REPOSITORY ARGONNE NATIONAL LABORATORY

COLLECTION HUMAN HEALTH REDEARCH LOMMITTEE RECORDS

BOX No. ---

727366



FOLDER HEALTH PHYSICS JOURNAL VOL 12 Pp 444-473

dealth Physics Pergamon Press 1966. Vol. 12, pp. 449-473. Printed in Northern Ireland

THE CONSEQUENCES OF INGESTION BY MAN OF REAL AND SIMULATED FALLOUT*

GEORGE V. LEROY, JOHN H. RUST and ROBERT J. HASTERLIK

Departments of Medicine and Pharmacology, The University of Chicago, and the Argonne Cancer Research Hospital, ‡ Chicago, Illinois

(Received 19 April 1965; in revised form 1 July 1965)

Abstract—Real and simulated particulate fallout and solutions of ⁸⁵SrCl₂ and ¹³⁴CsCl were fed to 102 healthy volunteers. Absorption and retention of ingested radioactivity was measured by whole-body counting using the gamma-ray spectrometer at the Argonne Cancer Research Hospital. An average of 3 per cent of the radioactivity of week-old local fallout was absorbed: the range was 0–9 per cent. Strontium and cesium leached or dissolved from simulated fallout behaved in the same way, metabolically, after absorption as they did when the tracer was swallowed in a solution or injected intravenously. The large number of subjects studied provided additional information on the range of variation of intestinal motility, biological availability of strontium, cesium and barium following ingestion of fallout, and retention of the radionuclides of these elements.

449

I. INTRODUCTION

IN 1961 CONCERN about some of the problems of the internal deposit of fission products (FP) following ingestion of local fallout led us to propose studies using real fallout from the Nevada Test Site (NTS); simulated fallout particles that contained ⁸⁵Sr, ¹³³Ba or ¹³⁴Cs; and solutions of ³⁵SrCl₂ and ¹³⁴CsCl. The proposal involved feeding the test materials to healthy adult volunteers and estimating retention and absorption from measurements of their gamma radiation using the whole-body spectrometer (WBS) of the Argonne Cancer Research Hospital. The results reported here are concerned with the transit time of particles in the gastrointestinal tract (Section II); the biological availability of radioactivity following ingestion of real local tailout (Section III); and the extent to which the process of leaching from simulated fallout may modify the metabolic behavior of strontium, cesium and barium (Section IV).

The fallout that we studied and attempted to simulate is designated variously as local, early or close-in. After land-surface detonations in the United States at the NTS such fallout consists of particles ranging in size from less than 1μ in diameter to more than 1 mm. The particles are composed of FP, and soil materials and components of the nuclear device in which radioactivity has been induced by neutrons. Near the point of detonation the particles are large and silicaceous with radioactivity distributed throughout and condensed on the surface. The ratio of biologically available activity to total radioactivity is reported to be small. As one moves away and downwind from the point of detonation the particles become smaller, relatively more of the radioactivity is condensed on their surfaces, and biological availability of the FP is increased. The extent of the distribution of fallout depends on the size of the nuclear explosion, meteorological conditions, and the relation of the fireball to the surface of the earth. With high-yield thermonuclear weapons under average weather conditions local fallout may extend to distances exceeding 500 miles, and the

1190514

1

BEST COPY AVAILABLE

[•] Supplemented in part by funds under Contract OCD-OS-62-214.

 $^{^{-1}}$ An extended version of this study appeared in $^{10}6^{+}$ as Argonne Cancer Research Hospital Report No. 102

[‡] Operated by the University of Chicago for the United States Atomic Energy Commission under Contract AT(11-1)-69.

area contaminated by a single weapon may exceed thousands of square miles.

The principal hazard from this variety of fallout is external exposure to gamma radiation, the intensity of which may exceed 1000 R/hr in some locations at the time when fallout is completed. The risk of internal deposition of FP is real but the magnitude of the risk is uncertain. Fallout particles enter the body by inhalation and ingestion. Because of the relatively large size of the particles, the majority of those inhaled are promptly trapped in mucus and quickly cleared from the respiratory passages either to be expectorated or swallowed. Fallout ingested with food or water (plus that from the respiratory tract) passes through the gastrointestinal system where the particles are exposed to a changing chemical environment in which some fraction of the FP may be dissolved and thus become available for absorption and subsequent deposition in internal organs and tissues.

The factors responsible for absorption of FP determine the biological availability of the radionuclides in fallout. These factors include: (1) the physical and chemical characteristics of the particles, i.e. the leachability and solubility of the nuclides they contain; (2) the absorbability of the chemical compounds formed in the intestinal contents; and (3) the transit time of particles through the gastrointestinal tract. Various estimates have been made of the biological availability in man of local fallout. According to Effects of Nuclear Weapons⁽¹⁾ 10 per cent of FP in local fallout is available, but there are no experimental data to support this estimate. MILLER⁽²⁾ has calculated the potential solubility of the principal nuclides as a function of particle size. In the case of ¹³¹I the estimate ranges from 0.016 (fraction soluble) for particles deposited close to the detonation, to 1.0 for the more distant ones. For 90Sr, theoretical solubility ranges from 0.16 close in, to 1.0 at the outer limits of the distribution.⁽³⁾ The chemical solubility of local fallout collected at NTS after tests of various nuclear devices ranges from nil to more than 50 per cent. Some writers assume that biological availability parallels the chemical solubility in dilute solutions of acids. In some estimates of the hazard from ingested fallout the biological availability is taken as equal to the abundance of a particular radionuclide.

The possibility of obtaining suitable simula for fallout occurred to us after reading a report that described the "radiating microspheres developed by the Minnesota Mining and Man facturing Co. (3 M). These are "ceramic bodies of spherical shape and selected size which contain and immobilize relatively large quantities many useful isotopes". In reply to our inquiry, T. N. LAHR of the 3 M Radiochemical Project said that he could make microspheres in which radionuclide would not be completely immobili lized so that some fraction would leach out in a weak acid solution. In December 1961 two a us (JHR and GVL) met with a group of scientists interested in fallout at Atomic Energy Commission headquarters in Germantown. group (KERMIT LARSON, J. Z. HOLLAND, W.P. LOTZ, H. D. BRUNER, S. H. COHN and J.S. ROBERTSON) agreed that it should be possible to prepare satisfactory simulants with the following properties: the close-in simulant should be glassy or ceramic, and about 10 per cent of the radioactivity should be leachable; the distant simulant should resemble a sintered oxide with a solubility of about 70 per cent; a suitable particle size would be in the range 20-60 μ in diameter. In addition the group agreed that the proposed studies were feasible and worth doing, and that the elements of greatest interest were strontium, cesium and barium.

The radionuclides of these elements (and iodine) in FP are considered critical nuclides because metabolic processes lead to concentration in bone, muscle and the thyroid gland. Once concentrated, each turns over slowly so that the combination of selective deposition, long effective half-life (with the exception of ¹⁸¹) and energetic nuclear radiations can result in a significant internal radiation dose. The estimate of internal dose following ingestion is based on assumptions regarding the fraction absorbed, the fraction of the amount absorbed that goes to the target organ, and the rate of removal from the target organ. For most purposes the values assigned to these assumptions are those given in ICRP Publication No. 2,⁽⁵⁾ and listed in Table Li. Although these are convenient averages the is that absorption, concentration in a target organ, and biological turnover may be pofoundly modified by such factors as the motify of the gastrointestinal tract, the chemical

Element	Average		Ha	alf-life (days	;)	Fraction	Fraction from blood to	Fraction reaching organ of reference	1	
and radio- nuclide	daily intake (g/day)	Organ of reference	Physical (Tr)	Biological (T_{b})	Effective (T)	to blood (f_1)	reference (f_2)	ingestion (f _w)	R/hr-mc at 1 cm	Ē _đ MeV
	10-3	Total body	- <u>-</u>	1.3 × 104		0.3	1.0	0.3		
		Total body	65	1.3×10^4	64.7	0.3	1.0	0.3	3.2	0.014
-Sr		Bone	65	1.8×10^4	64.8	0.3	0.7	0.21		
1	2×10^{-4}	Thyroid		138		1.0	0.3	0.3		
131		Thyroid	8		7.6	1.0	0.3	0.3	2.18	0.19
Cs	Trace	Total body		70		1.0	1.0	1.0		
134Cs		Total body	840	70	65	1.0	1.0	1.0	8.0	0.116
Ba	9×10^{-4}	Total body		65		0.05	1.0	0.05		
133Ba		Total body	2.6×10^{3}	65	63	0.05	1.0	0.05	2.0	0.014
Ba		Bone	2.6×10^3	65	63	0.05	0.7	0.035		

Table I.1 Biological and related physical constants

Source: Table 12, Report of Committee II, ICRP;(5) and HINE and BROWNELL.(5)

compounds formed in the intestinal contents, the route of administration which may regulate the amount that goes to the target organ, and the physiological state of the individual.

There is an extensive literature on fallout,* most of which deals with (1) the external radiation hazard from local fallout; (2) the body burden of critical nuclides in animals collected near a test site; (3) the movement through the biosphere of critical nuclides from local as well as world-wide fallout; and (4) the metabolism of iodine, strontium and cesium following oral and parenteral administration of solutions, and alter accidental contamination. We are not aware of any report of the feeding of real, local fallout to volunteers. To our knowledge this is also the first study where strontium, cesium or barium were administered in a fashion that substated the actual ingestion of particulate fallout. Since it was debatable if there was any advantage to the use of simulants, an important pojective of our study was to compare the behavior of radionuclides leached from the simuant with that of simple solutions of strontium and cesium.

Plan of experiments and methods

The design of the experiments was simple: whole-body counting was used to measure the amount of γ -emitting activity that remained in the bodies of the volunteers at intervals after ingestion of the test materials. The real fallout and the simulants were swallowed in gelatin capsules; the solutions of ¹³⁴CsCl and ⁸⁵SrCl₂ were swallowed with several ounces of water. All test materials were administered without regard to meals or times of day. The identity and the amount of the radionuclide in each dose was determined by one of us (GVL) before administration using an Autogamma y-ray spectrometer. Immediately before the dose was taken, the body background of the subject was measured for 40-60 min in the WBS. At some time during the few hours following the dose-and before the subject voided-a 2-min whole-body count was made. The net counting rate at the time of the first post-dose count was taken as 100 per cent of dose for the purpose of calculating retention at later times. Thereafter, at intervals that varied from a few days to a few weeks serial whole-body counts were made: the counting time was adjusted to the amount of activity remaining so that the probable error of counting was small. Serial counts were continued until: (1) the net counting rate decreased to less than 1.0 per cent of the first post-dose count.

451

10.000

[•] For a concise statement about fallout, see $E_{ij}^{(c)}$ of Nuclear Weapons;⁽¹⁾ for a more detailed discussion, see Fallout and Radiological Counter-Training (2)

(2) sufficient counts were obtained for a reliable estimate of rate of excretion or (3) the volunteer was no longer available. In a few cases samples of stool and urine were collected during the first few weeks after simulants were fed, and these were measured in a properly calibrated largevolume well counter. The information from these measurements was not commensurate with the effort expended and collections of excreta were discontinued early in the study.

The volunteers were healthy adults—University students and members of our staff—who were properly informed of the nature of the experiments and the implications of the study for Civil Defense. None reported any gastrointestinal symptoms following ingestion of any of the test materials. The experiments were approved by the Committee for the Human Use of Radioisotopes of the University of Chicago Hospitals and Clinics. The total number of volunteers was 102.

All measurements of radioactivity retained in the subjects were made with the whole-body gamma-ray spectrometer (WBS) of the Argonne Cancer Research Hospital. This instrument, which has been described in detail elsewhere⁽⁷⁾ consists of four 5 \times 5-in. thallium-activated NaI crystals coupled to photomultiplier tubes arranged above a couch in an iron-shielded room. The output of each crystal detector is transferred through a mixing circuit to a multi-channel pulse height analyzer adjustable for readouts of 3, 6 or 12 keV per channel. The readout is stored on punched paper tape. When the volunteers were counted the four crystals were placed approximately equidistant from each other along the midline of the subject and 18.5 in. above the couch. For one-half the counting time the subject was supine: for the other half prone. Preliminary trials demonstrated that there was only a few per cent difference in the efficiency with which the WBS measured a point source (such as a capsule) inside a subject's body and the same amount of activity distributed throughout the body as an extended source. The practice of counting in the prone and the supine position adequately compensated for any inequality in the anterior-posterior distribution of radioactivity in the course of an experiment.

The Argonne Cancer Research Hospital

instrument is very stable: twenty-two counts of a standard 85 Sr source (four vials taining a total of 0.57 μ c) gave a mean 12,398 \pm 190 net cpm. Variations of the by ground of the iron-shielded room were equi small, and when contamination occurred it cleaned up promptly.

The dead time of our WBS is corrected automatically by a proportional increase in live time of the duty cycle. At counting less than about 500,000 cpm the response of instrument is a linear function of the amoun radioactivity present. Since few of our conexceeded 100,000 cpm no correction for cocidence was necessary. The background counting rate of the in shielded room was measured each day b 40- or 60-min count, and the performance of detectors was checked using a sealed calibra source that contained 2.0 μ g of ²²⁶Ra in equirium with its daughters.* The efficiency of WBS for the various test materials and forenergy ranges used to count them is shown Table I.2.

Processing the data

The fraction of dose retained at any time, **1**, **da** was

$$f_{\rm R}^{(\dagger)} = \frac{\text{net cpm at } t}{\text{net cpm at } t_0} ,$$

where t_0 is the time of the first count after a dose was swallowed.

The fraction of dose absorbed, f_A , was obtained from serial whole body counts made during the second through the sixth week after the dose we given. We assumed that 99+ per cent of gested insoluble material was eliminated from the intestinal tract before the end of the seven day (see Section II).

In the case of ¹³⁴Cs serial values for net concorrected for decay were plotted on a semichart and the *y*-intercept for day 1 (the day affithe dose was given) was obtained by extrapoling a line fitted by eye for the period 1–7 week For ⁸⁵Sr and ¹³³Ba serial counts were plotted.

^{*} Supplied by the National Bureau of Standay

[†] The symbols $f_{\rm R}$ and $f_{\rm s}$ were selected because do not duplicate any of the symbols used in Publication No. 2.⁽⁵⁾

Radionuclide:	⁸⁵ Sr	¹³⁴ Cs	133Ba	²²⁶ Ra*
Photopeak of characteristic radiation (MeV)	0.510	0.605; 0.796	0.302; 0.355	0.610
photons/dist	(1)	(0.85) (0.77)	(0.26) (0.74)	
Energy range over which counts were summed (MeV)	0.378-0.570	0.504-0.900	0.198-0.432	0.096-1.250
Background, room plus average subject (cpm)	340	465	684	
(alibration factor, \ddagger (net cpm/ μ c)	13,200	16,940	6,160	21,025
Efficiency of detector (%) §	0.59	0.96	0.38	0.95

Table 1.2 Response of whole hody spectrometer

• This was a sealed calibrated source containing 2.0 μ g of ²²⁶Ra in equilibrium with its daughters, obtained from the U.S. National Bureau of Standards.

† Source: HINE and BROWNELL.⁽⁶⁾

 \ddagger Net cpm per μ c ingested.

net cps

 \times 100. Photon yield for ¹³⁴Cs was taken as 0.8, for ¹³³Ba as 0.74, and for \S Efficiency = y-photons/sec 26Ra as 1.0.

a log-log chart, and the y-intercept for day 1 was read off the eye-fitted line. The fraction of dose absorbed was

$$f_{\Delta} = \frac{\text{extrapolated net cpm at day l}}{\text{observed net cpm at }t_0} . (I-2)$$

Although it is the convention to assume that biological elimination of most elements follows a simple exponential function, it is well known that many data support the view that the fraction of the body burden excreted per day may vary inversely with time and may be best represented by a power function. Following a single injection of certain bone-seeking radionuclides-such as those of strontium and barium-the body burden can be expressed as

$$R(t) = At^{-n}, \quad t \ge 1 \text{ day}, \quad (I.3)$$

where $R t_{t} =$ fractional retention t days after injection: A = normalized fraction of injected dose retained at the end of unit time; and n = aconstant.

We have chosen not to employ this expression for individual subjects because there is some midication that a power function does not represent precisely the true situation, since the exponent y has been found to vary with time. In addition to this, its metabolic significance remains unexplained. For the sake of simplicity we expressed long term retention of strontium,

1190518

barium and cesium as the per cent of the amount absorbed (f_A) remaining in the body at times longer than 50 days. In the case of cesium where elimination appears to follow a simple exponential function indefinitely, the rate of elimination is reported as the half-time in days.

II. TRANSIT TIME OF PARTICLES THROUGH THE HUMAN GASTRO-INTESTINAL TRACT

It is common knowledge that the time required for some recognizable articles of the diet (corn, for example) to appear in the feces varies from a matter of hours to several days. In fact, the variation in intestinal motility of healthy people is so great that few clinicians will agree on an average time for half or all of a suitable indicator to pass through the gut. For radiological health purposes, most workers use the assumptions proposed for the Standard Man which are given in Table II.1. These were not particularly useful for our purpose since we were less concerned with the radiation dose to portions of the gut than we were about the time at which we could confidently expect that 99 per cent or more of ingested particulate material had been excreted. The Standard Man value-an average of 31 hr from ingestion to excretion-is obviously too short and does not provide any indication of the variability which must occur. We were interested in obtaining a reliable value

453

「ないなながい」という

14 C 14

したいというというできたいとうないで

Portion of interest	Mass of contents (g)	Time food remains (hr)	Suggested revision* (hr)
Stomach	250	1	1
Small intestine	1100	4	4
Upper large intestine	135	8	13
Lower large intestine	150	18	31

Table II.1.	The	gastrointestinal	tract of	f the	standa	rd	man
-------------	-----	------------------	----------	-------	--------	----	-----

• HAYES, CARLTON and BUTLER⁽⁸⁾ studied the elimination of an insoluble tracer and suggested changes the entrance time into the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 13 to 18 hr; and the in-residence time in the lower large intestine from 14 to 31 hr.

Source: Table 11, Report of Committee II, ICRP.⁽⁵⁾

for the 99 per cent excretion time because we could then assume that any radioactivity subsequently remaining represented nuclides released from particles and absorbed from the intestinal tract *minus* the fraction of absorbed activity excreted in urine and feces up to that time. Obviously it was to our advantage to start making serial measurements to estimate the rate of excretion of absorbed material as soon as possible after the dose was administered. In this report, then, we define *transit time* as the time required to excrete more than 99 per cent of a dose of insoluble material.

When we began this study the report of HAYES, CARLTON and BUTLER⁽⁸⁾ had not yet been published. The only guidance available to us was the experience of several groups of investigators with oral doses of ⁵⁹Fe to study iron absorption using whole-body counting. There was general agreement among them that all of the unabsorbed tracer was excreted by 10-14 days. The experiments reported here were designed to find a time between 31 hr and 10 days which would satisfy our requirements. In addition to information about the distribution of transit times we were interested to find out if any significant fraction of insoluble particles was trapped in anatomical features of the gut such as the vermiform appendix. Finally we wanted to know if the stable microspheres were as insoluble in vivo as in vitro.

Materials and methods

Radiating microspheres containing ⁸⁵Sr or ¹³⁴Cs or ¹³³Ba were prepared for us by the Radiochemical Project of the Minnesota Mining and Manufacturing Company to be used as insoluble controls for the simulants of fallout. The microspheres are milky white in appearance and almost perfectly spherical in shape. The have an absolute density of about 3.0 g/cm and a bulk density of 2. They are complete ceramic in nature and have a melting point excess of 1500°C. They are physically insoluin all organic and inorganic solvents excepboiling 57 per cent HF. Soak tests in water and in 0.01 N HCl for 7 days at 50°C demonstrate slight leaching of the nuclide label: on the order of 0.001 to 0.004 per cent.⁽⁴⁾ The specific activity was about 50 μ c/g, and the partice size was in the 30-40 μ range. Three groups of volunteers—fourteen in all were used for a pilot study to see if there we any retention of radioactivity at 10 and 20 day after ingestion of the labeled microspheres. Of the basis of these results, three additional groups (a total of twelve) were fed ¹³⁴Cs-labeled microspheres and counted every 2 days until the amount of activity remaining was less than 01 per cent of the dose. Some of the subjects we asked to keep diaries recording their bound movements: this information was less important than the counting data so the practice we discontinued.

The amount of radioactivity fed—from **abo** 0.5 to 14.0 μ c—was a compromise: on the **c** hand we were obligated to keep the radial dose to the gut to a minimum; and on the **oth** hand we wanted to use sufficient activity so the retention of less than 1.0 per cent could measured with an acceptable probable error.

Results

The counting data for the three pilot studies are shown in Table II.2. In Experiment No. the amount of ⁸⁵Sr microspheres fed way

		Day 0	Day -	+ 10	Day +	11	Day +	- 20	Day + 23
Subject	Tracer	net cpm	Net cpm*	fr†	Net cpm	fu	Net cpm	fR	Net cpm
7-1	⁸⁵ Sr	6886	59	0.0085			46	0.0066	
7_7	85Sr	4688	44	0.0093			38	0.0081	
7-3	85Sr	5475	63	0.0115			68	0.0145	
7.4	85Sr	5827	56	0.0102			-48	0.0087	
7-3	85Sr	4565	78	0.017			54	0.0118	
4.1	133Ba	86,221			18	< 0.001			
9.2	133Ba	141,379			3	< 0.001			nsc‡
9.3	133Ba	132,407			nsc				
9-4	133Ba	123,414			10	< 0.001			
1-1	134Cs	199,835	79	< 0.001			68	< 0.001	
1-2	134Cs	242,053	110	< 0.001			85	< 0.001	
1-3	134Cs	190,692	56	< 0.001			75	< 0.001	
1-4	134Cs	262.418	92	< 0.001			89	< 0.001	
1-5	134Cs	271.314	593	0.021			72	< 0.001	

Table 11.2. Excretion of stable microspheres

Probable error for $f_{\rm R} = 0.01$ of dose: (1) ⁸⁵Sr = 55 net cpm; PE = 4.9 per cent, (2) For ¹³³Ba = 1200 net opm; PE = 0.4 per cent, (3) For ¹³⁴Cs = 2330 net cpm; PE = 0.3 percent. PE = $\frac{67.45}{\sqrt{G}} \left(\frac{\sqrt{K^2 + K}}{K - 1} \right)$, where G = gross counts; and $K = \frac{\text{brow optimized}}{\text{background cpm}}$

Standard error of background rate: (1) For ⁸⁵Sr experiment = 17 cpm; (2) For ¹³³Ba experiment = 26 cpm; (3) For ¹³⁴Cs experiment = 21 cpm. SE = \sqrt{cpm} .

• Net cpm not corrected for decay; $\dagger f_{\rm R}$ = fraction of dose remaining; \ddagger nsc = No significant counts, i.e. equal to or less than background rate.

small that the probable error of the net counting rate 'PE) if 1.0 per cent of the dose remained e.g. about 55 net cpm) was 4.9 per cent. When the net cpm on D + 10 and D + 20 are corrected for decay it appears that the average retention was about 1.5 and 1.0 per cent of dose, respectively. From these data alone it is not possible to decide if the fraction that remained represents microspheres trapped in the gut, or adiostrontium leached from the supposedly instuble microspheres and absorbed from the intestinal contents.

In Experiment No. 9 when larger amounts of ingla microspheres were fed, the net counting ^{14:es} on D + 11 and D + 23 were distinctly f than the standard error (SE = \sqrt{cpm}) of the background counting rate: 26 cpm. In this case we concluded that no microspheres "mained in the gut, and that no 133Ba had been cached from the particles and absorbed.

With the 134Cs microspheres (Experiment No. 1, the fraction of activity remaining on D and D + 20 was less than in the case of

⁸⁵Sr but may be significant. The PE of these net counting rates was less than 5 per cent, and in every case the observed net cpm was several times the SE of the background counting rate: 25 cpm. Since it is unlikely that any microspheres remained in the gut as long as 20 days, we concluded that some radiocesium-less than 0.2 per cent-had leached from the particles and been absorbed. The net counting rates were so low, however, that it was not feasible to attempt an estimate of the fraction of leached material that was absorbed.

The pilot study was satisfactory because the results indicated that the upper limit of the distribution of transit times was less than 10 days.

The results of three additional experiments with ¹³⁴Cs microspheres are given in Table II.3. Here the findings are expressed as per cent of dose remaining on Days +2, +4, +6, +8 and +10. The data indicate that on the average about 60 per cent of the particles were excreted during the first 48 hr following ingestion. The range, however, was quite large: from 2 to 100

		Per cent of	dose remaini	ng on day	
Subject	2	4	6	8	10
1	54	< 0.1	< 0.1		
2	67	< 0.1	< 0.1		
3	69	2.5	< 0.1	< 0.1	
4	2	0.2	<0.1	< 0.1	
5	8	0.3	< 0.1	< 0.1	
6	8	< 0.1	< 0.1		
7	12	0.1	< 0.1	< 0.1	
8	20	< 0.1	< 0.1		
9	53	2.5	< 0.1	< 0.1	
10	9	9.0	< 0.1	< 0.1	
11	100	83.0	26.0	6.0	0.2
12	94	12.0			0.9
Average*	41	9.0	2.5	0.6	0.2
Average,					
without No. 11	36	2.5	< 0.1	< 0.1	<0.1
Median	20	0.3	<0.1	<0.1	<0.1

Table II.3. Excretion of ¹³⁴Cs microspheres

• To calculate averages, <0.1 = 0.1.

per cent remained. By the sixth day less than 0.1 per cent of the dose was still retained by ten of twelve subjects. Combining the results of these three experiments with those of Experiment No. 1 (Table II.2) we see that by the tenth day only three of seventeen subjects retained more than 0.1 per cent of the dose of ¹³⁴Cs. The actual values for these were 202, 593 and 895 net cpm, respectively, i.e. 0.19, 0.22 and 0.95 per cent of the dose.

Discussion

Our findings indicated that we could begin serial measurements in the WBS to evaluate absorption as early as the sixth day after the dose was swallowed if we were willing to accept a small amount of uncertainty because of individual variations. By waiting a day or two longer the likelihood became substantially less that any particulate material remained in the gut. Our results suggest that the Standard Man assumption of 31 hr as the average time that ingested material remains in the intestinal tract (Table II.1) is too short, thus leading to underestimates of radiation dose to the intestinal mucosa from ingested radioactivity. There does not appear to be any simple way to use the data obtained by whole-body counting to adjust the Standard Man assumptions.

It is interesting to compare the findings HAYES et al.⁽⁸⁾ with those presented here. The fed an insoluble tracer, 140La citrate, to fifty-io patients and measured the radioactivity in ea stool as it was passed. For each subject at le 90 per cent of the dose was recovered ultimately A graph from their report showing cumulative excretion as a function of time after ingestion redrawn as Fig. II.1. On the same figure the is a comparable curve showing the average fraction of dose remaining in our subject HAYES et al.⁽⁸⁾ state that for radioactive materi with a half-life longer than about 12 hr Standard Man assumptions require revision An average residence time of 49 hr fits their dat better than the published value: 31 hr. Read ing the points off their curve (Fig. II.1) appears that about 60 per cent of the activit was excreted during the first day; and the time for 98 to 99 per cent excretion was about 4 day It is evident that bowel motility was somewing more rapid in their patient-subjects than in healthy active volunteers. Perhaps bowel had are more regular in hospital than elsewher Actually the difference in intestinal motility, the two groups is not very important. Had used their data to determine the time when than 1 per cent of the dose of insoluble mater remained in the gut we would have selected

「「「「「「「「「」」」」

いたち ちょうかい

456



FIG. II.1. Transit time of particles in the human intestinal tract. The upper curve is the cumulative excretion of ¹⁴⁰La citrate by hospitalized subjects whose median age was less than 53 years. This curve is redrawn from HAYES, CARLTON and BUTLER.⁽⁸⁾ The lower curve is the average for twelve of our subjects who received ¹³⁴Csmicrospheres (Table II.3).

fifth or sixth day. On the basis of the results presented here we waited about 48 hr longer before we assumed that excretion of insoluble particles was complete—that is, that less than ! per cent of the dose remains.

The 3 M stable radiating microspheres were described by the manufacturer as insoluble. Some leaching occurred in vitro, however, when tracer-labeled microspheres were submitted to a soak test. In the case of 137Cs and 90Sr microspheres, for example, the manufacturer reported that the leach rate was the same in water and in dilute HCl. In prolonged tests "leaching contimites at a constant rate after 7-14 days, and is independent of sample size and solvent volume within reasonable limits."(4) Our data suggested that more leaching occurred in the intestinal contents of man than in vitro. After 7 days it appeared that about 1 per cent of 85Sr had been retained. In other experiments (see Section IV) when a dilute aqueous solution of 85SrCl₂ was ed the fraction absorbed was about 17 per cent. Such being the case it is proper to assume that as much as 4 or 5 per cent of the radiostrontium cached out of the "stable" ⁸⁵Sr microspheres during their residence in the gut. This is much

1190522

more than the 0.001 per cent reported for soak tests using ⁹⁰Sr microspheres prepared in the same fashion as the ones we used. Our own soak tests (Table IV.1) were performed with 0.1 N HCl and 0.1 N NaOH at 37°C for 2 hr. No measurable activity was leached from any of the stable microspheres. The procedure used could detect at least 0.001 per cent of the activity present. We have no information about the physiological mechanisms that are responsible for the discrepancy between the *in vivo* and the *in vitro* results.

When solutions of ¹³⁴CsCl were fed (see Section IV) about 90 per cent of the tracer was absorbed. The amount of radiocesium leached from the ¹³⁴Cs microspheres and absorbed ranged from about 0.03 to 0.2 per cent. This is significantly more than the 0.004 per cent leached *in vitro* reported by the manufacturer for ¹³⁷Cs microspheres manufactured in the same fashion as those supplied to us.

The radiation dose to the lower large intestine was calculated using 31 hr as the average inresidence time. The largest dose was 696 mrad, and the average was less than one-half that amount. The estimates of radiation dose for all experiments are given in Table II.4.

III. THE AVAILABILITY OF RADIO-NUCLIDES IN LOCAL FALLOUT

The biological availability for man of the radioactivity contained in local fallout is not well documented. The factors that determine availability include: (1) the physical and chemical characteristics of the particles, i.e. the leachability and solubility of the nuclides they contain; (2) the absorbability of the chemical compounds formed in the intestinal contents; and (3) the transit time of particles through the gut. It is customary to assume that 10 per cent of the activity of local fallout that is swallowed is available. Measurements of the fraction of activity leached by acid, alkaline and neutral solutions have been reported that range from none to more than 50 per cent. Such data display wide variations depending on particle-size, solvent, age of the material, duration of contact with the solution, and conditions associated with the nuclear explosion. The present study was undertaken to measure the biological availability of activity in local fallout collected at a single

457

					Estim	ates of ra	adiation dose,	mrads		
	Number of subjects	Amount administered (µc)		To mu large	To mucosa of lower large intestine*		To whole bodyt		ne and t arrow†	
Test material		subjects	subjects	Minimum	Maximum	Per μc	Maximum‡	Per µc	Maximum‡	Per μc
Real fallout	10	0.8	2.3							
Strontium-85				11.7	20	13.9	10.4	36.4	19	
Microspheres	5	0.4	0.6							
Local simulant	13	0.9	1.5							
Distant simulant	9	0.7	1.3							
*SrCl.	18	0.9	2.5							
Cesium-134				49.7	696	42.8	165			
Microspheres	18	5.5	14.0							
Local simulant	ŝ	7.0	9.0							
Distant simulant	5	2.4	4.7							
134CsCl	4	0.5	0.6							
Barium-133				9.3	214	10.3	11	28.6	21	
Microspheres	4	13.5	23.0							
Local simulant	3	4.0	6.0							
Distant simulant	8	4.0	7.0							
Total	102									

Table II.4. Amounts administered and estimates of radiation dose

* Mucosa dose: $d(\beta + \gamma)/hr = 0.5 C (2.13 \tilde{E}_{\beta} + 10^{-3} \tilde{g}\tau)$ rad, using 31 hr for residence time. \dagger Whole body to bone + bone marrow: $D_{\beta} + \gamma = CT (73.8 \tilde{E}_{\beta} + 0.0346 \tilde{g}\tau)$ rad, using T from Table I.2. These formulae are frequence $tal.^{(9)}$ ‡ Factors used to estimate maximum radiation dose to any subject, from Table I.2 or experiment data, whichever was larger: ³³Sr, whole body = $0.3 \times 2.5 \times 13.9$; bone = $0.21 \times 2.5 \times 36.4$; LLI = $0.7 \times 2.5 \times 11.7$; ¹³⁴Cs, whole body = $0.82 \times 4.7 \times 42.8$; LLI = 14×49.7 ; ¹³³Ba, whole body = $0.15 \times 7 \times 10^{-3}$ bone = $0.105 \times 7 \times 28.6$; LLI = 23×9.3 .

location following the land-surface detonation of a nuclear device: SMALL BOY, on 14 July 1962 at the Nevada Test Site of the United States Atomic Energy Commission.

Material and methods

1190523

The samples of fallout were sent to us by CARL F. MILLER (Stanford Research Institute) with the approval of the Office of Civil Defense. The material we received on D + 5 had the characteristics shown in Table III.1A. For the feeding experiments only samples No. 4, 5, 6, 7 and 8 were used. According to MILLER the fallout was collected at a distance of 4500 ft (1390 m) east of the site of detonation. The radiation intensity of the fallout field at that location, corrected to H + 1 hr was 34 R/hr. There were no measurements of the solubility of the samples we received, but data are available for other collections at nearby stations. These are also given in Table III.1B.

Portions of each sample were placed in soft gelatin capsules (dissolving time in stomach less than 10 min) to be swallowed by the volunteers, or used to estimate the decay correction factor for the sample. At the time of ingestion (D + D +7, or D + 9) the gross gamma activity each dose was equivalent to about 10⁵ disintegrations/sec. This amount of activity was selected because: (1) we assumed acid-solubility we about 10 per cent; (2) we assumed the factor for the decay rate was approximately $t^{-1.2}$; (2) the beta:gamma ratio was taken to be unity (4) we did not want the radiation dose to the lower large intestine to exceed 100 mrad;* and (5) we wanted to give enough activity to provide reliable counting rates during the third and fourth weeks after ingestion. 「山田市村ののためをないい」の東京市内を

For these studies the output of the WBS we summed for the energy range 0.096 to 1.25 MeV. In this range the efficiency of the WBS 0.95 per cent for ²²⁶Ra (see Table I.2), and presumably is the same for FP. When the subjects were counted the crystal arrangement ve

• To approximate this dose we followed DUNNING who calculated that $48 \mu c$ of 1-day old fallout wo deliver about 1.0 rad to the lower large intestine.

	A. Characteristics o	f the fallout samples	*
Size-sample number	Size (µ)	Mass (g)	Activity, as per cent of total
1	2830 +		
2	2830-1410	0.0528	14.7
3	1410-710	0.2016	22.4
4	710-350	0.2051	26.2
5	350-177	0.0674	23.9
6	177-88	0.0630	4.9
7	88-44	0.2356	2.8
8	++-0	0.3880	5.1
	B. Solubility of co	omparable samples†	
		Time of contact	
	Age of material	with acid‡	Per cent
Mesh size	(days)	(days)	leached§
+24	5.9	10.1	8.5
+42	5.9	10.1	3.8
+ 80	5.9	10.1	19.0
+170	5.9	10.1	42.0

T	able	Π	I.	1

• Description of Collection Number 100 PC 5 (SIS PC 5), supplied by C. F. MILLER.

† This is another collection from the same location as 100 PC 5.

‡ HCl solution at pH 1.0.

§ 7-activity only.

Another sample from the same location, was tested when 1.1 days old: acid-solubility ranged from 20 to 58 per cent.

as described in Section I. When the samples were counted the crystals were arrayed in a cluster directly over the source. Correction factors for radioactive decay were obtained by counting each sample at frequent intervals between D + 11 and D + 39. These measurements, corrected for background, are shown as a log-log plot in Fig. III.1. The curves which are linear, were fitted by eye and extrapolated back to the time the fallout samples were fed. A decay correction factor for each subject was obtained in the following manner: If the sample was fed on D + 6 the corresponding value for net cpm was read off Fig. III.1 (see the arrows on Curve No. 7) at the midpoint between 6 and 7 days (315,000), and the 14-day value was read off at the midpoint between 20 and 21 (56,000). The factor used in the case of LO was (315,000/66,000) 4.8. Individual decay factors were calculated for each sample and for the time period involved in each experiment.

Results

Net counting rates for the first count after ingestion of the samples are shown in Table III.2. The gamma-ray spectra for four of the five samples used are shown in Fig. III.2 (a satisfactory curve for Sample No. 4—the largest sized particles—is not available). There are minor differences among these spectra which may be due to variations in nuclide composition associated with particle size. The spectrum for Sample No. 6 has a prominent peak at about 1.2 MeV (yttrium-91,?) which is not evident in the others. Additional evidence for variation in composition of the different sized samples is scen in Fig. III.1, the graphic representation of the decay rates.

The results of the measurements of all ten subjects are summarized in Table III.2. The values for $f_{\rm R}$ —the fraction of dose retained on the last day—represent the amount of activity absorbed from the gut minus the fraction of



FIG. III.1. Decay rate of samples of real fallout.

been subtracted from any of these spectra,

45

.

Subject	<u> </u>	Fir	st count	Las	st count	Deserve	Connected		
	(No.)	Day	Net cpm	Day	Net cpm	factor	net cpm	$f_{\mathbf{R}}$	f_{\perp}
	4	<u>– 6</u>	26,400	+20	210	6.2	1302	0.049	0.06
	- 4	+7	18.860	+21	120	4.9	588	0.031	0.04
	5	-9	26.140	+22	94	3.2	301	0.012	0.01
	5	÷6	27,700	+22	11	5.4	59	0.002	0.00
	6	-6	31.800	+20	114	4.7	536	0.017	0.02
	6	÷-6	24.350	+20	50	4.7	235	0.010	0.0
	7	÷9	38,950	+23	nsc	4.8			
	7	÷6	19,960	+20	205	4.8	984	0.049	0.0
	8	+6	29.470	+20	348	5.5	1914	0.065	0.0
	8	+7	49.220	+21	nsc	5.0			
	-						Mean:	0.024	0.0

Table III.2. Retention of radioactivity from real fallout

PRIVACY ACT MATERIAL REMOVED

1190525

absorbed material that was excreted up to the time of the final count. The values for $f_{\rm A}$ —the fraction of dose absorbed-were estimated from $f_{\rm R}$ multiplied by an arbitrary constant, c = 1.35. This constant was derived from our study of simulated fallout that is described in Section IV. Using simulants labeled with 85 Sr or 134 Cs, f_{A} was obtained by extrapolation to Day + 1 of the curve for $f_{\rm R}$ versus time for the period 7-50 days after the simulant was swallowed. The relationsnip: $f_A = c \times f_R$ at 14 days, was determined. For ${}^{s5}Sr, c = 1.56$; and for ${}^{134}Cs, c = 1.13$. We used the average, c = 1.35, to estimate $f_{\rm A}$ for the real fallout. The average amount of dose absorbed, 3.2 per cent ($f_{A} \times 100$), was much less than we expected and even the largest value-8.8 per cent in the case of -was smaller than the amount customarily assumed. and -the final counting In two casesrate for all energies was the same or less than the subject's 60-min background count (nsc) and in these cases we have assumed that no activity was absorbed.

On the last day of the experiment the highest counting rate for all energies was 348 net cpm for on D + 20. The probable error of this rate is 1.5 per cent. The lowest final count—11 net cpm for on D + 22—has a PE of 34 per cent. Several subjects were counted again a few weeks later, but in every case the rate was not significantly different from the background count.

Discussion

1190526

Our findings demonstrate some of the vicissiudes encountered in a study of the consequences ingestion of week-old silicaceous local fallout. These include: (1) variations in the size and me nuclide-content of the particles; (2) rapid fadioactive decay; (3) uncertain correlation between chemical solubility is sitro and biological availability as measured by the fraction of activity absorbed; (4) biological variability in the function of the human gut; and (5) the limitations of whole-body counting. In spite of these difficulties and the attendant uncertainties, the information obtained provides a better basis for evaluation of the relative hazards of ingestion i local fallout than is possible by indirect methods and studies of wild animals.

When the samples were received on D + 5

we had no information about their composition: we only knew that the material was local fallout from land-surface detonation and that the particles had been sorted into several categories of size. Standard references allow one to make an educated guess about the relative abundance of FP at the time the samples were fed. The complex decay pattern of FP mixtures makes analysis of the γ -ray spectrum difficult even under the most favorable circumstances. The spectra obtained with the WBS after ingestion (Fig. III.2) are further complicated by mass absorption effects, Comptonscattering, and uncertain geometry. Under the conditions of our study it was not feasible to analyze rigorously such spectra. Division of the output of the WBS into arbitrarily selected energy ranges was tried and was not particularly helpful. When the final counts of the volunteers were tabulated on the basis of energy range (see Fig. III.2), range B was the only one where there were significant net counting rates for each subject. Since this range includes the principal photopeak of ¹³¹I it is tempting to attribute the average of 31 net cpm to that radionuclide. If we do this we can estimate that most of the ¹³¹I that should have been in these samples of local fallout was available for absorption.*

It appears from the data in Table III.2 that there was no correlation between availability and particle size: in each size-class there was a low value for f_R and a high one. As mentioned above we had no information about the solubility of the samples we received, although fallout collected nearby was found to have solubilities ranging from 3 to 42 per cent in HCl at pH 1.0 (see Table III.1B). It is customary to assume that the average solubility of local fallout is

PRIVACY ACT MATERIAL REMOVED

[•] The amount of activity ingested was about 2.0 μ c: the abundance of ¹³¹I between D + 7 and D + 15 is approximately 0.07. If all the ¹³¹I was absorbed and the fraction retained in the thyroid was 0.3, then there should have been 2.0 × 0.07 × 0.3 = 0.042 μ c (42 nc) in the gland shortly after the dose was swallowed. Assuming an effective half-life of 7 days, on the last day of the study there should have been about 10 nc in the thyroid. If all the efficiency of the WBS was about 0.005, then 31/0.005 = 6000 dpm, or about 3 nc at the time of the final count.

10 per cent, and that biological availability is of the same order. Our findings suggest that availability, on the average, is about 3 per cent of the total γ -emitting activity. Only four of ten subjects retained more than 3 per cent when measured 2 weeks after fallout was ingested, and two had almost none.

The interval of 2 weeks between feeding and the first examination in the WBS to assess absorption was chosen because we believed that some insoluble material might remain in the gut for as long as 10 days. The studies described in Section II were performed after these experiments with real fallout; and they indicated that retention of insoluble particles (30-40 μ in diameter) after the sixth or seventh day was the exception. Our experience with real fallout would have been more satisfactory had counts been made at 8, 10, 12 and 14 days instead of just at 14 days. Unfortunately we did not anticipate that the fraction retained would be as small as it was, so that we planned to obtain a series of measurements between the fourteenth and twenty-eighth day after feeding. For the twenty-eighth day after D + 6 the decay correction factor—using $t^{-1\cdot 2}$ —was about $\times 25$, and we expected to find counting rates during the fourth week on the order of 100 net cpm for all energies. Better data could have been obtained with larger doses, but we believed that we were obligated to limit the exposure of the lower large intestine to about 100 mrad.

IV. STUDIES WITH SIMULATED FALLOUT

Our experiments with simulated fallout were designed to answer the question: Does the fact that radionuclides of strontium, cesium or barium are released slowly into intestinal contents from real or simulated fallout modify their metabolic behavior during and after absorption from the gut? A corollary to this is the related question: Is it really necessary to use simulants to study the metabolism of critical nuclides from ingested fallout? There does not appear to be any good reason to ask these questions in the case of the radioisotopes of iodine since there is abundant evidence that soluble salts of iodine are absorbed rapidly from any portion of the gut. There is also good evidence that the fraction of iodine in the blood

that is trapped by the thyroid gland is not fluenced by the manner in which the element enters the blood. There are, however, sev reasons for asking the questions about strontin cesium and barium: First, there are many in our understanding of intestinal absorption the familiar mineral elements that are classed essential. The situation with respect to stre tium-which also surely applies to cesium barium—was epitomized by J. F. Loutrr⁽¹⁾ 1961: "How strontium . . . (is) absorbed fi the gut is still a matter of considerable debate indeed is the whole subject of intestinal absor tion. What appears to be certain is that only the small intestine which absorbs these ion the rate is greater in the uppermost part of long tract, though the greater mass may absorbed down where the flow is much slow Later in the same lecture he speculated the "the time-course of entry of strontium into circulation may affect its fate". And second although we have no satisfactory data, i reasonable to suppose that the leaching of rac nuclides from silicaceous local fallout in changing chemical environment of the gut relatively slow process. The same is also probably true for the rate at which the sinte oxides of distant fallout dissolve in intest contents. Lacking many facts we will not far wrong if we assume that dilute solutions salts of strontium, cesium and barium are mediately available for absorption after they swallowed, while compounds formed in 1 intestinal contents from radionuclides leach or dissolved from particulate fallout may somewhat less available. In these circumstan it is appropriate to wonder-as LOUTIT did the time-course of absorption does affect fate of critical nuclides. If it can be shown there is indeed no significant difference in rate of elimination of strontium, cesium barium regardless of how they gained entra to the blood, then valid studies of metabolic can be performed using the intravenous 🙀 for administration to avoid the inevitable certainties associated with experiments with the tracer is given by mouth.

Materials and methods

In accordance with the recommendation of our *ad hoc* advisors (see Section

1190527

Radio-chemical Project of the Minnesota Mining and Manufacturing Company prepared two types of simulants with the following characteristics.

Local simulant (LS) was a specially prepared microsphere fabricated from leachable glass that contained tracer amounts of ⁸⁵Sr, ¹³⁴Cs or ¹³³Ba. The material was manufactured so that about 10 per cent of the radioactivity could be leached from it by 0.1 N HCl at 37°C in 2 hr. The activity was approximately 50 μ c/g, and particle size was in the 30–40 μ range. Although not entirely carrier-free, the amount of carrier was small.

Distant simulant (DS) was either strontium oxide + 85 strontium oxide, or cesium silicate + 134 cesium silicate, or barium oxide + 133 barium oxide calcined at about 900–1000°C. The calcined mass was pulverized and particles in the 30-40 μ range were used. The solubility specified was from 50 to 100 per cent in 0.1 N HCl. The specific activity of the DS was about 50 μ c/g at the time of preparation. In contrast to the LS the amount of carrier was relatively large.

Soak tests were performed by adding a few milligrams of simulant to 10 ml of 0.1 N HCl or 0.1 N NaOH in test tubes. The tubes were agitated constantly in a water bath at 37°C for 2 hr. After filtration the filtrates were counted in a well-type scintillation spectrometer (Auto-34mma). The results are shown in Table IV.1.

Solutions of ⁸⁵SrCl₂ and ¹³⁴CsCl were prepared from carrier-free material using water as the diluent. No effort was made to obtain a soluble salt of barium for these studies.

The range of doses administered is shown in Table II.4. The amount of radioactivity fed was a compromise: we wanted to keep the radiation dose to a minimum, and we wanted enough activity absorbed to permit reliable whole-body counting for several hundred days. We assumed that biological availability paralleled solubility and that the particulate nature of the material had no influence on availability. All doses were given without regard to meals or to time of day.

Results

The γ -ray spectra of three subjects who had ingested one or other of the radionuclides are shown in Fig. IV.1. The portions of the spectrum over which counts were summed for measurement of the whole body counting rate are indicated in each case, (See also Table I.2.)

Strontium-85. The solution of ⁸⁵SrCl₂ was fed to twelve subjects; the average per cent of dose absorbed ($f_A \times 100$) was 17 with a range of 8– 34 per cent (see Table IV.2). The rate of elimination of the fraction of dose absorbed was such that the average retention after 2 weeks was 52 per cent with a range of 34–77. At 50 days average retention decreased to 36 per cent (range: 17–65); after 100 days it was 26 per cent (range: 18–30); and after 150 days only an average of 19 per cent remained (range:

	Per cent dissol	leached or ved in:*	Average per cent absorbed by subjects
Simulant	0.1 N HCl	0.1 N NaOH	$(f_{\rm A} \times 100)$
LS- ⁸⁵ Sr [†]	6.0	8.0	16
DS-85Sr‡	72.0	15.0	13
LS-134Cs	1.4	5.0	31.0
DS-134Cs	38.0	12.0	82.0
LS- ¹³³ Ba	5.0	0.7	Nil
DS- ¹³³ Ba	91.0	23.0	6.0

Table IV.1. Availability of radionuclides from simulants on the basis of soak tests

*After soaking for 2 hr at 37°C.

† The soak test results refer to the second batch; the per cent absorbed by subjects refers to the first batch.

[‡] All results apply to the second batch. Soak tests were not performed with the first batch, the average absorption of which was 32 per cent.

and the second second second

いまではないましいいないない ここのあるまちあん ちょうう



100 C 100

「ういいというから」のないであることで

FIG. IV.1. Gamma-ray spectra after ingestion of simulants. The counts for these spectra were summed for 6 keV/channel. The heavy portion of each of the top three indicate the energy range over which counts were summed to measure retention. The bottom spectrum was obtained in February 1963 before the ¹³³Ba simulant was fed (spectrum directly above). The two small peaks in the third spectrum (¹³³Ba) at about 0.600 and 0.780 MeV are thought to be due to a trace of ¹³⁴Cs that contaminated the BaO + ¹³³BaO during manufacture. After about 100 days these peaks were no longer seen.

	Observed	Descrived Extrapolated et cpm net cpm D - 0 on $D + 1$	ć	Appa	$\begin{array}{c} \text{ rent reten} \\ \text{ of } f_{\mathbf{A}} \end{array}$	cent	Duration of test	Net cpt	
Subject	net cpm on D $- 0$		$f_{\mathbf{A}}$	D + 14	D + 50	D + 100	D + 150	(days)	day
13-1	12 700	2500	0.20	44	27			77	123
13-2	13,700	2700	0.20	54	40	18	6	135	46
13_3	11 400	1300	0.11	77	65			76	253
13-3	13,700	4600	0.34	59	43	29	20	135	260
10-1	31,600	3300	0.10	67	33	30	24	151	157
10.2	30,900	4000	0.13	50	34			75	587
10-3	33 300	9800	0.29	41	29	25	22	153	412
10_4	32,600	4200	0.13	43	33	30	24	149	204
20-1	22,600	1900	0.08	34	17			61	159
20-1	21,300	4000	0.18	47	29			61	604
20-2	18,300	2700	0.15	54	41			58	544
Averag	e		0.17	52	36	26	19		

Table IV.2. Retention after oral dose of ${}^{85}\mathrm{SrCl}_2$

464

÷

Subject	Observed net cpm on D – 0	Extrapolated net cpm on D + 1		Apparent retention as per cent of fraction absorbed				Duration	Net cpm
			JA	D + 14	D + 50	D + 100	D + 150	(days)	day
15-1	19,000	4800	0.25	30	22	21		199	136
LS-2	17,000	2400	0.14	52	38	37	29	247	57
18-3	17.000	1800	0.11	43	29	24	21	164	95
15-4	18,000	2100	0.24	52	38	28		242	139
LS-5	12,000	700	0.06	34	19	14		245	1.4
Averag	e		0.16	42	29	25			
lst									
Batch	11.000	2000	0.21	33	21	18		164	144
DS-1	14,000	7200	0.21	20	9	10		166	150
DS-2	15,200	1500	0.30	37	22	17		90	433
DS-3	16,700	4300	0.30	31	18	13		168	140
DS-4 DS-5	13,400	6400	0.20	27	16	11	_	166	183
Averag			0.32	30	17	12		-	
2nd									
Batch									
DS-6	14,100	2000	0.14	53	53	15	_	114	nsc
DS-7	13.000	1700	0.13	71	59	54		145	nsc
DS-8	13,500	2100	0.16	52	38			97	nsc
DS-9	9500	700	0.07	67	70	_		53	219
Averag	ge		0.13	61	55			-	

Table IV.3. Retention of ⁸⁵Sr released from simulants

6-24). The trials were not all of the same duration because of factors beyond our control such as vacations, graduation and dropping out of school. The lowest value for the final total body count in this group was 46 net cpm, not corrected for decay (subject 13.2 on D + 135). The PE of this net counting rate is 5.8 per cent.

The first batch of LS labeled with ⁸⁵Sr was fed to two groups of volunteers but only the first group yielded satisfactory data. As shown in Table IV.3 an average of 16 per cent of the activity was absorbed ($f_A \times 100$) and the range varied by a factor of 4—from 6 to 25 per cent—as was the case when the solution of ⁸⁵SrCl₂ was fed. A second batch of LS was fed to four subjects but the fraction absorbed was no more than a few per cent and was very little different from that found when stable microspheres were fed we Section III). This result was surprising because our soak tests showed that about 6 per (ent of the activity was leached in 0.1 N HCl and 3 per cent in 0.1 N NaOH. The first batch of DS was fed to five subjects (Table IV.3) and an average of 32 per cent of the activity was absorbed, with individual values that ranged from 21 to 47 per cent. When the second batch was fed less activity was absorbed: the average was 13 per cent. The soak tests of the second batch showed that acid solubility was 72 per cent. (The solubility of the first batch of DS was not measured but the specifications called for a solubility in this range.) A statistical test* showed no significance to the difference between the average f_A for the second batch of DS, the second batch of LS, and the

* The test used was the WILCOXON two-sample test⁽¹²⁾ in which the normal variable, K, is determined from the rank order of the individual measurements of two samples: $K = \frac{2R \pm 1 - n(N+1)}{n(N+1)(N-n)/3}$ where R = sum of the ranks of the smaller sample; N = total number of cases; n = number of cases in the smaller sample.

「おい」で、「たけたい」となるない。「ない」で、「いった」「ほうまい」を見ていたが、「ない」」では、「ない」」では、「ない」」では、「ない」」というない。

1190530

,

solution of chloride. When the results with the first batch of DS (average $f_A = 0.32$) were compared with all the others the difference was highly significant: P = 0.004.

After absorption of ⁸⁵Sr from the gut the rate of elimination was evaluated on the basis of per cent retention at 14, 50, 100 and 150 days. In practice, the measured values for activity retained were plotted as net cpm corrected for



FIG. IV.2. Unexpected variations after ⁸⁵Sr and ¹³³Ba. The topmost curve—DS-¹³³Ba—is subject No. 1, Table IV.5, who absorbed the largest amount of barium. The curve for LS-2, Table IV.3 shows what we expected to find on serial counting to measure retention of ⁸⁵Sr. The other three curves demonstrate unexpected variations which—like those in the top curve—could not be attributed to malfunction of equipment or other errors.

decay on log-log paper. The value for f_A expressed in net cpm was entered on the ordinate for Day 1. Net counting rates for Days 14, 50, 100 and 150 were read off the curve and expressed as per cent of the extrapolated value for Day 1. The statistical test showed no significant difference between retention of activity from the chloride solution, LS, or the second batch of DS. These data therefore were combined and plotted as open circles on Fig. IV.4. The average for retention of 85 Sr from the first batch of DS are indicated by closed circles. The range of values within the various groups was so great that any distinction between the sources

was of doubtful significance. Accordingly in median value for per cent retention by all sujects receiving ⁸⁵Sr at 14, 50, 100 and 150 da was plotted as the crosses on Fig. 1V.4. To line fitted by eye to these points intersects abscissa at about 1.5 days. This is a reasonantime for absorption to be complete. The eqution for this line is: $R(t) = 60 t^{-0.2}, t \ge 1$ we This relationship predicts that after 52 we retention of ⁸⁵Sr will be approximately 16 cent.

An unexpected increase or decrease in whole-body counting rate occurred at so time among the serial measurements of sixte of the thirty-five subjects who received ⁸⁴ Some of these are illustrated in Fig. IV.2 wh net counting rates corrected for decay are plotte against time in days on log-log coordinate Initially, we supposed that these abnormal count were caused by malfunction of the equipme and discontinued measurements of those subject When careful review of the operation of the WBS failed to provide an explanation for majority of the abnormal counts the practice terminating experiments where they occur was stopped. The same phenomenon was served when DS-133Ba was used (see below but there were only occasional unexpect variations of the counting rates with any of the ¹³⁴Cs tracers, and none with any of the standa sources used to validate the performance the instrument.

Cesium-134. The solution of ¹³⁴CsCl was for to four volunteers who absorbed $(f_A \times 100)$ 90 90, 89 and 87 per cent respectively. When the LS was fed, absorption was 36, 29 and 29 per cent, respectively, In our soak tests of the LS about 2 per cent of the activity was leached in 0.1 N HCl at 37°C in 2 hr, and 5 per cent in 0.1 N NaOH under the same conditions. With the DS the average absorption for five subjects was 82 per cent. The solubility of the sintered ¹³⁴Cs silicate in our soak tests was 38 per cent in acid and 12 per cent in alkali. In all of the subjects who received ¹³⁴Cs the rate of eliminate tion of the fraction of dose absorbed followed single exponential function for as long as mean urements of $f_{\rm R}$ were made. Table IV.4 gr the values for f_A , and for rate of elimination f_A , and for rate of elimination f_A , and for rate of elimination f_A , f_A , expressed as the half-time in days. The me half-time (T_b) for the twelve subjects was 91

ころうちをいいたのにある、気をなるというとなるので、「ないない」ので、いたのでなってものです。

466

Subject	Tracer used	∫ _A	Measured half-life (days)	Half-life corrected for decay* (days) ^T b	Duration of test (days)
	Cl	0.90	63	70	207
2	Ĉi	0.90	77	97	43
$\overline{\overline{3}}$	Cl	0.89	110	129	173
4	Cl	0.87	96	112	107
5	DS	0.84	63	70	-49
6	DS	0.83	71	80	159
7	DS	0.82	69	78	88
8	DS	0.80	92	107	159
9	DS	0.73	71	80	159
10	LS	0.36	79	90	107
11	LS	0.29	74	84	105
12	LS	0.29	81	92	105
		Mean: Standar	79 d deviation :	91 ±18	

Table IV.4. Biological turnover of ¹³⁴Cs

* $T_{\rm p} = 840 \, {\rm days}.$

13 days. The elimination rate was not influenced by the form in which the tracer was administered: the results were the same for the solution and the particles (P = 0.40).

Barium-133 The local simulant labeled with ¹³³Ba was fed to three subjects: in one case retention ($f_{\rm R} \times 100$) of approximately 1.0 per cent was found on the tenth day, but 11 days later no activity could be detected. On our soak tests 5 per cent of the activity was leached from the LS in 0.1 N HCl, and 0.7 per cent in aikali. The DS was fed to eight subjects, four of whom retained more than 1.0 per cent of the dose at the tenth day. The data for these four are given in Table IV.5. Absorption ($f_{\rm A} \times 100$) was 15, 6, 3 and 1 per cent, respectively. Elimmation was slow and at 100 days the amount retained was 46, 33 and 42 per cent, respectively. In our soak tests the solubility of the DS was 91 per cent in acid and 23 per cent in alkali. Three of the four subjects who received the DS displayed unexpected variations in the serial counts which were as marked as in the case of those tested with ⁸⁵Sr (see Fig. IV.2).

Dose from internal radiation. Estimates of the internal radiation dose received by the volunteers are given in Table II.4. The largest dose to the lower large intestine—696 mrad—occurred when the stable ¹³⁴Cs microspheres were used to study transit time. This is about onehalf of the maximum permissible internal dose to a single organ (1.2 rem in any 13-week period) according to the recommendations of ICRP for persons in Exposure Category B. The average

Nubject	Observed net cpm on $D = 0$	Extrapolated net cpm on D + 1	Apparent retention as per cent of fraction absorbed					Duration	Net cpm
			Ja	D + 14	D + 50	D + 100	D + 150	(days)	day
1	31.000	4800	0.15	81	58	46	42	340	1190
2	28,800	1800	0.06	67	36	33		106	677
3	24.200	740	0.03	73	43	42	18	150	127
-1	22,700	200	0.01	75				85	142
Avera	ige		0.06	74	44	40	25		

Table IV.5. Retention of 133Ba released from distant simulant

1190532

こうまちのが、あいなかったいないい、「あまた」、なれるたちのないまちのままで、

radiation dose to the LLI from ¹³⁴Cs was probably less than 300 mrad. The largest whole body dose was also supplied by ¹³⁴Cs following absorption from the DS: approximately 165 mrad, using T = 70 days. This is about 1/10 of the maximum permissible internal dose recommended by ICRP for persons in Category B. The largest radiation dose to bone + bone marrow was 18 mrad from ⁸⁵SrCl₂, using T =65 days.

Discussion

1190533

The principal objective of these experiments was to answer the question: Does the fact that radionuclides of strontium and cesium are released slowly into intestinal contents from real or simulated fallout modify their metabolic behavior during and after absorption from the gut? As a preliminary it was necessary to feed solutions of the chloride salts to obtain baselines for comparison of the time-course of elimination of radioactivity leached or dissolved from particulate simulants. Although we have no data to support the assumption, we assumed that absorption of the solutions occurred rapidly and predominantly in the upper small intestine, while absorption of leached or dissolved activity occurred slowly from more distal portions of the gut where the in-residence time of the simulant was relatively long (see Table II.1). The null hypothesis that we tested can be stated as follows. The metabolism of strontium and cesium that enters the blood from the intestinal tract is not affected by the site at which intestinal absorption occurs nor by the rate of absorption. Our results support the null hypothesis since we found that the rate of elimination (or retention as a function of time) was not significantly different, whether the tracer was given in solution or as a particulate simulant. To evalute the significance of such differences as were observed we used the WILCOXON two-sample test which is based on the rank order of the results rather than their means and standard deviations. On the basis of our findings we conclude that it is not necessary to use simulants to study the behavior of FP absorbed from ingested fallout.

Although we did not administer either ⁸⁵Sr or ¹³⁴Cs by intravenous injection the results we obtained for rate of elimination after intestinal absorption were sufficiently similar to those reported in the literature for intravenous injection to warrant the conclusion that the parenteral route is satisfactory for most studies of the metabolism of these elements. This is an important conclusion since the parenteral route has obvious advantages over the oral, the chief of which are that the amount of activity entering the blood can be known with certainty, and that zero time is definite.

Even though our principal objective was to examine the need to use simulants to study internal deposition of FP from fallout, considerable information was obtained about two aspects of the metabolism of strontium, cesium and barium: intestinal absorption and retention in a small group of healthy active adults. In addition, our findings demonstrate some of the problems encountered when the metabolism of bone-seeking radionuclides is investigated by whole body counting. We shall consider each element in turn.

Cesium. The metabolism of cesium has been studied by many investigators: after accident, with ¹³⁷Cs, and after parenteral and oral administration of the statement of istration of salts of ¹³⁴Cs and ¹³⁷Cs.⁽¹³⁾ The values reported for biological half-time (T_b) range from 65 to 135 days. Cesium chloride is readily ab sorbed, and our results ($f_A = 0.90$ for the chlo ride, and 0.82 for the oxide) are in good agreement with those reported by others, There is general agreement that a single exponential function is adequate to express the rate of elimination. In our group of twelve subjects (Table IV.4) the time of disappearance of onehalf of the ¹³⁴Cs absorbed ranged from 70 to 129 days with a mean of 91 and a standard deviation of ± 18 days. The extreme values $(T_b = 70 \text{ and } 129 \text{ days})$ were found in the two longest experiments: 207 and 163 days, respectively, after taking ¹³⁴CsCl. Although the series is not large it affords a good estimate of the biological variability of the excretion of cesium: coefficient of variation = 0.20.

Since most of the radiocesium is in muscle and soft tissue the efficiency with which the whole body detectors can "see" it should change little—if at all—with time. In all twelve cases the time course of retention was quite regular, and only in the two shown in Fig. IV.3 was there, any unexpected deviation of the net counting rate.

Strontium. There are many reports in the



FIG. IV.3. Unexpected variations after ¹³⁴Cs. These are the only two instances among more than sixty whole-body counts where the result deviated significantly from the value expected. Except for the abnormal value in each curve, elimination followed a single exponential function.

literature about the metabolism of strontium using the stable isotope as well as ⁸⁵Sr, ^{87m}Sr, ³⁹Sr and ⁹⁰Sr.⁽⁹⁾ In ICRP Publication No. 2 (see Table I.1) the value for f_1 —the fraction entering the blood from the gastrointestinal tract—is given as 0.3. In twelve of our subjects fed the solution of ⁸⁵SrCl₂ the average fraction absorbed (f_A) was 0.17 with a range 0.08-0.34. COHN, SPENCER, SAMACHSON and ROBERTSON⁽¹⁴⁾ reported absorption of 20.7 and 16.4 per cent, respectively, by two patients with osteoporosis. SPENCER et al., (15) using 90Sr in contaminated food as the tracer, found a mean absorption of 5.3 ± 3.01 per cent in a group of twelve young munteers. Lourir⁽¹¹⁾ believes that "around 20 per cent seems to be a fairly representative figure to take for absorption of soluble strontium salts". It is of interest that when he gave 100-200 mg of strontium as a soluble salt in water, some 35 per cent was absorbed. He attributed the larger absorption under these conditions to a mass effect. In our study, the largest value for $f_{\mathbf{A}}$ (0.47) was found in one of the subjects who received the first batch of the DS which contained abundant carrier SrO. The biological variability of absorption of strontium is thus much greater than that for cesium. When we fed

1190534

⁸⁵SrCl₂ absorption ranged from 8 to 34 per cent, and in the entire group of subjects the range was 6 to 47 per cent. If we set aside the five subjects who received the first batch of DS because their average absorption was significantly different from all the others (P = 0.004), then we can treat the remaining twenty as a sample that received tracer amounts of strontium. For this group, the mean absorption was 16.4 ± 7.9 per cent, and the median was 14.0 per cent. The coefficient of variation, 0.48 is greater than that reported by SPENCER *et al.*⁽¹⁵⁾; and is also much larger than that for cesium.

The general agreement about the extent of absorption of strontium from the gut does not extend to retention. Reports in the literature vary so greatly that it is not possible to cite a consensus. Biological half-life is often an ambiguous concept and the wide range of values reported for Sr turnover may be due in part to imprecise definition of the term. Using an exponential model COHN *et al.*⁽¹⁴⁾ reported biological half-times ranging from 210 to about 1000 days. LOUTTT⁽¹¹⁾ estimated that about one-seventh of a dose of strontium is due for slow turnover along with the bone in which it is incorporated. He calculated the rate of bone



FIG. IV.4. Long-term retention of ⁸⁵Sr absorbed from gut. The open circles (\bigcirc) are averages for retention by twenty subjects who received either ⁸⁵SrCl₂, LS, or the second batch of DS. The closed circles (\bigcirc) are the averages for the five subjects who received the first batch of DS. The median of the values for retention by all twenty-five subjects is indicated by the crosses (+).

replacement as 6 per cent per year, so that T_b for strontium in bone would be about 12 yr (4400 days). This is less than the value proposed by Committee II of ICRP: $T_b = 18,000$ days. In general, our results are consistent with Lourrr's estimate that about one-seventh of the strontium absorbed remains in the body at the end of the first year. If excretion by our subjects had continued at about the rate that we observed during the first several hundred days, then about 16 per cent of the ⁸⁵Sr absorbed would have been retained at the end of the first vear (see Fig. IV.4).

We were perplexed by the occasions when the net counting rate was significantly different from that expected on the basis of previous serial measurements. After excluding all possible operating errors, unexpected results were obtained in sixteen of the forty-five subjects who received ⁸⁵Sr. Examples of these are shown in Fig. IV.2. The curve for LS-2 is satisfactory, while that for DS-8 demonstrates almost complete disappearance of ⁸⁵Sr between the Days 62 and 100. Four others behaved in the same fashion, all occurring during the summer months. Aside from the possibility that vigorous physical activity may have been responsible we have no explanation for the occurrence. In several subjects there was an unexplained increase in the net counting rate as illustrated by the curve for LS It is conceivable that redistribution of ⁸⁵Sr the skeleton permitted the detectors to "see" in activity with greater efficiency. This couhappen if strontium deposited in the verteb and ribs was relocated to arms or legs when there is a smaller amount of overlying soft tissue. All subjects were counted in the prone and the supine position to compensate for unequal at terior-posterior distribution such as may occur in the trunk region. Obviously such variation complicate any study of long-term retention. Barium. Because of their cardiotoxicity life is known about the long-term metabolism barium salts beyond the fact that approximat 70 per cent of the fraction absorbed from the goes to bone where the half-life is reported to 65 days (Table I.1). We fed DS consisting $BaO + {}^{133}BaO$ to eight subjects, but only for absorbed a sufficient amount to permit an mate of f_A . Long-term retention could followed in three of these for times ranging fi 100 to 340 days. Unexpected variations of net counting rate occurred in all three resembled those seen with ⁸⁵Sr. The data for subject who absorbed the most barium (fl 0.15) are shown in Fig. IV.2. The configura of the curve resembles that for strontium, retention measured at about 1 yr (340 days)

15 per cent of the amount absorbed. The DS made with ¹³³Ba was the most soluble of the annulants used.

V. CONCLUSIONS

After $2\frac{1}{2}$ years of work and the planned exposure of 102 healthy volunteers to small amounts of internal radiation it is appropriate to ask what we have learned that has relevance for Civil Defense and health physics.

First, we are not in much better shape than when we started with respect to our ability to predict the magnitude of the hazard from internal deposition of radioactivity following ingestion of fallout particles. It appears that local silicaceous fallout is not so soluble as it was assumed to be-if our sample was at all typical. The biological availability of radioactivity from such fallout was about one-third of the value usually quoted. This small advantage is offset by the uncertainty of intestinal motility which may vary over quite a wide range. There is also evidence-not too conclusive perhaps-that the intestinal contents of man are much more effective than dilute acid or alkali as a leaching agent for the glassy simulant that we used. We have not studied the reactions responsible for this and cannot say whether enzymes, chelating agents or intestinal microflora are responsible. On the basis of our limited experience it does net seem proper to equate biological availability win solubility or leachability as measured by soak tests using 0.1 normal acid and alkali.

Second, we have demonstrated that it is not necessary to use particulate simulants of fallout to study the metabolism of such critical nuclides as those of strontium, cesium or barium. The metabolic fate of strontium and cesium that gain rutrance to the blood is apparently not influrutrance to the blood is apparently not influrutrance to the blood is not trivial, however, and presumably the same is true for atom. This conclusion is not trivial, however, atom. This conclusion is not trivial, however, atom there might be quantitative or qualitative afferences in the fashion in which the body desposes of material injected intravenously, and maknown chemcial compounds of the same torment absorbed from the gut.

Third, we learned quite a bit about the distribution of such values as the fraction of dose at surfield from the gut and the rate of elimination

of strontium, cesium and barium. This was possible because of the large number of volunteers who were willing to participate in a study that concerned an aspect of Civil Defense. It was also possible because of the comparative ease of measuring long-term retention of γ -emitting nuclides using the whole-body counter. The important finding was a better delineation of biological variability, which is considerable for cesium, and much greater for strontium and barium. The range of variability was greater than we had anticipated.

Fourth, there appears to be little difference between our findings with healthy active volunteers and the results reported by others who studied hospitalized patients whose metabolic state was not greatly disturbed by illness.

Finally, our results may be useful to health physicists by providing additional empirical information about absorption and elimination of strontium, cesium and barium. When our values are compared with those of the ICRP (see Table I.1) there is good agreement for the absorption of cesium: $f_A = 0.89$ vs. $f_1 = 1.0$. Our mean value for f_A of strontium—0.17—is probably significantly lower than the ICRP's $f_1 = 0.30$. We had too few subjects to warrant any conclusion about the absorption of barium except that it varied remarkably.

The biological half-time (T_b) for cesium in our healthy young adults was 91 \pm 18 days which is probably not significantly greater than the ICRP value: 70 days. It is difficult to evaluate T_b for strontium since we used the relatively short-lived ⁸⁵Sr, and have no information about long-term retention (i.e. longer than 1 yr): our longest study was 247 days. We can say with some confidence that about 40 per cent of strontium absorbed from the gut was eliminated during the first week after a single ingestion, and that about one-sixth of the fraction absorbed remained in the body at the end of the first year. This burden which is equivalent to about 3 per cent of the amount ingested (0.16×0.17) is surely available for long-term retention. During the first year after ingestion we can estimate a $T_{\rm b}$ of about 50 days for short-term turnover for 10 per cent of the amount ingested (0.17×0.60) since 60 per cent of the fraction absorbed remained at 7 days, and about 30 per cent at 50 days. (See Fig. IV.3.) Following LOUTIT⁽¹¹⁾

471

「たいたからうちょう

we may assume that strontium remaining after about 1 yr is subject to long-term turnover as bone is replaced at a rate that approximates 6 per cent per year in adults ($T_b = 12$ yr). Using these values in the case of a single ingestion of ⁹⁰Sr would result in a much smaller estimate of radiation dose to bone than is found when the ICRP values are used: $f_w = 0.21$, and $T_b =$ 1.8×10^4 days.*

Barium should behave like strontium with respect to long-term retention, and the fraction ultimately deposited in bone should turn over at a rate that approximates that of bone replacement. The ICRP value for T_b of barium is 65 days, and is quite different from what we found in three satisfactory studies (see Table IV.5). Approximately one-half the fraction absorbed (f_{Δ}) was eliminated during the first 50 days, as was the case with strontium. At the end of 1 y the measured retention of barium was 25 per cent of the amount absorbed in one subject, and in another the extrapolated retention was about 10 per cent.

Individual variation in the absorption of strontium and barium was remarkable and we have no explanation for the cause of it. Even more difficult to explain is the unexpected reduction of strontium burdens in five of eighteen

 $f_{\omega} = 0.21$, T(bone) = 6400 days, and using $\Sigma \text{ EF}(\text{RBE})n$ for ${}^{90}\text{Sr}-{}^{90}\text{Y}$ for E_{β} in bone = 5.5 MeV

$$D_{\beta} = 73.8 C E_{\beta} T$$
 rad

$$= 73.8 \times 0.003 \times 5.5 \times 6400$$

= 7793 rad.

- (2) Using values suggested by this study: "Short-term" turnover:
 - $f_{w} = 0.07$ (i.e. 10 per cent of ingested remains presumably in bone after 1 week; and 3 per cent remains after 1 year),

T = 50 days, and $E_{\beta} = 5.5$ MeV

$$D_{\beta^1} = 73.8 \times 0.001 \times 5.5 \times 50$$

= 20 rad.

r

1190537

- "Long-term" turnover: f = 0.02 T = 4400 day
- $f_w = 0.03, T_b = 4400 \text{ days}$ $T = 3000 \text{ days}; E_B = 5.5 \text{ MeV}$
- $D_{d^2} = 73.8 \times 0.0004 \times 5.5 \times 3000$
- $\gamma_{\beta}^{-} = 487 \text{ rad.}$

$$D_3 = D_8^1 + D_8^2 = 507 \text{ rad.}$$

subjects who were studied for longer than months. In each case the drop occurred affia vacation, and may well have been associate with accelerated remodeling of bone provole by unusual physical activity. This variability in the metabolic behavior of strontium and barium is in striking contrast to that of cesium which was almost completely absorbed from the gut and eliminated at strictly exponential rate by all twelve subjects.

SUMMARY

(1) The behavior of real and simulated fail out following ingestion was studied in 10 volunteers using the whole-body gamma spectrometer.

(2) Ten volunteers ingested samples of typical local fallout. The average biological availability of the radioactivity in this material was 3, per cent. Two of the subjects absorbed almost nonand only four of the ten absorbed more that 3 per cent. (3) The rate of excretion of insoluble particulate material from the gastrointestinal trace was followed in twenty-seven subjects. On the average about one-half the material was creted during the first 2 days following ingestion. In all but three of seventeen subjects 99.9+ per cent was gone by the seventh day.

(4) Simulants of fallout labeled with ⁸⁵Sr,¹³⁴Cs or ¹³³Ba were fed to forty-three subjects, and solutions of ⁸⁵SrCl₂ or ¹³⁴CsCl were fed to twenty-two. The rate of elimination of strontium and cesium was the same whether the tracer was given in solution or whether it was absorbed from the simulant.

(5) On the average, about 17 per cent of the strontium tracer was absorbed from the gut. Of the amount absorbed, about 60 per cent was retained at the end of the first week and about 16 per cent at the end of 1 yr. Carrier strontium enhanced absorption but appeared to decrease retention to some extent. The biological variability of absorption of strontium was large: when ⁸⁵SrCl₂ was fed the fraction absorbed ranged from 0.08 to 0.34.

(6) In twelve subjects the biological half-time for elimination of 134 Cs was 91 \pm 18 days. When 134 CsCl was fed about 90 per cent of the trace was absorbed.

(7) Using BaO + ¹³³BaO as a distant simulant

[•] Total dose (D_{β}) to bone after a single ingestion of 100 μ c of soluble ⁹⁰Sr:

⁽¹⁾ Using ICRP values for biological and physical constants:

DS) average absorption was 6 per cent in eight subjects, with a range of 1-15 per cent. Elimination of barium by one subject was slower than for strontium: 25 per cent of the amount absorbed was retained at the end of 1 yr.

(8) There was no correlation between the biological availability of the simulants and leachability or solubility as measured by soak tests in 0.1 N HCl and 0.1 N NaOH.

Actinowledgements—The authors are indebted to Dr. CAROL M. NEWTON for assistance in the early phase of this study. They are especially grateful to the 102 volunteers who hoped that they were making a contribution to Civil Defense.

REFERENCES

- 1. Effects of Nuclear Weapons. U.S. Department of Defense, U.S. Government Printing Office, Washington, D.C. (1962).
- 2. C. F. MILLER, Fallout and Radiological Countermeasures, Vol. 1. Stanford Research Institute (1963).
- 3. Damage to livestock from radioactive fallout in event of nuclear war, National Academy of Sciences National Research Council Publication 1078, Table XIII, p. 53. Washington, D.C. (1963).
- 4. T. N. LAHR and J. P. RYAN, Properties and Medical Uses of a Unique Ceramic Carrier for Radioisotopes, Paper presented at the Central Chapter, Society of Nuclear Medicine, Rochester, Minnesota, 29 October 1961; see also Chem. Eng. News, p. 20, 3 October (1960).

1190538

- 5. ICRP Publication 2, Report of Committee II on Permissible Dose for Internal Radiation. Pergamon Press, London (1959).
- 6. G. J. HINE and G. L. BROWNELL, Radiation Dosimetry. Academic Press, New York (1958).
- 7. Whole-Body Counting, Proceedings of the Symposium on Whole-Body Counting, International Atomic Energy Agency, Vienna (1961).
- 8. R. L. HAYES, J. E. CARLTON and W. R. BUT-LER, JR., Health Phys. 9, 915 (1963).
- 9. E. H. QUIMBY, S. FEITELBERG and S. SILVER, Radioactive Isotopes in Clinical Practice. Lea & Febiger, Philadelphia (1958).
- 10. G. M. DUNNING, Am. Ind. Hyg. Ass. J. 19, 111 (1958).
- J. F. LOUTIT, Strontium 90, Irradiation of Mice and Men, Chap. 3. University of Chicago Press, Chicago (1962); see also K. WILLIAMS, Strontium Studies, AERE-R-3423.
- 12. W. A. WALLIS and H. V. ROBERTS, Statistics, p. 594. Free Press, Glencoe (1960).
- C. R. RICHMOND, J. E. FURCHNER and W. H. LANGHAM, Health Phys. 8, 204 (1962); S. E. HAM-MOND, F. O. BOLD and N. S. MACDONALD, Health Phys. 8, 523 (1963).
- S. H. COHN, H. SPENCER, J. SAMACHSON and J. S. ROBERTSON, *Radiat. Res.* 17, 173 (1962).
- H. SPENCER-LASZLO, J. SAMACHSON, E. P. HARDY, JR. and J. RIVERA, Clin. Sci. 24, 405 (1963).
- Radiological Health Handbook. U.S. Department of Health, Education and Welfare, Washington, D.C. (1960).