	2630	11733	2.2	5357	398	4.7

PRIVACY ACT MATERIAL REMOVED

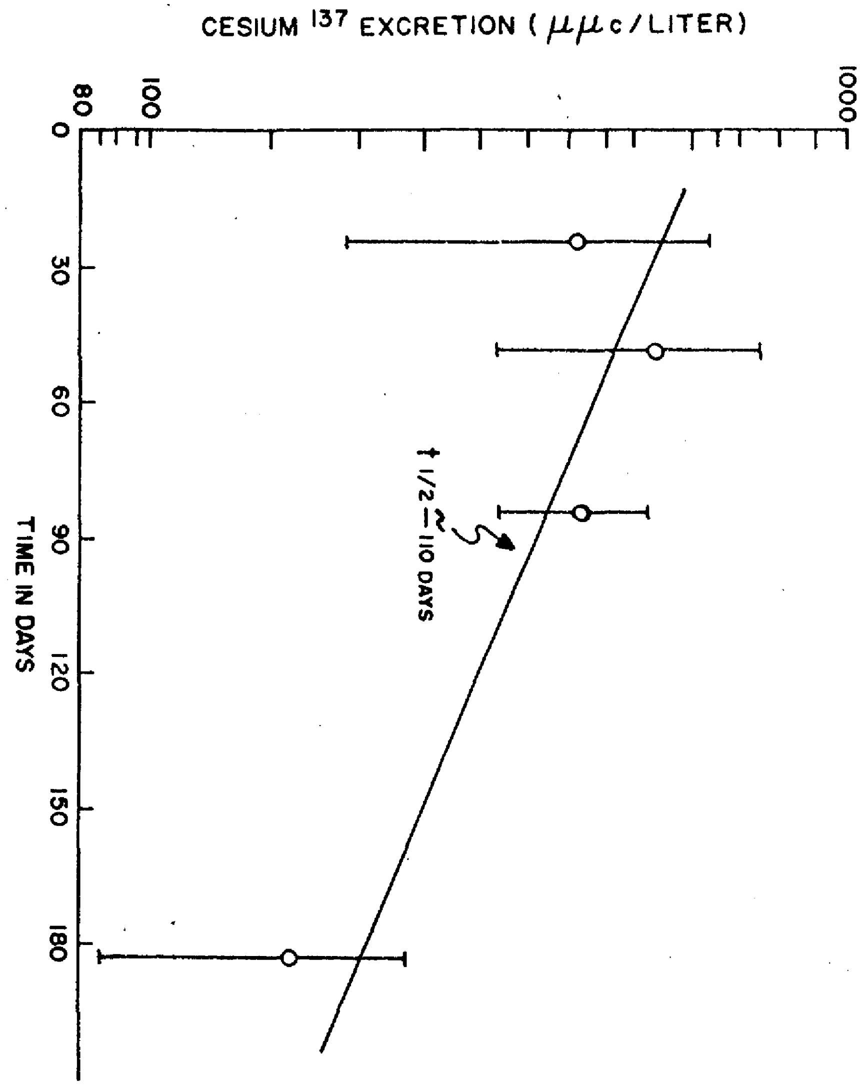

Figure 1. Excretion Levels of Urinary Cesium137 at Various Times After Exposure

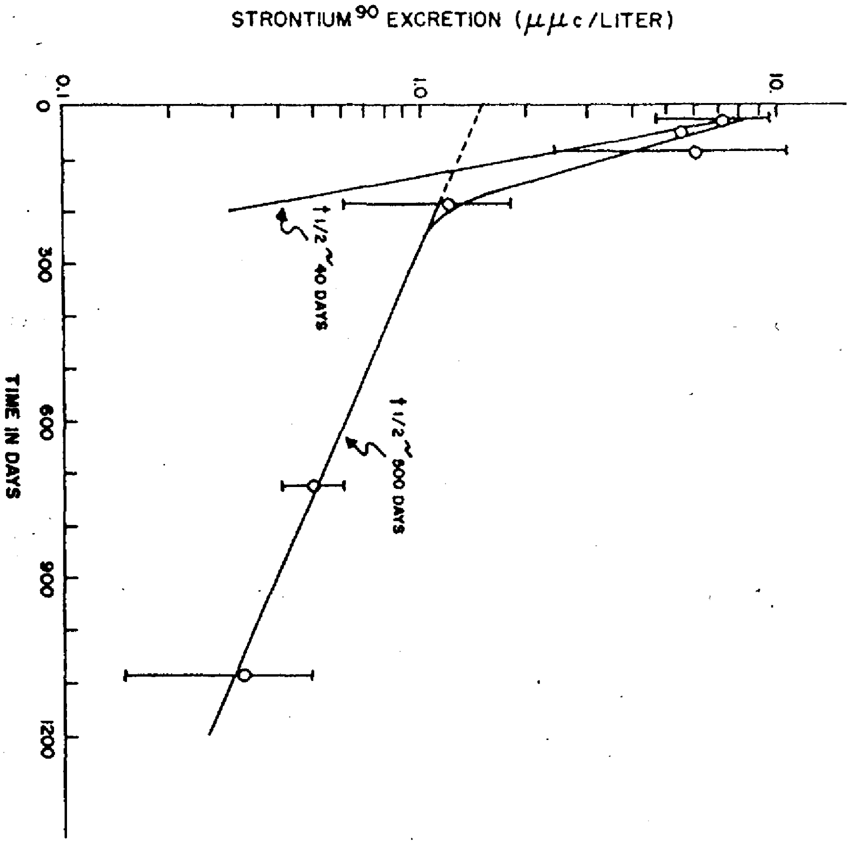

Figure 2. Excretion Levels of Urinary Strontium^{90} at Various Times After Exposure

APPENDIX

I. In the case of strontium elimination, the following assumptions were made:

a. The population was returned to the contaminated atoll at time $t = o$, with a zero strontium body burden.

b. The population absorbs a daily increment of x uc, and x is considered to be a constant independent of time.

c. The amount of strontium excreted in the urine each day is given by $P(t) = \frac{1}{k}E(t)$, where $E(t)$ is the total excreted by all routes each day, and k is a constant independent of time.

d. The body is considered to be a two compartment system, A and B, where $A + B = 1$. The excretion rates for each compartment are a and b $days^{-1}$ respectively. The portion of $E(t)$ excreted from each compartment is proportional to the burden remaining in that compartment. For cesium and zinc elimination similar assumptions are made, except that only one compartment is assumed.

e. Now:

$s(t)$ is total strontium body burden at time t in uc

$s_1(t)$ and $s_2(t)$ are the portions in each compartment

and $s(t) = s_1(t) + s_2(t)$

Considering each compartment seperately and adding the results,

$$\frac{ds}{dt^1} = Ax - E_1(t) = Ax - k\,P_1(t) = Ax - as,$$

hence $s_1(t) = \frac{Ax}{a}(1 - e^{-at})$ and $s_2(t) = \frac{Bx}{b}(1 - e^{-bt})$

Since $E(t) = as_1 + Bs_2$

$$P(t) = \frac{1}{k}(as_1 + bs_2)$$

equilibrium body burden $M = \lim_{t \to \infty} s(t)$

$$s(t) = x\left[\frac{A}{a}(1 - e^{-at}) + \frac{B}{b}(1 - e^{-bt})\right]$$

$$M = x\left(\frac{A}{a} + \frac{B}{b}\right)$$

$$P(t) = \frac{x}{k}\left[A(1 - e^{-at}) + B(1 - e^{-bt})\right] = \frac{x}{k}(1 - Ae^{-at} - Be^{-bt})$$

1. $y = \%$ of equilibrium $= \frac{s(t)}{M} = \frac{\frac{A}{a}(1 - e^{-at}) + \frac{B}{b}(1 - e^{-bt})}{\frac{A}{a} + \frac{B}{b}}$

2. $s(t) = kP(t)\,\frac{\frac{A}{a}(1 - e^{-at}) + \frac{B}{b}(1 - e^{-bt})}{A(1 - e^{-at}) + B(1 - e^{-bt})}$

3. $x = \frac{kP(t)}{A(1 - e^{-at}) + B(1 - e^{-bt})}$

4. $M = kP(t)\,\frac{\frac{A}{a} + \frac{B}{b}}{A(1 - e^{-at}) + B(1 - e^{-bt})}$

f. The following values for strontium metabolism were obtained from Supplement No. 6 of the British Journal of Radiology:

$A = \frac{7}{12}$ $B = \frac{5}{12}$ $k = 1$ and $s_o = o$

$a = 3.65 \times 10^{-3}$ days^{-1} and $b = 1.73 \times 10^{-4}$ days^{-1}, corresponding to a half-time of elimination of 190 and 4000 days respectively.

$x = 0.6\, x'$ and x' is total daily intake.

At $t = 270$ days:

$P(t) = 3.45 \times 10^{-6}$ uc/day (1954 Exposed Rongelap Subjects)

3.9×10^{-6} uc/day (control Rongelap Subjects - Unexposed 1954)

II. **In the case of cesium and zinc:**

$$s(t) = \frac{x}{a}(1 - e^{-at}) + s_o e^{-at}$$

x is the daily accretion in uc/day, and s_o is the body burden in uc at t = o.

$$\frac{ds}{dt} = -as + x = -a\left[s_o e^{-at} + \frac{x}{a}(1 - e^{-at})\right] + x$$

$$= -E(t) + x$$

$$kP(t) = E(t) = a\left[s_o e^{-at} + \frac{x}{a}(1 - e^{-at})\right]$$

$$M = \lim_{t \to \infty} s(t) = \frac{x}{a}$$

Zinc[65]

$a = 6.3 \times 10^{-3}$ days^{-1} (t1/2 = 110 days) k = 10

s_o = 0.03 uc

Cesium[137]

$a = 4.6 \times 10^{-3}$ days^{-1} (t1/2 = 150 days) k = 1#

s_o = o

#The urinary/fecal ratio of radiocesium for human subjects is about 5/1, so that estimates of body burden are too low by about 20 percent.

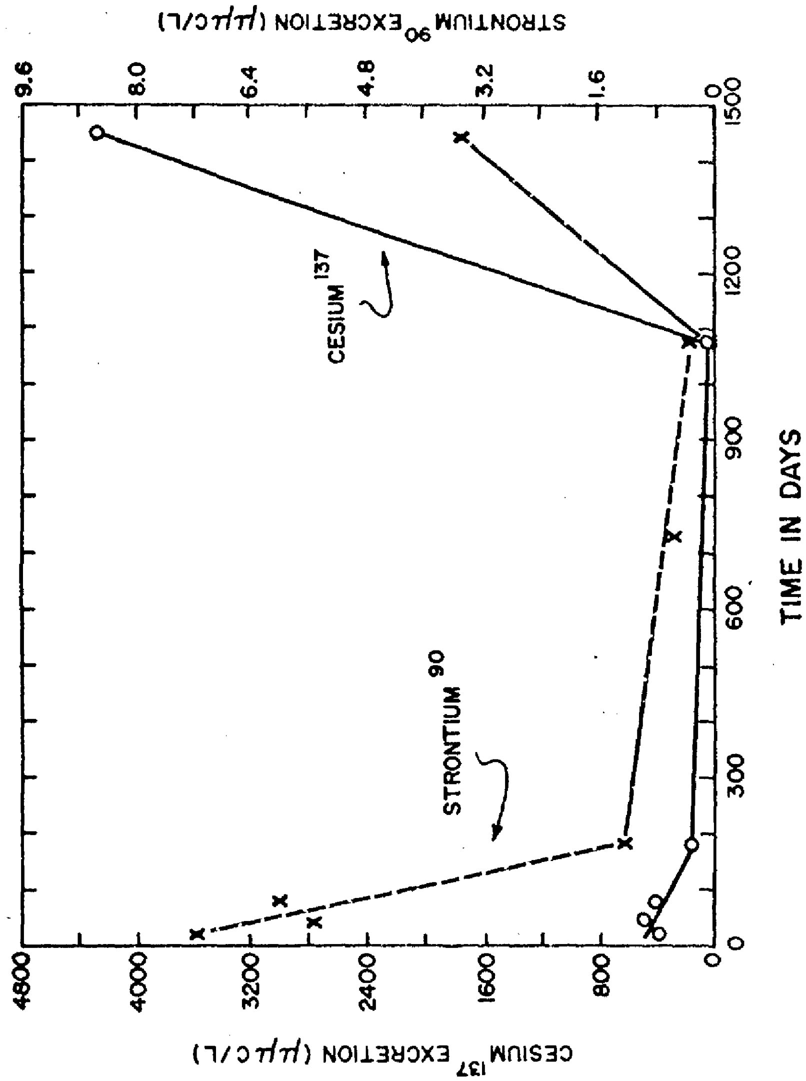
STRONTIUM 90 EXCRETION (μμc/L)
9.6
8.0
6.4
4.8
3.2
1.6
0
CESIUM 137 EXCRETION (μμc/L)
4800
4000
3200
2400
1600
800
0
TIME IN DAYS
0
300
600
900
1200
1500
CESIUM 137
STRONTIUM 90

Figure 3