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SEMIANNUAL PROGRESS REPORT

FOR PERIOD ENDING JULY 31, 1955

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For Period Ending July 31, 1955

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HEALTH PHYSICS DIVISION
SEMIANNUAL PROGRESS REPORT

APPLIED RADIOBIOLOGY

E. G. Struxness

DISTRIBUTION AND EXCRETION OF URANIUM IN MAN

S. R. Bernard J. R. Muir
J. C. Gallimore G. J. Dodson
G. W. Royster, Jr. N. L. Gillum
C. S. Banks

The intravenous administration of fissionable uranium in man for its possible application to neutron-capture therapy[1-3] has continued during the period of this report. To date, eight terminal brain-tumor patients have been studied, seven of which have died. Blood, cerebrospinal fluid, urine, and fecal samples were collected at Massachusetts General Hospital in accordance with established routine and sent to ORNL for analysis. Autopsy tissue samples were obtained in six of the seven terminations. Table 1 summarizes the pertinent injection data.

The data which have been collected on the first six patients have been tabulated and plotted. Figure 1 shows graphs of the disappearance of uranium from the blood. On an average, 99% of the injected dose is removed from the circulation in about 20 hr, assuming uniform distribution at injection time and a blood volume of 7.14% of body weight. Once reduced, the blood values remain relatively constant until time of expiration. Figure 2 shows the urinary excretion data obtained in this study. These data are presented as log-log plots to compare the excretion rates on an identical time scale. Most of the excretion takes place in the first 24 hr after administration. Essentially all the uranium is excreted in the urine, since less than 1% of the injected dose is found in the feces.

Table 2 lists the autopsy and biopsy findings in terms of per cent of injected dose per gram of tissue. This method of reporting values was necessary because it was impossible to obtain weights of all the organs and tissues sampled. If the weight values of standard man[4] are assumed, and if these values are converted to per cent of injected dose per organ or per tissue, it is evident that all soft tissues, excluding kidney, contain approximately 20% at 60 hr postinjection and less

[1] W. H. Sweet and M. Javid, *J. Neurosurg.* 9, 200–209 (1952).

[2] M. Javid, G. L. Brownell, and W. H. Sweet, *J. Clin. Invest.* 31, 604–610 (1952).

[3] L. E. Farr *et al.*, *Am. J. Roentgenol., Radium Therapy Nuclear Med.* 71, 279–291 (1954).

[4] S. Kinsman *et al.*, *Radiological Health Handbook*, NP-4071, p 127 (Sept. 1952).

TABLE 1. SUMMARY OF INJECTIONS IN THE DISTRIBUTION AND EXCRETION STUDY

Patient	Expiration Time (days)	Injection Compound	Injection Dose (μc)	Autopsy Samples Obtained
I	2½	$UO_2(NO_3)_2 \cdot 6H_2O$	12.2	Yes
II	74	$UO_2(NO_3)_2 \cdot 6H_2O$	9.8	Yes
III	566	$UO_2(NO_3)_2 \cdot 6H_2O$	10.1	Yes
IV	136	$UO_2(NO_3)_2 \cdot 6H_2O$	33.4	No
V	139	$UO_2(NO_3)_2 \cdot 6H_2O$	47.2	Yes
VI	18	$UO_2(NO_3)_2 \cdot 6H_2O$	3.3	Yes
VII		UCl_4	2.6	
VIII	21	UCl_4	3.1	Yes

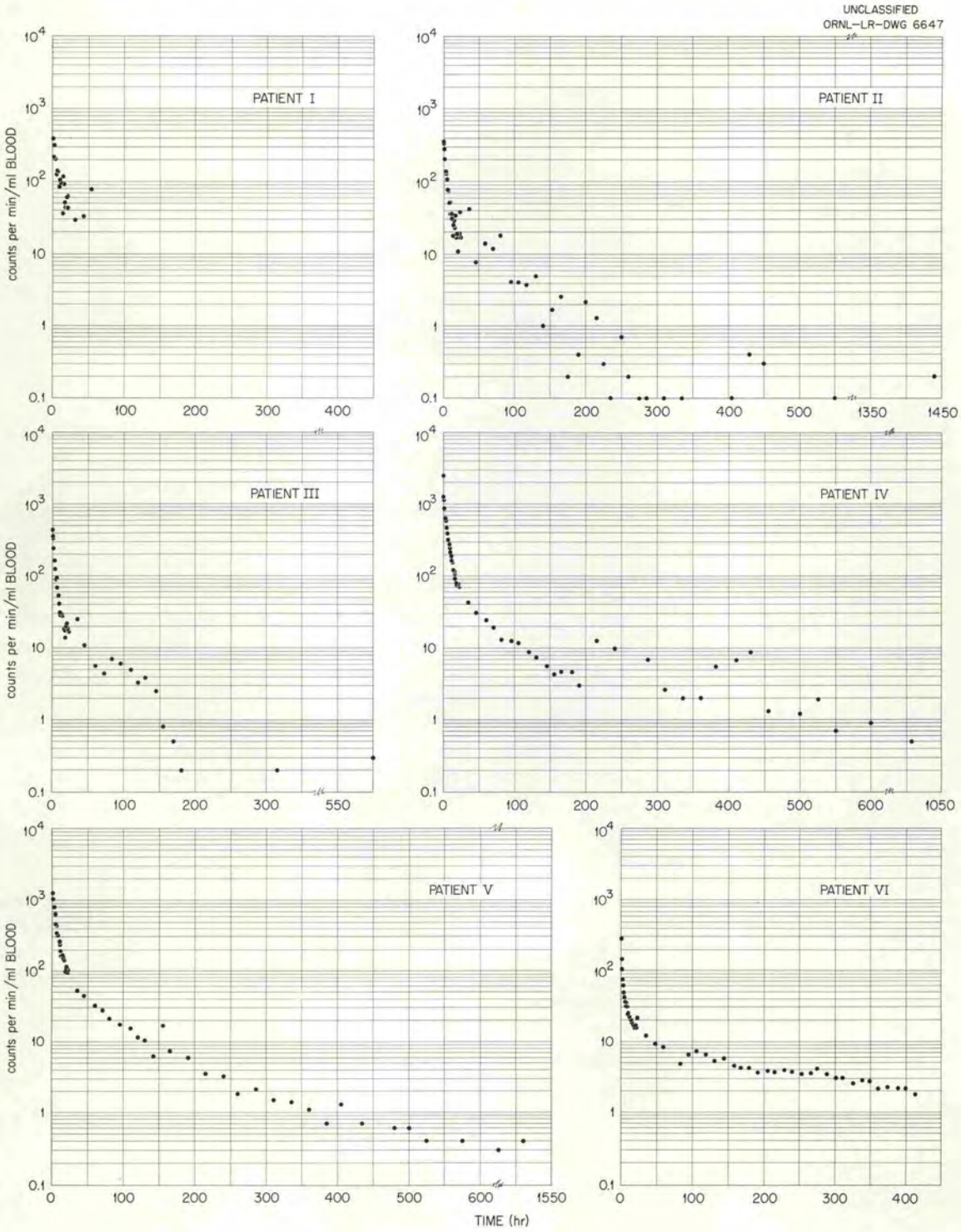

Fig. 1. Uranium Disappearance from Blood.

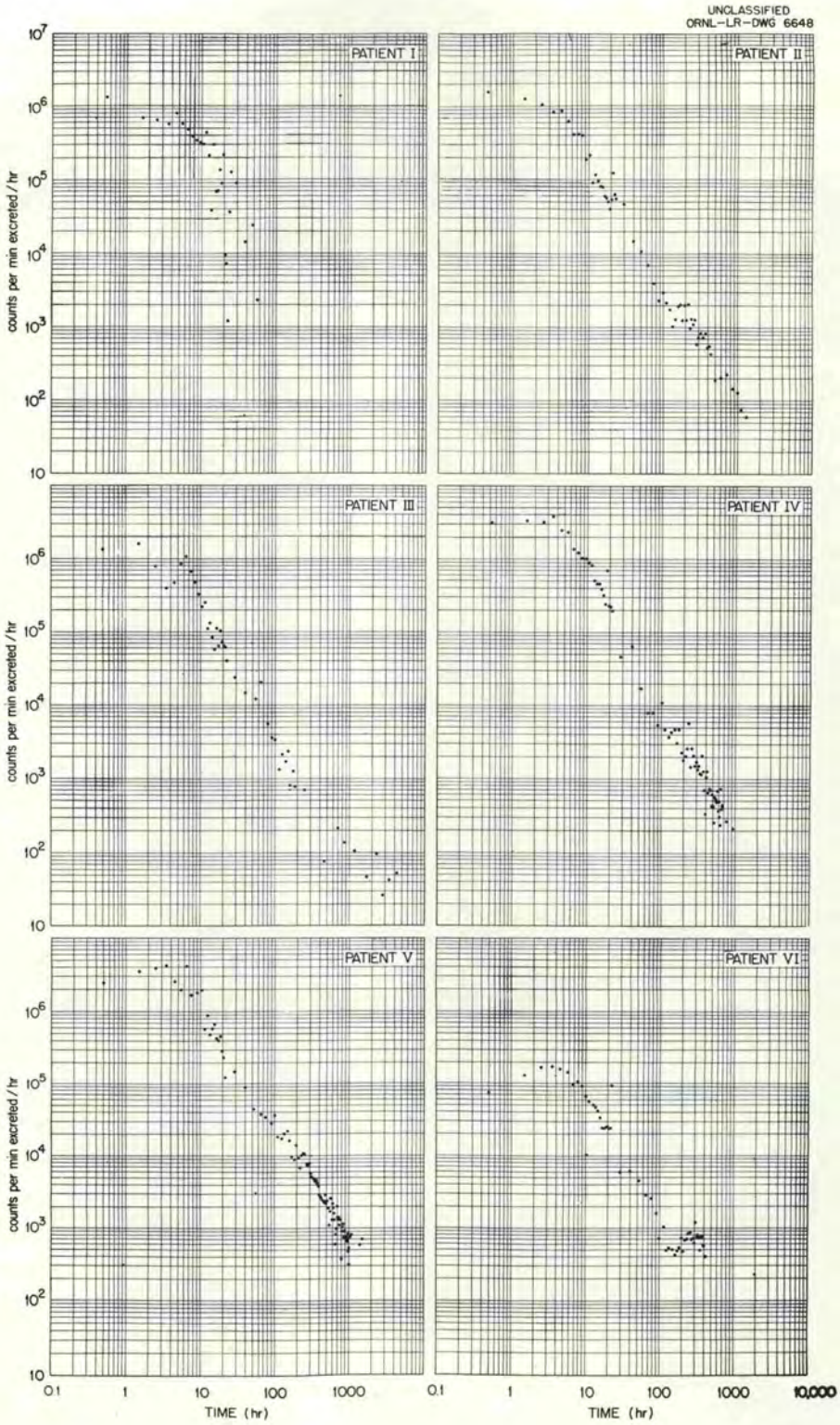

Fig. 2. Urinary Excretion of Uranium.

than 1% at 3100 hr. These residual percentages are shown in Fig. 3.

Total body content of uranium, determined by cumulating the per cent of injected dose in urine and subtracting from 100, is shown in Fig. 4 and is seen to be slightly different for each patient.

TABLE 2. BIOPSY AND AUTOPSY RESULTS FOR PATIENTS INJECTED WITH U(VI)
PER CENT OF INJECTED DOSE PER GRAM $\times 10^{-4}$

	Patient Number					
	I	II	III	IV	V	VI
	Bone Biopsy Results					
0–24 hr	8.7	11.2	12.9	10.8	8.0	8.8
	7.9	6.7	3.3		6.3	9.3
	23.4	4.4	2.0		5.9	
	16.2	15.9				
	8.2					
	13.4					
	17.9					
24–48 hr	7.0				2.2	
	15.1					
	Autopsy Data					
Expiration Time (days)	2½	74			139	18
Organ						
Adrenal gland	11.1	1.6			0.6	6.0
Aorta	7.1				0.01	7.1
Bladder	2.2	0.1			0.09	
Brain					0.05	13.8
Brain (frontal lobe)	270.8	0.2				
Brain (parietal lobe)	28.2					
Brain (temporal lobe)		0.1				
Cartilage					0.1	
Cartilage (rib)		0.9				
Cartilage (tracheal)						3.5
Fat (subcutaneous)	0.6					0.6
Fat (from skin)						8.7
Femur (II & V bone from shaft)		0.5			0.6	
(I & VI bone from distal end)	5.8					4.4
Gall bladder	4.8				0.2	13.7
Heart	2.0	0.1			0.2	0.7
Intestine (small)	2.1	0.3			0.1	1.8
Kidney	554.0	22.1			39.3	239.9
Liver	10.4	1.4			0.9	6.5
Lung	4.7	0.3			0.2	3.6
Lung (necrotic)	23.9					
Marrow (bone)		0.1			0.2	
Muscle	0.4	0.3			0.09	1.0
Muscle (psoas)						0.3
Pancreas	96.0	1.2			0.08	1.1
Prostate		0.2			0.2	1.6

TABLE 2 (continued)

Organ	Patient Number I	II	III	IV	V	VI
Rib	19.7	2.6			0.9	42.1
Skin	3.0	0.2			0.1	1.7
Skull	7.9	11.9			1.9	
Spleen	18.8	3.6			0.5	7.7
Sternum					0.5	
Stomach	3.2	0.1			0.05	0.8
Testis		2.1			0.05	2.9
Thyroid		0.1			0.07	1.5
Tumor (tissue from edge)	24.8	0.6			0.4	
Tumor (viable)		0.6				
Vertebrae (lumbar)		1.8				
Vertebrae (thoracic)	53.8					

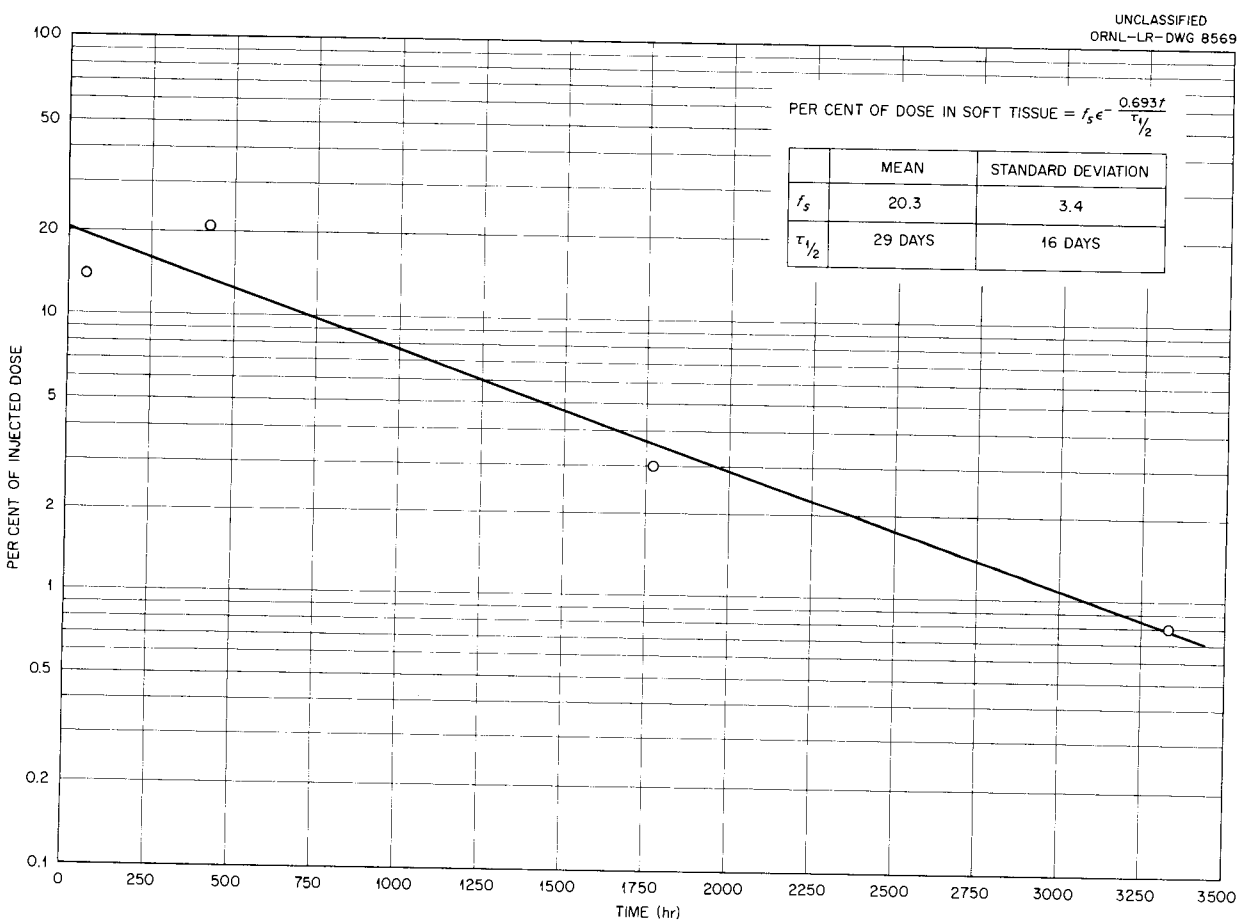

Fig. 3. Per Cent of Injected Dose in Soft Tissues (Other Than Kidney) at Autopsy.

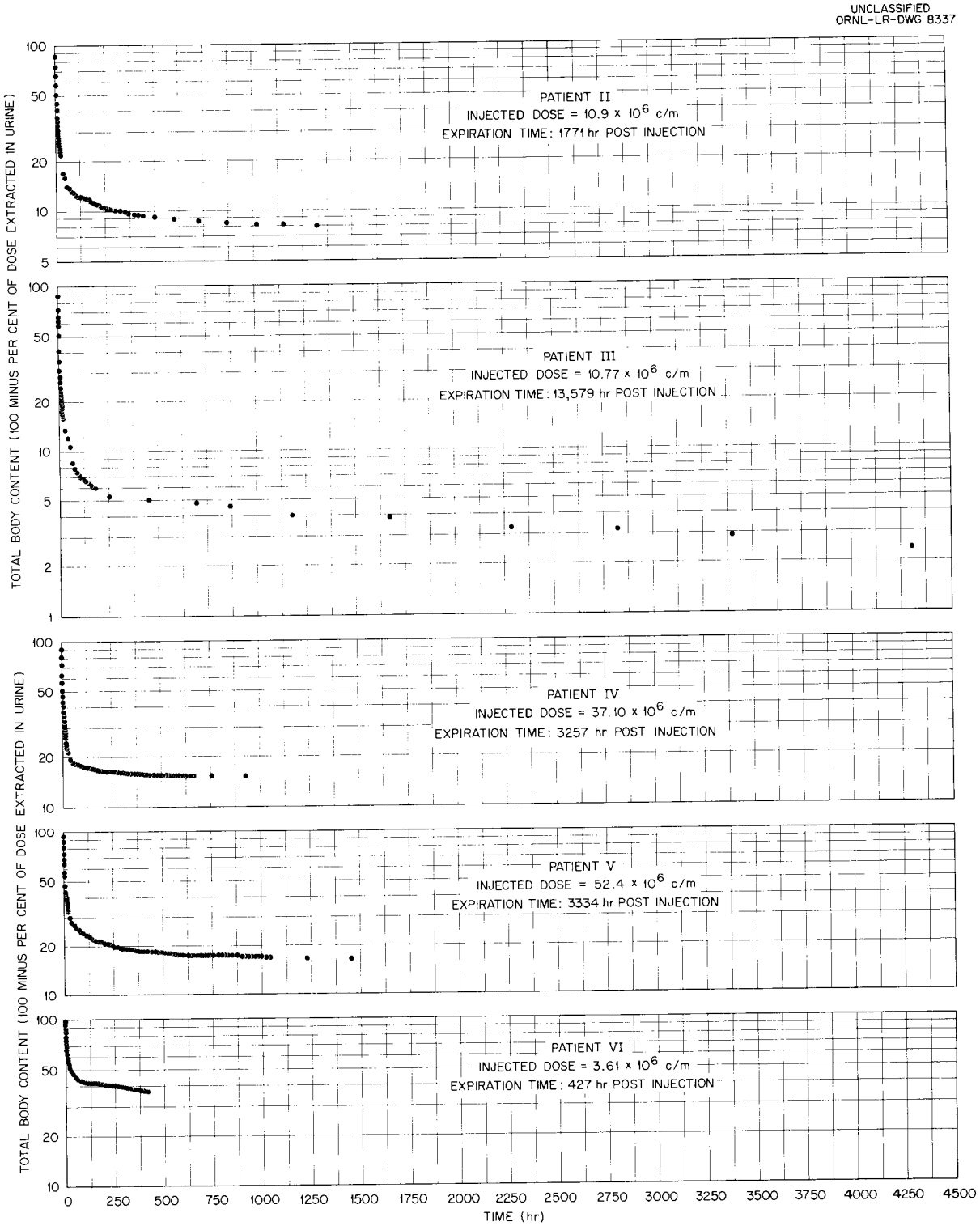

Fig. 4. Per Cent of Injected Dose Retained in the Body as a Function of Time After Injection.

ECOLOGY

S. I. Auerbach

H. F. Howden[5] M. D. Engelmann[6]
V. L. Sheldon[5] R. J. Davis[6]

Studies of the Uptake of Fission Products by Earthworms

Uptake of Sr^{89} by *Eisenia foetida* (Savigny). – Earthworms are a group of organisms that play an important role in the maintenance of soil fertility. Their value in this regard was first extensively discussed by Darwin in 1882.[7] More recently, in 1948, their ecology in relation to agriculture has been reviewed by Hopp.[8] These organisms are also important food items in the diets of many higher animals and as such are links in natural food chains.

In view of the ecological significance of earthworms, it was considered advisable to begin a study of the effects on earthworms of the fission products which might, at some future time, be released into the soil environment. The primary objective of the first experiment was to test the ability of certain species of earthworms to take up from local soils and to concentrate the radionuclide Sr^{89}. The studies were initiated with Sr^{89}, not only because it is an important fission product, but also because it has certain technical advantages in an initial series of experiments. These advantages include a relatively short half life and an ease of analysis. The earthworms used in these experiments were the common red worm, *Eisenia foetida* (Savigny),[9] and were obtained from commercial bait stores.

Red-yellow podsolic soil was brought in from the laboratory area, air-dried, and sieved through 10- and 40-mesh screens to yield a soil mass of uniform particle size. Organic matter was provided in the form of pulverized peat. The chemical analysis of this soil was as follows: organic matter, 5%; P_2O_5, 31 ppm; K, 11 ppm; Mg, 13 ppm; Ca, 110 ppm; pH, 4.1; total hydrogen, 10 milliequivalents per 100 g; cation exchange capacity, 16.82 milliequivalents per 100 g; per cent base saturation, 40.5.

One kilogram (corrected for moisture content) of soil was placed in each of 25 red-clay flower pots. These were divided into 20 experimentals and 5 controls. The experimental pots were subdivided into five series of four pots; each series contained a different activity level of Sr^{89}. The isotope was added to the soil by taking aliquots at the required activity level, diluting them to 100 ml with water, and thoroughly slurrying this solution with 100-g aliquots of soil from each of the experimental pots. The initial amounts of strontium used were sufficient to produce the following activities (in counts per minute per gram) in each series of pots: 350, 700, 1050, 1400, and 1750 – all at 10% geometry.

After the isotope was mixed with the aliquots of soil, the solutions were dried, pulverized in mortars, and homogeneously blended into the remaining 900 g of soil from each experimental pot, by putting both dry mixtures into a stainless steel cylinder, sealing it, and then rotating it for 15 min on a lathe. The soil was then returned to the pots, and sufficient water was added to bring the soil to optimum moisture. Controls were treated in similar fashion except that the isotope was not added. One day later ten adult worms were added to each of the pots. Moisture was added as needed. After 30 days the pots were examined for the surviving worms. A total of 168 out of an initial inoculation of 250 worms were found after 30 days. Forty-one of the survivors were taken from the experimental and the control pots and were immediately killed and prepared for counting. The remainder were placed in moist sphagnum moss for three days to "scour" their intestinal tracts of any soil that might be within them. This was done because the activity of Sr^{89} in the worms was so low that the isotope-containing soil held in the gut might produce misleading results. This was proved to be the case, as is shown by Table 3. After removal from the moss, the worms were sacrificed.

All worms were prepared for counting by digestion in concentrated HNO_3 with H_2O_2 added. After all the organic material was destroyed, the residue was dissolved with 15 to 20 drops of 1 *N* HNO_3 and transferred to a stainless steel counting dish of 3.2 mm depth and 25 mm diameter. All samples were counted on the second shelf of a

[5]Research participant.

[6]Temporary summer employees.

[7]C. R. Darwin, *The Formation of Vegetable Mould, Through the Action of Worms, with Observations of Their Habits*, Appleton, New York, 1882.

[8]H. Hopp, *Soil Sci. Soc. Amer., Proc.* 12, 503–507 (1948).

[9]The authors are indebted to Dr. William Murchie, Thiel College, Greenville, Penn., for the identification of the earthworms.

TABLE 3. ANALYSIS OF Sr^{89} ACTIVITY IN EARTHWORM, *EISENIA FOETIDA* (SAVIGNY), AFTER 30 DAYS

Series No.	Initial Activity of Soil at 10% Geometry (counts/min/g)	With Soil Present in Gastrointestinal Tract			Without Soil in Gastrointestinal Tract		
		Number of Individuals	Average Activity (counts/min)	Per Cent Activity of Total in Soil	Number of Individuals	Average Activity (counts/min)	Per Cent Activity of Total in Soil
I	350	7	10.9 ± 2.7	3.1×10^{-3}	21	1.3 ± 2.4	3.7×10^{-4}
II	700	7	22.4 ± 2.9	3.2×10^{-3}	20	5.9 ± 2.5	8.4×10^{-4}
III	1050	6	34.3 ± 3.1	3.3×10^{-3}	24	6.7 ± 2.7	6.6×10^{-4}
IV	1400	7	42.3 ± 3.5	3.0×10^{-3}	21	9.5 ± 2.9	6.7×10^{-4}
V	1750	8	51.6 ± 3.2	2.9×10^{-3}	11	11.7 ± 2.6	6.7×10^{-4}
Controls		6	0.6 ± 2.3		30	0.4 ± 2.3	
		41			127		

thin-window G-M counter. Samples were counted for 40 min and backgrounds for 20 min. Statistical error at the 95% confidence level was calculated for each sample. The results are summarized in Table 3.

These results indicate that there was some uptake of Sr^{89}. Under the particular experimental conditions there was some correlation between the amount of radioactivity in the earthworms and the environment in which they were kept. In all cases the order of magnitude of uptake was very small in comparison to the uptake of Sr^{89} by vertebrate animals.

If the average radioactivity in each series of experimental worms is expressed as the percentage of the total activity available to them in the soil mass for the 30-day period, it is apparent that under the conditions of this experiment there was no concentration of Sr^{89}.

Studies of the Effectiveness of Various Types of Containers in Maintaining Tree-Hole Arthropod Populations in the Laboratory

A necessary prerequisite for experimentation on the delayed effects of ionizing radiation on tree-hole arthropod populations is a suitable container which will maintain the populations in their natural substrate for extended periods of time. Four types of containers were tested over a three-month period for their ability to fulfill this need; they were (1) two artificial tree holes bored in a 3-ft *Liriodendron tulipifera* log kept in the laboratory, (2) two artificial tree holes bored in two separate trees of the same species as the log, (3) two wooden boxes constructed of the same wood as the logs, and (4) two 6-in. red-clay flower pots.

In January, 400 g of *L. tulipifera* tree-hole mold was placed in each of the containers. To prevent excessive moisture loss, the boxes and flower pots had glass covers and the artificial holes had wooden lids with a screen opening. One of the boxes and one of the flower pots were placed in an airtight chamber into which air of 85% relative humidity was piped. The other box and flower pot were kept in the laboratory, where the relative humidity was about 30%. All the containers were examined weekly for moisture content, and water was added when needed. At intervals of 30, 60, and 90 days samples of mold were taken from each of the containers and from the original tree holes, the latter serving as controls. These samples were weighed and were processed in Berlese funnels to separate the arthropod populations, and the arthropods were counted and then separated into major taxonomic categories.

The pertinent data are summarized in Table 4. These data include the fresh weight of the sample from each container, the moisture content of the mold sample at the time of "berlesing," the number of arthropods per gram of dry weight of mold, and the frequency of the taxonomic categories in each sample.

TABLE 4. SUMMARY OF TESTS OF CONTAINERS OF TREE-HOLE MOLD (FOR THE PERIOD FROM JANUARY TO JUNE 1955)

Type of Container	30-day Berlese				60-day Berlese				90-day Berlese			
	Wet Weight of Sample (g)	H_2O (%)	Arthropods per Gram of Dry Weight	Frequency of Categories	Wet Weight of Sample (g)	H_2O (%)	Arthropods per Gram of Dry Weight	Frequency of Categories	Wet Weight of Sample (g)	H_2O (%)	Arthropods per Gram of Dry Weight	Frequency of Categories
I. Wood box (in airtight container)	64	21.2	2.8	5	56	26.1	1.52	6	87	46.2	7.3	8
II. Wood box	62	17.8	0.94	3	57	7.4	0.72	1	77	15.4	6.7	6
III. Flower pot (in airtight container)	75	34.9	24.1	8	66	25.9	22.4	5	99	52.1	12.8	7
IV. Flower pot	86	38.2	19.0	7	77	38.9	16.8	4	98	56.6	18.3	5
V. Log hole (laboratory)	73	30.1	33.2	7	63	27.4	27.9	3	77	41.0	38.8	7
VI. Log hole (laboratory)	72	26.6	20.9	6	73	31.4	59.5	6	81	34.9	23.6	6
VII. Log hole (field)	75	41.8	18.2	5	174	74.7	8.9	5	137	74.5	12.6	6
VIII. Log hole (field)	94	35.7	14.1	7	86	39.7	19.4	7	101	52.0	46.0	6
Control	82	33.8	23.9	8	64	28.7	13.3	6	62	22.0	17.5	11
Control					54	11.8	6.5	6				

The results indicate that the flower pots and the artificial tree holes in the laboratory are the most satisfactory of the containers. The arthropod populations were maintained at levels equal to, or in excess of, the populations of the artificial holes in the field and of the controls. There were fluctuations in the frequencies of taxonomic categories, but the data are too few for any conclusions to be drawn. In general, in all the containers the numbers of individuals, and in some cases the frequency of taxonomic categories, increased during the experiment. This was due to new individuals being hatched from eggs either already present or oviposited during the course of the experiment.

It was also found that the amount of moisture lost was least – and that optimum moisture levels could best be maintained – in the flower pot in the airtight chamber. In addition, in this type of container there is less possibility of contamination by extraneous organisms, not originally present at the start of the experiment, moving into the pot.

URINALYSIS RESEARCH

L. B. Farabee

The development of a procedure for the determination of radioactive strontium in urine specimens has been completed, and a detailed report has been prepared for publication.[10] Although this procedure was designed primarily for analysis of urine for strontium, it recovers radioactive barium equally as well. This is to be expected, because the stability constants of the versene chelates of barium and strontium are similar. Since this urinalysis procedure does not provide a chemical separation of strontium from barium, identification of the radioactive isotope can be made by half-life studies.

The inorganic constituents of the final sample from a 24-hr urine specimen consist of tracer amounts of calcium, magnesium, and sodium and essentially all the inert strontium and barium in one day's excretion. Preliminary tests show that about 0.3 to 0.4 mg of inert strontium is excreted per day. This strontium can be precipitated from all the other elements except barium by two $Sr(NO_3)_2$ precipitations from an 84% nitric acid solution. This precipitation can be used as an adjunct to the procedure. Since the sample can be counted a short time after the separation of the rare-earth daughter products of Sr^{90} and Ba^{140} by this precipitation, growth and/or decay studies can be made to identify the radioactive isotope.

[10] L. B. Farabee, *Procedure for the Radiochemical Analysis of Strontium and Barium in Human Urine*, ORNL-1932 (Sept. 6, 1955).

DISTRIBUTION OF RADIOISOTOPES IN ANIMAL TISSUE

M. J. Cook F. G. Karioris[11]
K. Z. Morgan

A previously reported study on continuous and single internal exposures of mice to Co^{60} has shown that single-exposure data may be used to obtain approximate maximum permissible values for continuous exposure.[12] An experiment similar to the Co^{60} study is in progress in which Sr^{90}-Y^{90} is administered orally to DBA-strain mice. In the first group of approximately 200 mice each mouse is given a single 10-μc dose, and for the second group of approximately 200 mice the drinking water contains the radioisotope in a concentration of 1 μc/ml.

It is believed that when a hanging-drop bottle is used much water is wasted by dripping, evaporating, and the mice playing in it. A capillary drinking-water fountain was designed which prevented dripping, kept evaporation to a minimum, and prevented the mice from playing in the water, and this fountain is under test at the present time.

ISOTOPIC DISTRIBUTION IN MAN

I. H. Tipton M. J. Cook
D. K. Bowman J. C. Gallimore

Spectrographic Analysis of Human Tissue

The tissue-collection program has been established in Denver, Dallas, Miami, Atlanta, and Baltimore. All the samples have been received from Denver, but tissue is continuing to be received from the other four cities.

Medical examiners or coroners in St. Louis, Cleveland, New Orleans, Seattle, Tacoma, and Richmond have sent letters offering assistance.

[11] Summer research participant from the Physics Department, Marquette University, Milwaukee, Wis.

[12] M. J. Cook, K. Z. Morgan, and A. G. Barkow, "An Experiment Designed to Test the Validity of the Current Practice of Using Single-Exposure Data to Calculate Maximum Permissible Concentration in Water for Continuous Exposure to Radioisotopes," to be published in *Am. J. Roentgenol., Radium Therapy Nuclear Med.*

The wet-ashing procedure has been replaced by the dry-ashing procedure, which is a more rapid process. By the dry-ashing procedure approximately 50 samples per week may be prepared for spectrographic analysis.

The spectrographic laboratory for the analysis of human tissue at the University of Tennessee has refined the quantitative methods for 29 elements in soft tissue and 21 elements in bone. Analyses are being made at the rate of 40 samples per week.

The results of analyses of 61 bone samples are given in Table 5. These samples included those received from Boston, New York, Birmingham, Columbus (Ohio), and Knoxville from April 1951 through September 1954.

TABLE 5. SPECTROGRAPHIC ANALYSIS OF BONE ASH

Number of Samples: 62
Mean Per Cent of Ash from Bone: 17

Element	Limit of Sensitivity (ppm)	Coefficient of Variation (%)	Total Number of Samples Containing Element	Number of Samples Containing Trace Amounts	Mean Concentration (ppm)	Range (ppm)	Remarks
Al	25		62	31		260–<25	
Cr	4		28	13		27–<4	
Mn	1		45	31		8–<1	
Fe*	100	±15	46		1428	3000–100	
Ni	25		6	5		29–<25	
Cu	1	±15	62		8**	>300–3	Two samples contained >300 ppm
Sr	10	±15	62		100***	1000–10	One sample contained >>1000 ppm
Ag	2		55	5		11–<2	
Sn	15	±20	62		49	88–30	
Ba	1	±15	62		13	215–4	
Pb	10	±15	62		56	200–10	
No other elements detected							

*Only 46 samples analyzed for Fe.
**Two high values not included in mean.
***One high value not included in mean.

SANITARY-ENGINEERING RESEARCH

E. G. Struxness

FIELD INVESTIGATIONS AND EXPLORATIONS (LIQUID-WASTE DISPOSAL)

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Water Decontamination Study

Activated carbon is usually a good agent for removing traces of color, tastes, or other undesirable substances from water. However, activated carbon alone is ineffective for the adsorption of ionically dispersed metallic compounds. Ion-exchange methods can be used effectively for this purpose. Trace metals can be removed from solutions if an adsorbable radical is first attached to the metal ion and then the activated carbon is employed to remove the resulting metallic organic complex. This observation indicated that the removal of radioactive cations from water by this method should be investigated.

To 2.5 liters of Oak Ridge tap water (pH 7.8, hardness 114 ppm as $CaCO_3$) a sufficient amount of the particular radioisotope under investigation was added to give a reasonable counting rate (about 3000 counts/min/ml). Analytical-grade dimethylglyoxime, in weights of 0.5, 1.0, 2.5, and 5.0 g, was added to 800-ml glass beakers. A fifth beaker, containing no organic material, was used as a control. In order to wet the powdered dimethylglyoxime, 1 ml of 0.5% aerosol solution was added to each beaker. Samples of the contaminated tap-water solution were taken for chemical and radiochemical analysis. Five hundred milliliters of the solution was then added to each of the 800-ml beakers, and the beakers were stirred for 30 min at about 250 rpm. Fifty-milliliter samples were withdrawn from each beaker at regular 10-min intervals and filtered through a 6-in. depth of technical-grade granular bone charcoal. Three 1-ml samples of this filtered liquid were placed in aluminum counting dishes, dried under infrared heat lamps, and counted by using a G-M mica-end-window tube and 64 scaler.

After 30 min of stirring, the fourth set of samples was removed, and 50 ml from each beaker was added to five 50-ml Lusteroid tubes and centrifuged. The supernatant liquid from each tube was prepared and counted in the described manner, and its pH was determined by using a Beckman model G glass-electrode pH meter.

All glassware used in the experiment was precoated with Desicote.[1] Desicote is an organosilicon compound which removes even the slightest trace of water from silicate surfaces and leaves a water-repellent coating of molecular thickness. This coating protects the glassware from cross-contamination by the various radioactive materials used and prevents abnormally high removals of certain radioisotopes due to plating-out on the surfaces of the glassware.

The data are given in Table 6. Larger amounts of organic material resulted in increased removals of the radioisotopes. When no organic material was added, the process was simple surface adsorption by the activated charcoal.

For 6 of the 11 cationic radioisotopes studied, the removals obtained were greater than 99% when 10 g of dimethylglyoxime per liter had been used. Three of the remaining radioisotopes were removed in excess of 98%, and only Na^{24} showed low removals (63% maximum obtained).

The detailed results indicated that increased contact time does not result in significantly higher removals; for example, Ce^{144}-Pr^{144} and Na^{24} were removed almost as efficiently after 10 min as after 30 min of stirring.

The principle involved is the complexing of the radioactive cation and then, by using carbon, the adsorbing of both the isotopic complex and any excess organic reagent. Because the radioactive contaminants represent a concentration too low to obey the law of mass action, it was necessary to use larger-than-stoichiometric proportions of the organic material.

Other complexing organic reagents may also be used for this particular type of treatment; a Darco[2] article suggests mercaptobenzothiazole and xanthates. The article also states that chelating agents do not seem to be effective, probably because they are strongly soluble and thus are not readily adsorbable.

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[1] Desicote is a product of the Beckman Instruments, Inc., of S. Pasadena, Calif.

[2] "Darco Digest," *Chem. Eng. News* 33(12), 1187 (1955).

TABLE 6. REMOVAL OF RADIOACTIVITY FROM CONTAMINATED WATER BY ADSORPTION OF ORGANIC COMPLEXES ON CARBON

Radioisotope	Initial Activity (counts/min/ml)*	Removal of Contaminant** (%) Amount of Organic Material (g/liter)					Microcuries per Milliliter in 10-g Sample
		0	1	2	5	10	
Sr^{89}	928	68.2	94.0	94.3	95.3	98.1	8×10^{-5}
Ba^{140}-La^{140}	8,330	59.3	67.1	69.7	90.0	98.6	5×10^{-4}
Na^{24}	3,840	38.3	44.2	52.2	52.8	63.9	6×10^{-3}
Sb^{124}	13,980	57.4	58.8	67.7	70.0	87.5	8×10^{-3}
Zr^{95}-Nb^{95}	6,580	64.4	85.5	86.9	89.2	98.4	5×10^{-4}
Ca^{45}	3,600	84.4	90.4	91.2	93.8	99.6	7×10^{-5}
Y^{91}	3,180	81.0	91.2	93.9	95.2	99.0	1×10^{-4}
Ag^{110}	2,690	89.2	93.7	95.8	97.0	99.3	9×10^{-5}
Ce^{144}-Pr^{144}	3,980	88.8	98.2	98.7	99.0	99.2	1×10^{-4}
Co^{60}	4,010	22.6	98.3	98.8	99.2	99.3	1×10^{-4}
Ru^{106}-Rh^{106}	3,870	34.5	81.2	90.9	95.3	99.0	2×10^{-4}

*The data for activity in counts per minute per milliliter were not corrected for geometry (~10%).

**After 30 min of contact time.

Whether the treated material meets the maximum permissible concentration values depends upon the initial concentration of the particular radioisotope.

Field Explorations

The 4-acre site[3] in Melton Valley is located on the contact of the bottom red noncalcareous member of the Conasauga shale and the next higher member composed of gray calcareous shale containing a few thin beds of limestone. Study of the drill cores, outcrops, and exposures made by trenching shows that under the site these beds are folded, forming a somewhat complex anticline. This may be an unfavorable feature, as a more uniform attitude of the beds might promote a more uniform movement through the ground of any seeping radioactive liquid.

[3] *HP Semiann. Prog. Rep. July 31, 1954*, ORNL-1763, p 16.

The several diamond-drill test holes and the 6-in. well were pressure-tested with water to locate depths at which water could move into or out of them. The top 20 or 30 ft of the holes were cased. The most permeable zone was found between the bottom of the casing and a depth of about 100 ft. Below that level the rock was less fractured, and the holes lost little water. The shale itself, where it is unfractured, has a very low permeability. Most of the ground-water movement is along a few individual fractures and not through the mass of the rock.

Water-stage recorders were installed on the wells, and the pattern of natural variation of water levels was studied. There was an easily observable difference between the wells; some showed a marked and rapid rise of water level following a rain, and some did not. Some of the wells showed small variations in water level due to earth tides or fluctuations in barometric pressure, and some

did not. This lack of hydrologic uniformity, and in particular the apparent concentration of fluid movement in certain channels rather than through the whole body of the well, is in itself unfavorable for waste disposal, for a pit depends for its effectiveness on the adsorptive capacity of the solid matrix of the aquifer. Several pumping tests were made, and the data from this study also showed a concentration of water movement along certain channels and at least a detectable difference in the response of the water levels in the several observation wells. As yet it is not possible to say how seriously the departure from hydrologic homogeneity might affect the movement underground of liquid waste; no large volume of shale can be expected to be uniform.

Waste-Storage Pits

Liquid Budget for Waste-Disposal Pits. – Liquid-waste-storage pit No. 3 was put into service early in the year. By June 30 the pit had been filled with waste and rainwater and had just begun to cascade into pit No. 2, which had received no pumped waste in five months and had dropped very materially in stage. Comparison of the rates of change of pit stage of the two pits shows that pit No. 2 is leaking more rapidly than pit No. 3 and that evaporation is an important factor, particularly in pit 3. The difficulty of accurately determining evaporation loss has shown the need for a more comprehensive study of this part of the budgeting. Two methods of approach appear to be possible, and both will probably be used. One method is to calculate the rate of evaporation from meteorological parameters; the other is to determine the rate of fall of stage during times of negligible evaporation – at such times the fall of stage must be largely due to seepage. Once seepage is determined, evaporation losses are obvious.

The more rapid seepage from pit 2 may be due to more permeable rock near it, or it may be that steeper water-table gradients have been developed out from it. Only part of the slope of the water table can be determined from the few observation wells available, and much remains to be done to explain the operation of the pits.

Evaluation of Proposed Site for Pit No. 4. – At the present rate of waste discharge there is need for at least one more disposal pit, and a site was chosen just south of pit No. 2. Pit 3 is just to the north of pit 2. It is proposed to pump all waste directly into pit 3, let any overflow cascade into pit 2, and allow overflow from pit 2 to cascade into the new pit, No. 4. The new location is at the end of the low, flat ridge on which the pits are located, and the land surface will slope away steeply from the edge of the pit, particularly to the east. The depth to ground water from the pit bottom will be of the order of 20 to 25 ft, somewhat more than in the two previous units. These factors alone would suggest a more rapid rate of seepage from the new pit, which would be desirable as far as the prime purpose of the pit is concerned, and, as far as is now known, quite safe. In order to analyze better the operation of the pit after it is in service, it is planned to make hydrologic tests, including pumping tests, of the area. The construction of the observation wells is already under way.

Vertical Definition of Radiochemical Waste Movement from Disposal Pits

To define the location of radioactive wastes that seep from a disposal pit, a method for determining the vertical configuration of the resulting contamination stream is necessary. The contamination profile is being evaluated by means of observation wells around the pit site, radiologging equipment,[4] selected-depth sampling, mixing of the well contents, and flushing out the well contents.

The gamma-ray log obtained would be influenced by (1) activity in the well water, (2) variations in the diameter of the observation hole, (3) sorption of activity on the side wall of the hole, and (4) sorption of activity in the rock or in solution, in the strata through which wastes are traveling. Radiologging followed by sampling is not sufficient to differentiate these effects. In Fig. 5*a* radiologging without disturbing the liquid contents of the well is referred to as a normal run.

Mixing the well contents to eliminate stratification of wastes was accomplished by pumping from the well bottom and discharging to the liquid surface. Radiologging following this mixing procedure is indicated as a recycle run (Fig. 5*b*) and is followed by sample collection. Fluctuations in probe response due to variations in hole size were eliminated by flushing out the well contents. The contaminated liquid was pumped from the bottom,

[4] *HP Semiann. Prog. Rep. Jan. 31, 1954*, ORNL-1684, p 25.

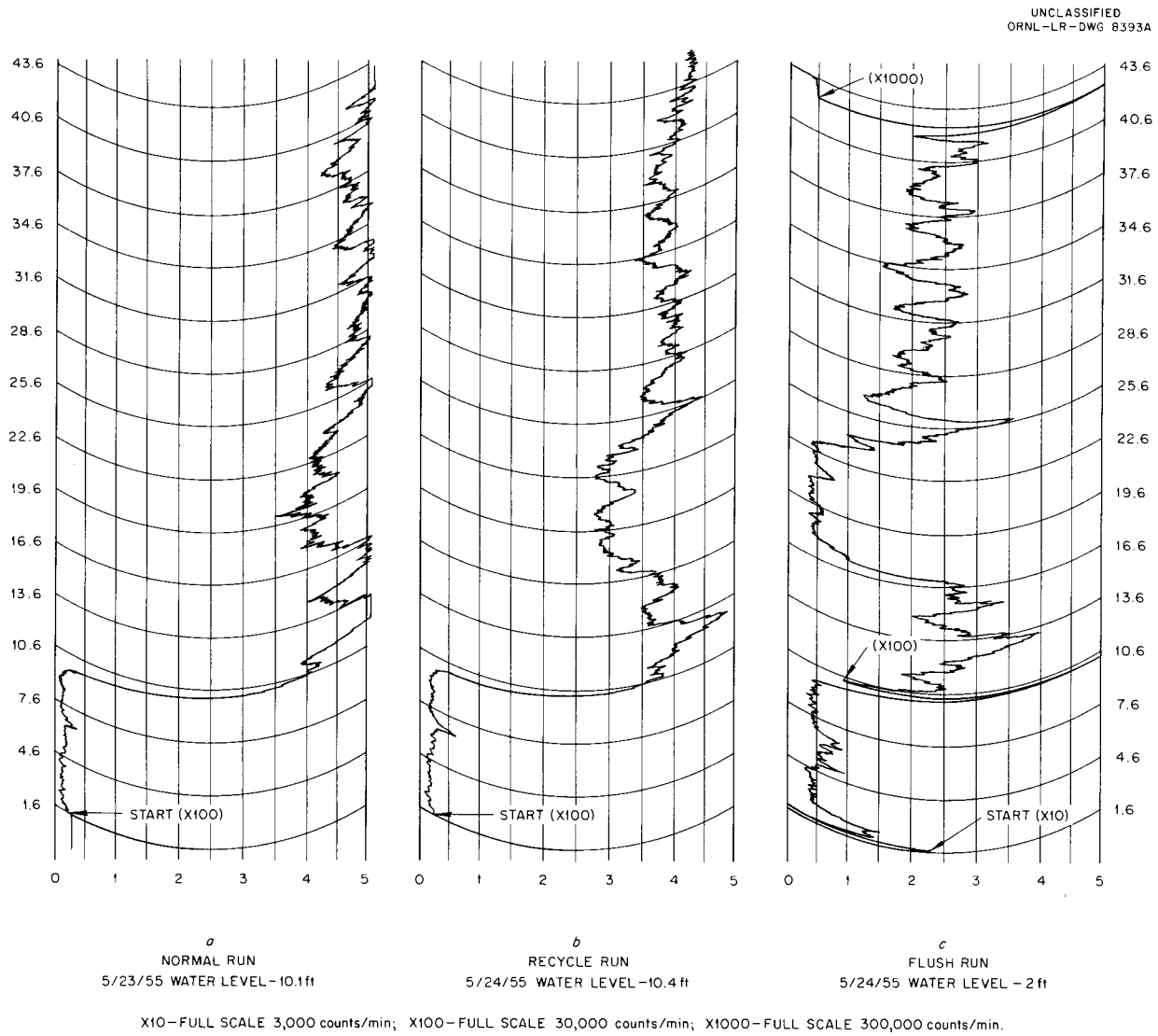

Fig. 5. Radiolog Charts; Well NX8. Line voltage, 115; tube voltage, 900; time constant, *MI*.

while a positive head of fresh water was maintained in the well. After flushing, a radiolog is obtained (Fig. 5*c*, flush run), the positive head of fresh water still being maintained. Samples were collected after each radiologging.

Reliability of the radiologging and sampling procedure has been shown with consecutive logs and duplicate samples. Analysis indicates that greater than 90% of the activity in the water is due to Ru^{106}-Rh^{106}.

The mixing procedure was effective in providing a uniform activity in the water. The location of maximum responses observed by radiologging remained unchanged after recycling. There was a lack of correlation between probe response and activity in samples collected. From the observations it appeared that the peaks in the radiolog charts were not a result of activity in the water. Flushing reduced the water activity to a negligible amount. Radiologging after flushing shows that peak responses remain at the same depths and substantiates the inference that the peaks are due to activity in the rock strata.

Hydrology and Geology of the Oak Ridge Area

Work on the report of the hydrology and geology of the Oak Ridge Area has consisted largely in transferring to maps of scale 1:24,000 the pattern of geologic formations shown on the recently compiled geologic maps of eastern Tennessee. Some information on land use has been compiled, and some diagraming of surface drainage patterns, as they might affect the movement of waste reaching the streams, has been completed. From what can be seen, it is clear that the areas now being used or being investigated are the best suited to waste disposal, and the report, when finished, will have value principally by confirming conclusions already reached.

A new topographic map of the area will be published at the end of July, and it is planned to use these quadrangles to prepare a new and better topographic base map of the government-owned area.

CHEMISTRY AND SOILS ENGINEERING

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Chemical Laboratory Studies

Procedures for the recovery from soil and identification of the major long-lived fission products have been evaluated. The efficiencies of the various leach solutions selected are indicated in Table 7.

These procedures were tested on 20 samples of river mud collected at various points below the Oak Ridge National Laboratory. The samples show that (1) only naturally occurring radioactivity was found in the mud from the Tennessee River, (2) the concentration in the mud of the Clinch River was approximately five times background, and (3) the concentration in the mud of White Oak Lake was about 1000 times that of the Clinch River mud. The predominant radioisotopes found were cesium, cobalt, and cerium. The methods of analysis will be published.

A technique was tested for analyzing the cobalt leached from soil to determine contamination by the major fission products; the procedure includes leaching with 1 *M* HNO_3 or HCl, precipitating with KOH, dissolving in acetic acid, precipitating with KNO_2, dissolving in HCl, scavenging with a precipitation of $Ce(OH)_3$ in basic NH_4Cl, precipitating with α-nitroso-β-naphthol from HCl solution, and igniting to Co_3O_4. The recovery of tracers, corrected for final yield of carrier, is reported in Table 8.

TABLE 7. RESULTS OF LEACHING EXPERIMENTS FOR THE RECOVERY OF ADSORBED RADIOISOTOPES FROM NO. 1 ORNL CLAY-TYPE SOIL

Radioisotope	Leach Solution	Recovery (%)
Cs^{137}	9 *M* H_2SO_4	98.8
Sr^{90}	0.5 *M* HNO_3	99.6
Sr^{90}	1.0 *M* HNO_3	99.8
Y^{90}	0.5 *M* HNO_3	98.6
Y^{90}	1.0 *M* HNO_3	99.6
Ru^{106}	1 *M* HCl + 0.2 g $KMnO_4$	98.2*
Zr^{95}	9 *M* H_2SO_4 + 0.5 *M* $H_2C_2O_4$	99.7
Nb^{95}	1.0 *M* HCl + 0.5 *M* $H_2C_2O_4$	99.6
Co^{60}	1.0 *M* HNO_3	97.7
Co^{60}	1.0 *M* HCl	99.8

*In distillate.

To obtain inexpensive simultaneous removals of the major long-lived fission products from an acid $Al(NO_3)_3$ solution by precipitation, the following procedure has been devised:

1. Boil 100 ml of solution. Add 200 mg of $Fe(CN)_6^{--}$, 60 mg of Ca^{++}, 32 mg of Zr_4^{+}, and 0.8 ml of 1:10 H_3PO_4. Boil and stir 5 min; filter.
2. Boil filtrate, add 1 g of Ba^{++}, and pour slowly into 100 ml of boiling water containing 8 g of Na_2SO_4. Boil and stir for ½ hr, and let settle.
3. Cool to approximately 24°F, dilute with 100 ml of H_2O, and add 15 mg of Ce^{+++}, 150 mg of $Fe(CN)_6^{--}$, 50 mg of Cu^{++}, and 1 g of $(NH_4)_2C_2O_4$. Stir in ice bath for 1 min, centrifuge, and decant.

The removals obtained are indicated in Table 9.

The removal efficiencies of the same radioactive tracers from 100 ml of acid $Al(NO_3)_3$ waste solution made basic with 80 ml of 18 *M* NaOH, with 100 mg of Ca^{++} and 100 mg of Na_2CO_3 added to carry the strontium tracer, are also listed, in the column headed "Caustic Solution."

TABLE 8. RECOVERY OF RADIOACTIVE TRACERS IN COBALT-ANALYZING PROCEDURE

Radioisotope	Activity on Soil (counts/min)	Activity in Leach Solution (counts/min)	Activity on Co_3O_4 Precipitate (Corrected for Carrier Yield) (counts/min)
Cs^{137}	546×10^4	116×10^4	0
Sr^{90}	42.3×10^4	40.3×10^4	0
Y^{90}	42.6×10^4	39.2×10^4	11
Ce^{144}	152×10^4	146×10^4	11
Ru^{106}	233×10^4	164×10^4	124
Zr^{95}-Nb^{95}	753×10^4	39.7×10^4	461
Co^{60}	384×10^4	349×10^4	341×10^4

TABLE 9. PER CENT REMOVAL OF FISSION PRODUCTS FROM AN ACID $Al(NO_3)_3$ SOLUTION

Radioisotope	Acid Solution (%)	Caustic Solution (%)
Cs^{137}	99.3	44.9
Sr^{90}	97.2	97.0
Y^{90}	86.3	99.0
Ce^{144}	99.3	98.3
Zr^{95}-Nb^{95}	99.5	91.5
Zr^{95}	100.0	88.0
Ru^{106} (2-month-old solution)	90.2	52.1

In both procedures, to determine the effect on removals of aging of the solution, the ruthenium carrier was added at least one month before the precipitation. It was found that the fraction of the ruthenium removed decreased with the age of the solution because of chemical instability of the ruthenium in the waste solution.

The cost of chemicals for treating the acid $Al(NO_3)_3$ solution was found to be 1.5 cents per liter and for treating the caustic waste (including the cost of NaOH), 5.0 cents per liter.

In studies on the effect of the concentration of cesium on its adsorption on local clays, it was found that the exchange capacity was a function of the concentration; a lowering of the cesium concentration resulted in a lower exchange capacity of the clays for cesium. To compare the adsorption of potassium on soil with that of the cesium reported above, known weights of irradiated and processed $K^{42}Cl$ were passed through columns containing 1 to 5 g of soil until the soil was saturated. Samples of the initial and final solutions and of the soil were counted to determine the amount of K^{42} adsorbed. Reducing the concentration of potassium from 52.3 to 0.0523 mg/ml reduced the amount of potassium adsorbed on the soil from 14.3 to 6.85 mg per 100 mg of soil.

Twenty AEC-Vanderbilt Fellowship students in training at the Laboratory are extending these studies to determine the effect of concentration on the adsorption of fission products on Conasauga shale.

Soils and Engineering Studies

Samples of semiweathered rock material from the 4-acre site have been obtained and their cation-exchange capacity and exchange complex determined. The cation-exchange capacity varies with the textural fraction, about 20 meq (milliquivalents) per 100 g for fractions between 10 and 80 mesh and dropping to 16 meq per 100 g for 6 to 8 mesh and for the finer-than-120-mesh fractions. The estimated total bases in the exchange complex comprise 4 to 6 meq per 100 g, and the H^+ ion 11 to 14 meq per 100 g by difference.

Permeability studies of natural rock materials from the 4-acre site were conducted. Cores were collected in such a manner that vertical, normal, and parallel-to-stratification permeabilities could be obtained. A vertical permeability (k_{20}) of

3×10^{-3} cm/sec and normal and parallel permeabilities of 1×10^{-3} and 4×10^{-3} cm/sec, respectively, were found.

Test sections of Tennessee ball clay and an asphalt membrane (along with test apparatus) were installed in waste pit No. 3 to determine the permeability and stability of the clay and asphalt liners under actual field conditions. There has been no detectable leakage through the ½-in. asphalt membrane; the clay liner is leaking to the extent of 10^{-7} cm/sec. The alkaline (pH = 10) $NaNO_3$ waste in the pit contains about 50% Cs^{137}, 40% $Ru^{103,106}$, and 1% Sr^{90}, with no significant variation of composition with depth. The material which leaches through the clay indicates approximately the same composition and level of activity.

The possibility of increasing the stability of the clay to weathering by diluting with sand has been tested on a laboratory model. Although the permeabilities of the clay-sand mixtures are comparable to the permeability of the undiluted clay (about 10^{-8} cm/sec), the most stable liner with respect to erosion was 100% clay.

Studies of strontium uptake by undisturbed core samples show a capacity of about 9 meq per 100 g of soil, as compared with values of about 23 meq per 100 g for 10- to 20-mesh screened samples of the same material. These values, which agree with the cation-exchange capacity of these fractions as determined by the ammonium acetate method, show the inefficiency of the undisturbed rock material for adsorption of cations as compared with the potential capacity. Of the textural fractions examined, the 6- to 8-mesh portion most closely approximates the ability of natural cores to exchange strontium. Cores saturated with tagged strontium were broken open, and activity was found to have concentrated along the bedding planes and crevices, indicating that at the flow rates studied (k_{20} approximately 5×10^{-4} cm/sec) exchange capacity of the entire rock fragments was not utilized.

Tests of the ability of semiweathered shale rock to neutralize acid aluminum nitrate solution show that 150 kg of shale is required to raise the pH of 1 liter of solution from less than 1 to 1.5; above the latter point, increased soil contact appears to have little if any effect.

By using tap water and acid aluminum nitrate solutions, the permeabilities of soil were determined. No consistent relationship between water flow rates and acid-waste flow rates through the same cores was observed in the five cores tested. This may be due to the chemical nature and the density and viscosity of the waste solution.

Studies of uptake by shale of critical radioisotopes from a solution of distilled water and mixed fission products have been partially completed. The solution contains carriers in concentrations predicted for an acid aluminum nitrate waste. A series of tests, conducted at pH 3.5, has been completed, and data indicate that the order of removal through shale columns from this solution is Nb > Y > Zr > Cs > Ru > Sr. A soil-to-solution ratio of about 0.5 g to 1 ml is sufficient to retain essentially 100% of the fission products in the solution. A second series at pH 5.0 shows the same order of removal as obtained in the first series, but a soil-to-solution ratio of only 0.3 g to 1 ml is required to remove all the fission products from solution at this pH. Further studies are under way at pH 1.5, and preliminary results indicate that the soil-to-solution ratio required for complete retention on the soil at this pH may be larger by an order of magnitude than the ratio required at the higher pH values tested.

Soil Fertility and the Uptake of Strontium by Bean Plants

In order to establish (1) the relationship between strontium and the other elements of soil fertility, especially calcium, on the nutrient-supplying power of the soil, (2) the relative uptake of Sr^{90} when the amount of carrier strontium is varied, (3) the distribution of these elements within the various portions of the plant, and (4) the uptake of the elements throughout the growth period, a pot-culture experiment was initiated in which the Conasauga shale was used as the medium and the garden bush bean as the indicator plant. This soil, being relatively infertile, was brought to 100% of its cation-holding capacity as follows: 5% potassium, 15% magnesium, and 80% calcium. The calcium was successively replaced with strontium to give seven levels or ratios of calcium to strontium. In addition, the soil was treated with 240 lb of nitrogen, 270 lb of phosphorus, and 120 lb of sulphur per acre.

Each culture contains 3 kg of dry soil into which five bush beans were seeded. Sufficient cultures were established to allow for five harvesting dates at approximately 12-day intervals and to allow

statistical analysis of the data. To date, two harvests have been completed, and analyses of the soils and plant parts have been initiated.

AIRBORNE-RADIOACTIVITY STUDIES

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A detailed study of the efficiency of one type of sand as an entrainment separator (aerosol filter) has been made. The sand was divided into five fractions: 8 to 12, 12 to 16, 16 to 20, 20 to 30, and over 30 mesh. Each fraction was tested with each of five dioctyl phthalate aerosols having radii of 0.20, 0.26, 0.41, 0.64, and 0.84 μ at linear flow rates varying from 0.05 to 2.0 cm/sec. In general, the data for upflow through the four larger sizes of sand were fitted to within ±25% by the following equation:

$$1000kD_g = (1 - \alpha)\,\frac{1 + 0.087/r}{r}\,(1.5V^{-0.50} + 12r^3V^{-0.90}) ,$$

where

$$k = -\frac{\ln F}{b} ,$$

F = fractional aerosol penetration, dimensionless,

b = height of sand column, cm,

D_g = average diameter of sand granule, cm,

α = void fraction of sand, dimensionless,

r = radius of aerosol particle, μ,

V = linear velocity through bed $= Q/A$, cm/sec,

Q = volumetric flow through bed, cm^3/sec,

A = cross-sectional area of the empty sand holder, cm^2.

The form of this semiempirical equation was derived by S. H. Jury of the University of Tennessee. Over the range for which the equation was fitted, the size for maximum penetration is independent of sand granule size and varies monotonically from a 0.3-μ radius of aerosol particle at V = 0.0545 cm/sec to a 0.45-μ radius at V = 2.18 cm/sec. The calculated performance of a column of sand 1 ft (30 cm) thick, at the aerosol size for maximum penetration, is given in Table 10 as a function of the linear velocity through the sand (V) and the size fraction of sand.

TABLE 10. SAND-COLUMN EFFICIENCY AT AEROSOL SIZE FOR MAXIMUM PENETRATION

Air Velocity, V (cm/sec)	Fractional Aerosol Penetration, F			
	8- to 12-mesh Sand	12- to 16-mesh Sand	16- to 20-mesh Sand	20- to 30-mesh Sand
0.0545	0.017	0.0032	0.0003	0.00001
0.109	0.076	0.026	0.0057	0.0007
0.218	0.196	0.101	0.038	0.0096
0.545	0.415	0.287	0.169	0.079
1.09	0.565	0.449	0.320	0.198
2.18	0.697	0.600	0.487	0.357

RADIATION DOSIMETRY

G. S. Hurst R. H. Ritchie

EXPERIMENTAL PHYSICS OF DOSIMETRY

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Beta Spectrometer

Further data have been obtained on the distribution of energy losses in a beam of 328-kev electrons after passage through thin foils. Under high resolution (0.2%) slight discrepancies appear when the theories of Landau and of Bluch-Leisegang are applied. These discrepancies are probably due to approximations made in the theories. In an effort to obtain better data, a smaller counter having a much lower background than the present counter has been developed. When reliable operation is secured, a more precise investigation of the disagreements will be made.

Accelerator

The data on the excitation of the conduction electron plasma in metals by the passage of a high-energy electron have been evaluated and compared with the theory of Pines and Bohm. For aluminum, agreement with the theory is found, with respect to both the excitation energy and the mean free path for the interaction. The excitation energy in magnesium is also found to check with theoretical expectations although the experimental mean free path disagrees with theory, possibly due to uncertainties in the thicknesses of the readily oxidized magnesium foils. Data on copper and silver fail to show the characteristic sharp absorption peaks indicative of plasma behavior, and it is believed that if the plasma effect exists in these metals it must be strongly modified by the interaction with inner-shell electrons. A report has been submitted to *The Physical Review* embodying the results of this research, and the project will be temporarily discontinued in expectation of further theoretical developments.

Measurements on the distribution of dose as a function of depth for 100-kev electrons striking a slab of aluminum have been completed. Attempts are being made to correlate results with a new electron-diffusion theory advanced by Spencer.[1] The pumping system and the electron gun on the accelerator have been modified to facilitate more rapid taking of data by using a new ion chamber.

Construction of an ion source for the accelerator has been started as an initial step in a program of study of the ionization of gases by heavy ions. Ion energies will be of the order of those found from neutron recoils, and the principal purpose of the experiment will be to measure the average energy required to create an ion pair, a quantity of importance in neutron dose studies.

X-Ray Exposure Facility

Several exposures of various test film badges were made for the Applied Health Physics Section. Exposures were also made to test the energy dependence of a scintillation counter to be used for the river surveys. Several quartz oscillator crystals and samples of pure and impure fused silica were given a series of long exposures at high dose rates for projects in this division and the Solid State Division.

THEORETICAL PHYSICS OF DOSIMETRY

J. Neufeld R. H. Ritchie
W. S. Snyder

Disordering of Solids by Neutron Radiation

By using the scattering theory of Bohr, the number of interstitials produced in a monoatomic solid by neutrons has been calculated. It was found that for neutron energies such that ionization could be neglected, half the energy was expended in producing interstitials. For higher energies the fraction of energy spent in this way was found to be much smaller.

The paper has appeared in *Phys. Rev.* **97**, 1636–1646 (1955).

A Note on the Half Life of Fission Products Produced by Uniform Irradiation in a Reactor

According to Way and Wigner,[2] the energy released in unit time by fission products produced

[1] L. V. Spencer, *Phys. Rev.* **98**, 1597 (1955).

[2] K. Way and E. P. Wigner, *Phys. Rev.* **73**, 1318 (1948).

at time $t = 0$ varies roughly as $t^{-1.2}$, where t is the time since the fission occurred. While this formula does not represent the rate of energy release very precisely, it does give an order-of-magnitude estimate which is valid for times from a few seconds after the fission has occurred to times of the order of several years. Actually the exponent varies and seems generally to increase as the period of time lengthens. If a reactor is operating at a constant power level, the mixture of fission products which has accumulated at the end of a certain period of time will decay at a different rate than will the products formed from a single fission. The problem is to estimate the half life of such a mixture at any time subsequent to the period of irradiation.

Assume that fissions are occurring at a constant rate in a material during the time interval $0 \leq t \leq t_1$. Then the energy released by these fission products per unit of time at time $t_1 + t_2$ is given by

$$(1)\quad \int_0^{t_1} A(t_1 + t_2 - t)^{-1.2}\, dt = \frac{A}{0.2}\left[\frac{1}{t_2^{0.2}} - \frac{1}{(t_1 + t_2)^{0.2}}\right],$$

where A is a constant depending on the units chosen, as well as upon the conditions of the irradiation.

If the rate of energy release at time $t = t_1 + t_3$ is to be a fraction p of the rate of energy release at time $t = t_1 + t_2$, then t_3 satisfies the equation

$$(2)\quad p\left[\frac{1}{t_2^{0.2}} - \frac{1}{(t_1 + t_2)^{0.2}}\right] = \frac{1}{t_3^{0.2}} - \frac{1}{(t_3 + t_1)^{0.2}}.$$

Taking t_1 as the unit of time, this becomes

$$(3)\quad p\left[\frac{1}{x^{0.2}} - \frac{1}{(x + 1)^{0.2}}\right] = \frac{1}{y^{0.2}} - \frac{1}{(y + 1)^{0.2}},$$

where x and y represent the lapses of time after the end of the period of irradiation in units of the period of irradiation, that is, $x = t_2/t_1$ and $y = t_3/t_1$.

By graphing the function

$$\frac{1}{x^{0.2}} - \frac{1}{(x + 1)^{0.2}},$$

the solution of Eq. 3 can be read from the graph (Fig. 6). This has been done for $p = \frac{1}{2}$, and the value of $y - x$ has been plotted as a function of x. Here $y - x$ represents the additional period of waiting required for the activity at time x to be decreased by half.

It is found that

$$y - x \cong 0.78x + 0.4\sqrt{x},$$

the fit being well within the limits of accuracy of the underlying assumptions for $0.01 \leq x \leq 1000$ but being about 40% too high at $x = 0.001$.

When the data as given by Wheeler[3] are used, Table 11 will give some idea of the accuracy to be expected from the treatment.

This paper has been issued as ORNL CF-55-4-104 (April 28, 1955).

Passage of Charged Particles Through Plasma

The electronic plasma has been treated as a dispersive medium characterized by a dielectric constant which is a function of both the frequency and the wave number of the disturbance. The passage of charged particles through the plasma was investigated by means of this approach, and formulas for the stopping power for low- and high-velocity particles have been derived.

This report has been published in *Phys. Rev.* 98, 1632–1642 (1955).

On the Density Effect

A rigorous justification was given for the statement by A. Bohr that for an incident particle of relativistic velocity the electrostatic effect of the surrounding electrons is negligible and the main factor in screening is due to electromagnetic interaction.

This paper is to be published in *Phys. Rev.* with the title "Density Effect in Ionization Energy Loss of Charged Particles."

[3] C. D. Coryell and N. Sugarman (eds.), *Radiochemical Studies: The Fission Products*, NNES-IV, vol 9, Book 1, p 349 (1951).

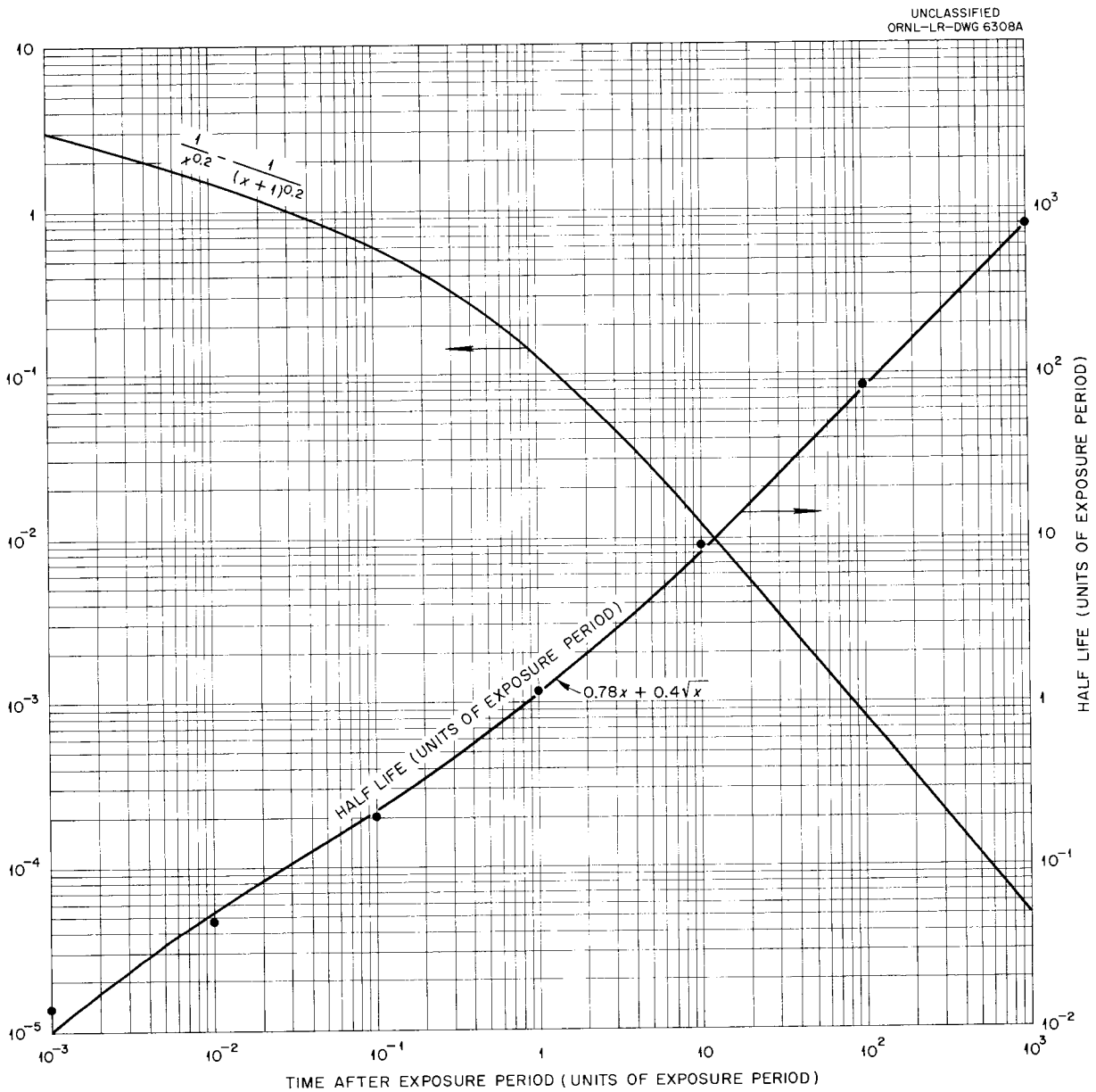

Fig. 6. Half Life of Fission Products Produced by Uniform Irradiation in a Reactor.

On the Number of Vacancies Created by Heavy Corpuscular Radiation

Kinchin and Pease[4] have suggested that a moving atom which collides with and displaces another atom may, in certain cases, take its place. Thus the number of displaced atoms is not the same as the number of vacancies, even when diffusion of interstitial atoms is neglected.

Let E be the energy of the striking atom before a collision and $E - y$ its energy after the collision. Altering the assumptions of Kinchin and Pease, it is assumed that if the struck atom receives energy y the probability of displacement is $p(y)$ and that if displacement occurs and if the energy of the striking atom after the collision

[4] G. H. Kinchin and R. S. Pease, *J. Nuclear Energy* **1**, 200–202 (1955).

TABLE 11. HALF LIFE OF FISSION PRODUCTS AT SUBSEQUENT TIMES AFTER IRRADIATION FOR VARIOUS PERIODS OF REACTOR IRRADIATION

Period of Irradiation, t_1 (days)	Cooling Period, t_2	Additional Time to Reach Half Value, $t_3 - t_2$	$(0.78x + 0.4\sqrt{x})\cdot t_1$ Period of Irradiation, $x = t_2/t_1$
180	1 day	17 days	6.1 days
180	10 days	30 days	24.8 days
10	1 hr	5 hr	7.0 hr
10	10 days	17 days	11.8 days
1	1 hr	2.2 hr	2.7 hr
1	10 days	16 days	9.1 days
1	100 days	60 days	82 days

is $E - y$ then the probability of the atom replacing the struck atom is $q(E - y)$. In general, $p(y)$ would be expected to be zero for small y and to increase to 1 at some higher energy, while $q(E - y)$ should decrease and become zero when $E - y$ is large.

Let $g(E)$ equal the mean number of vacancies created by the ejection of an atom of the medium whose energy after ejection is E. If $p(y) = 0$ for $y < \alpha$, then $g(E) = 1$ for $E < \alpha$. For $E < \alpha$, $g(E)$ satisfies the equation

$$(1)\quad g(E) = \int_0^E dy\, K(E,y)\,[p(y)g(y - \alpha) + g(y) - p(E - y)q(y)g(y)]\ ,$$

where $K(E,y)\,dy$ is the probability that a striking atom with energy E will lose energy y in dy in its next collision. The derivation of this equation follows closely the similar discussion of Eq. 7 in a previous paper of the authors[5] and assumes that ionization can be neglected. In accordance with the theory given[5] it should apply only for $E < Me^4/2\hbar^2$.

For low energies "hard sphere" scattering is assumed and $K(E,y)$ is taken as $1/E$. If $p(y) = 0$ for $y < \alpha$ and $p(y) = 1$ for $y > \alpha$ and if $q(y) = 1 - p(y)$, then by the methods developed[5] it is shown that

$$(2)\qquad \frac{E + \alpha}{3\alpha} < g(E) < 1.056\,\frac{E + \alpha}{3\alpha}$$

for all monoatomic substances and for $2\alpha < E < \beta$. The value β indicates the energy above which Rutherford scattering is assumed to apply and is given by $\beta = 2Z^2e^2/a$, with a the Bohr radius of the atom (ref. 5, p 1638). For Rutherford scattering, $K(E,y) = \beta^2/4Ey^2$ for $E\beta^2/(4E^2 + \beta^2) < y < E$, and $K(E,y) = 0$ otherwise. It is established that the inequality (Eq. 2) is valid for all $Z > 10$ for the region of Rutherford scattering and, with minor modifications, for lower Z.

The results for $E > Me^4/2\hbar^2$ are not complete but indicate a less rapid rate of increase for $g(E)$ due to ionization.

Thus the number of vacancies seems to be about two-thirds the number of displacements as calculated in the previous paper.[5]

Harrison and Seitz[6] have calculated the change in resistivity of copper irradiated by 17-Mev deuterons. By assuming that the number of displaced atoms was equal to the number of vacancies, they found that the calculated value was 5 times the observed value. The calculation of vacancies indicated above reduces this to 3.3 times the observed value.

The equation is being studied for cases when p and q change continuously from 0 to 1, and preliminary results indicate that the number of vacancies is much less than that given by Eq. 2.

[5] J. Neufeld and R. H. Ritchie, *Phys. Rev.* **97**, 1637–1646 (1955).

[6] W. A. Harrison and F. Seitz, *Bull. Am. Phys. Soc.* 30(2), 7 (1955).

Depth Dose Curves for Neutrons of Intermediate Energy

Depth dose curves for beams of neutrons normally incident on a 30-cm slab of tissue have been obtained by the Monte Carlo method. The neutron energies chosen were 100, 20, 5, and 0.1 kev. The energy absorbed from protons, heavier recoils, and gamma rays was tabulated separately. A dose curve was obtained by taking the RBE of protons and of heavy recoils as being 10 and 20, respectively, and gives a rem dose well in line with values obtained by interpolating in an earlier study.[7] The crude data will be smoothed and an error estimate will be calculated before the final release of the study.

Variation of Neutron Damage with Energy and Geometry

In connection with work on heavy particles for Subcommittee 4 of the National Committee on Radiation Protection, a preliminary study has been made of the variation of neutron dose with energy and with geometry of source and absorber. The discussion here attempts to give an indication of the trend and an order-of-magnitude estimate of the variation of damage.

It will be convenient to analyze the damage at any given site into (1) the "first-collision damage," which is the damage occasioned by the energy absorbed from the atoms with which the neutrons have their first collisions, (2) the "buildup damage," which is produced by recoils struck by the neutron in later collisions, and (3) the "absorption damage," due to energy released when a neutron is absorbed. The advantage of this way of analyzing the damage is that the first-collision damage is relatively easy to calculate when the energy of the incident neutrons is known, and it is possible to give some indication of the variability of the buildup damage and absorption damage in certain practical situations.

For a homogeneous phantom the first-collision dose is given in rem for a monoenergetic beam source by

$$(1) \qquad IEe^{-\sigma T}\left[\sum_i \sigma_i R_i \frac{2M_i}{(M_i+1)^2}\right],$$

where I is the intensity of the source (neutrons/cm^2), E is the initial energy of the neutrons, σ_i is the macroscopic cross section of the ith type of atom in the phantom, M_i is its mass number, R_i is the RBE for such a collision, $\sigma = \sum_i \sigma_i$ with all summations over the constituent elements of the phantom, and T is the thickness of phantom material a neutron must penetrate to reach the site in question. For an isotropic point source, it is necessary only to multiply by the factor $\frac{1}{4}\pi R^2$, where R is the distance from the source to the site of damage, and to take I as the number of neutrons emitted by the source. The values of the R_i can be obtained from other sources.[8,9]

At the irradiated surface of the phantom the buildup damage never exceeds the first-collision damage for any of the beams which have been calculated by Snyder and Neufeld.[10] Table 12 gives these values.

It should be emphasized that these figures are for damage at the irradiated surface, as the buildup dose is of much greater importance at sites farther below the surface. For the energy range indicated in Table 12, it is true, however, that the maximum damage can be taken as occurring at the surface, and the absorption damage can be neglected without serious error. Thus the rule for fast neutrons, that is, energy 0.1 to 10 Mev, can be stated: *The maximum damage may safely be taken as not more than twice the maximum first-collision damage.* Moreover, *the damage does not vary by more than a factor of 2 over the irradiated surface of the phantom.* When the size and shape of the phantom are varied, the first-collision damage remains unchanged at a point on the irradiated surface, and the buildup damage may be expected to decrease as the size of the phantom decreases. It therefore seems to be a safe rule that *the maximum damage from fast neutrons will not vary by more than a factor of 2 as the size of the phantom varies.* For example, the maximum damage received by a man and by a mouse subjected to the same beam of fast neutrons should

[7] W. S. Snyder and J. Neufeld, *Calculated Depth Dose Curves in Tissue for Broad Beams of Fast Neutrons*, ORNL-1872 (May 5, 1955).

[8] W. S. Snyder and J. Neufeld, *Proposed Program for Computing the Rem Dose Due to Irradiation by Fast Neutrons* (to be published; available from the authors).

[9] *Permissible Dose from External Sources of Ionizing Radiation*, p 48, NBS Handbook 59 (Sept. 24, 1954).

[10] W. S. Snyder and J. Neufeld, *Calculated Depth Dose Curves in Tissue for Broad Beams of Fast Neutrons*, ORNL-1872 (May 5, 1955).

TABLE 12. DAMAGE AT IRRADIATED SURFACE OF PHANTOM BY NEUTRONS OF VARIOUS ENERGIES

Initial neutron energy, Mev	10	5	2.5	0.5	0.1
First-collision damage at surface, rem/neutron $\times 10^{-8}$	5.8	5.0	4.0	1.9	0.68
Total damage at surface, rem/neutron	7.7	7.7	5.2	2.4	0.80

TABLE 13. ABSORPTION DAMAGE BY INCIDENT NEUTRONS OF VARIOUS ENERGIES

Energy of incident neutrons, Mev	10	5	2.5	0.5	0.1	0.005	10^{-4}	2.5×10^{-8}
Maximum absorption damage, rem/neutron $\times 10^{-9}$	0.7	0.8	1	1	1.2	1.3	1.4	1
Depth below surface where maximum occurs, cm	12	11	9	6	6	5	3	0.3

not differ by more than a factor of 2, and the variation of damage over the irradiated surface of either should not vary by more than a factor of 2. This rule is intuitively plausible if it is recalled that in a medium where collisions with hydrogen predominate, the first collisions account for, roughly, half the energy carried by the neutrons.

For thermal or epithermal neutrons the absorption dose is preponderant, and little information is available concerning the variation of this damage with the geometry of the phantom. For the practical case of a large phantom, for example, the trunk or even the head of a human body, it is possible to state a simple rule: *The maximum absorption damage in a large phantom is essentially constant at about* 1×10^{-9} *rem/neutron. The site at which the damage occurs varies in depth,* as Table 13, compiled from other data,[10] indicates.

The above data apply strictly only to broad-beam sources, but similar rough limits on the variability apply to the damage from an isotropic point source if allowance is made for the effect of the inverse-square law. For example, the maximum damage caused by an isotropic point source of neutrons will not differ by more than a factor of about 2, regardless of the size of the animal exposed to the damage, provided that the distance from the source is kept constant. The variation of damage over the irradiated surface when adjusted to eliminate the dependence on the inverse-square law should not vary by more than a factor of about 2.

Depth Dose Curves for Neutrons with RBE as a Function of LET

The National Committee on Radiation Protection has recommended[11] that the relative biological effectiveness (RBE) be based on linear energy transfer (LET). As a member of Subcommittee 4, W. S. Snyder has been requested to compute depth dose curves for neutrons by using the RBE as specified in *Handbook 59*. The linear energy transfer of protons and heavy recoils has been obtained by the use of the curves computed by the authors in a previous report.[12] To simplify the program, tissue is considered to consist of hydrogen and a fictitious element with the composite cross section required for the carbon, nitrogen, and oxygen atoms of tissue and with the weight of oxygen. The rem dose delivered by a proton will be taken as a fourth-degree polynomial in the energy of the recoil, for example,

$$\sum_{i=1}^{4} p_i E^i ,$$

and similarly for the rem dose of a heavy recoil. Only the first four moments of the energy of the proton will be tabulated for each depth region, and similarly for heavy recoils. Thus any future changes of RBE values may be substituted without recalculation, provided that the rem dose can

[11] *Permissible Dose from External Sources of Ionizing Radiation*, p 48, NBS Handbook 59 (Sept. 24, 1954).

[12] W. S. Snyder and J. Neufeld, *On the Energy Dissipation of Moving Ions in Tissue*, ORNL-1083 (Nov. 29, 1951).

be taken as a fourth-degree polynomial in the energy. It is planned to include a slab of bone (one additional heavier element) of arbitrary location in the slab and of any desired thickness up to 5 cm. The amount of energy absorbed from nonionizing heavy particles will also be estimated. The code for the Oracle is substantially complete. It is hoped that computation can begin within a few weeks.

A memorandum with further details on the program was prepared for the use of Subcommittee 4 and is available.[13]

Nondirectional Count-Rate Neutron Dosimeter

An analytic expression has been derived for the number of protons ejected with energies greater than B from the walls into the interior of a spherical cavity lined with paraffin by incident neutrons of energy E. This expression has been employed in the design of a nondirectional neutron counter whose counting response simulates the dose-energy variation for soft tissue. Calculations will be made by evaluating the uncertainties introduced by heavy atom recoils and wall losses in the high-energy region.

Effect of the Polarization Field on Capture by Moving Ions in Dense Media

A method has been formulated by using the impact parameter approach, for evaluation of the effect of polarization of a dense medium by the distant field of a charged particle on the capture of electrons by the incident particles. With the use of this method, calculations of the cross section for capture of protons moving in liquid helium are being carried out.

DOSIMETRY APPLICATIONS

F. J. Davis, J. A. Harter, P. W. Reinhardt, J. M. Garner, J. A. Auxier

Background Survey with Aircraft[14]

Flights over a series of cross-country traverse lines were made in order to study the variations in normal background gamma radiation. The equipment used was the scintillation detector developed by this Laboratory for the use of the U. S. Geological Survey in uranium prospecting. This equipment consists primarily of six NaI(Tl) crystals, each 4 in. in diameter and 2 in. thick. The counting rate of the crystals was recorded by two Esterline-Angus recorders, one recording the actual count rate and the other recording the count rate corrected by the function $e^{-\mu h}$, where h is the height above ground and μ is the absorption coefficient for radium gamma rays in air. This function has been shown to fit the variation of gamma-ray intensity with height in a previous report.[15] The correction is effected by a servomechanism which follows the height above ground as given by a recording radioaltimeter and controls the sensitivity of the rate meter to compensate for the altitude variations.

Flights were made over traverse lines during the last week in January and the first week in February 1955. The first line was north from Lafayette, Louisiana, to Moline, Illinois. The second line was south from Wichita, Kansas, to San Antonio, Texas. The third line was north from El Paso, Texas, through Albuquerque, New Mexico, and Farmington and Grand Junction, Colorado.

A portion (from St. Louis, Missouri, to Moline, Illinois) of the record of the first traverse line flown is shown in Fig. 7. The records run from right to left. The top record is the altitude in feet above ground, the second is the radiation without altitude corrections, and the third is the radiation corrected for altitude variation. The third record also has the soft-cosmic-radiation component subtracted so that the record shows only the reading proportional to the radiation from the ground. It is to be noted that, while the radiation from the ground is quite uniform, the second record shows variation due to altitude, and these variations are the mirror image of the altitude, that is, when the plane is low the radiation goes up and when the plane is high the radiation goes down. At point a is shown the record of the crossing of the Mississippi River near St. Louis. The radiation from the water is negligible compared to that from the land, and so the third record drops to zero. The second record drops only to 200 counts/sec (10 counts/sec/division). This represents the count due to soft cosmic radiation.

[13] W. S. Snyder and J. Neufeld, *Proposed Program for Computing the Rem Dose Due to Irradiation by Fast Neutrons* (to be published; available from the authors).

[14] In cooperation with the U. S. Geological Survey.

[15] F. J. Davis, P. W. Reinhardt, and J. A. Harter, *HP Semiann. Prog. Rep. Jan. 31, 1954*, p 11.

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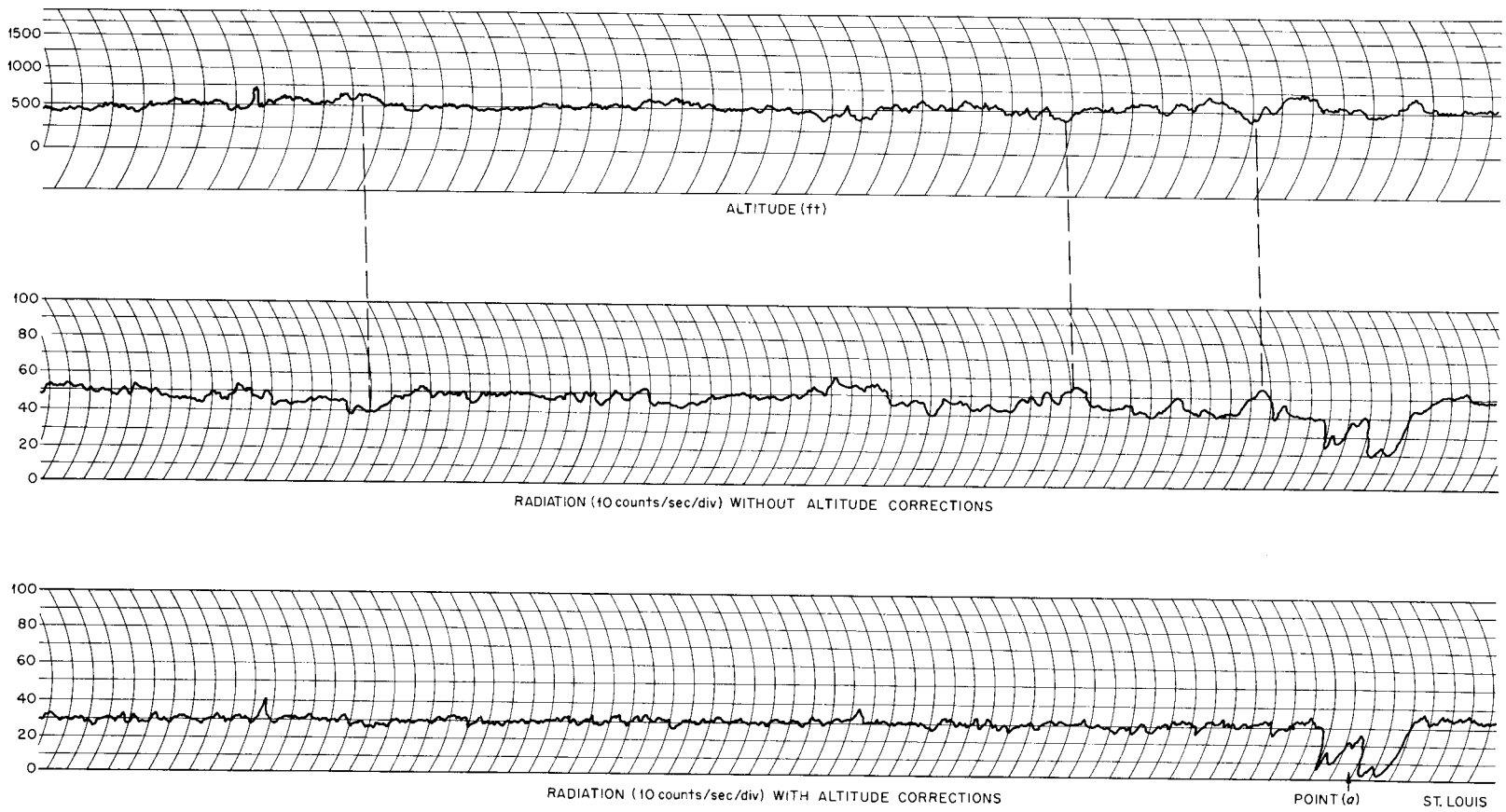

Fig. 7. Record of Radiation Measurement from St. Louis, Mo., to Moline, Ill.

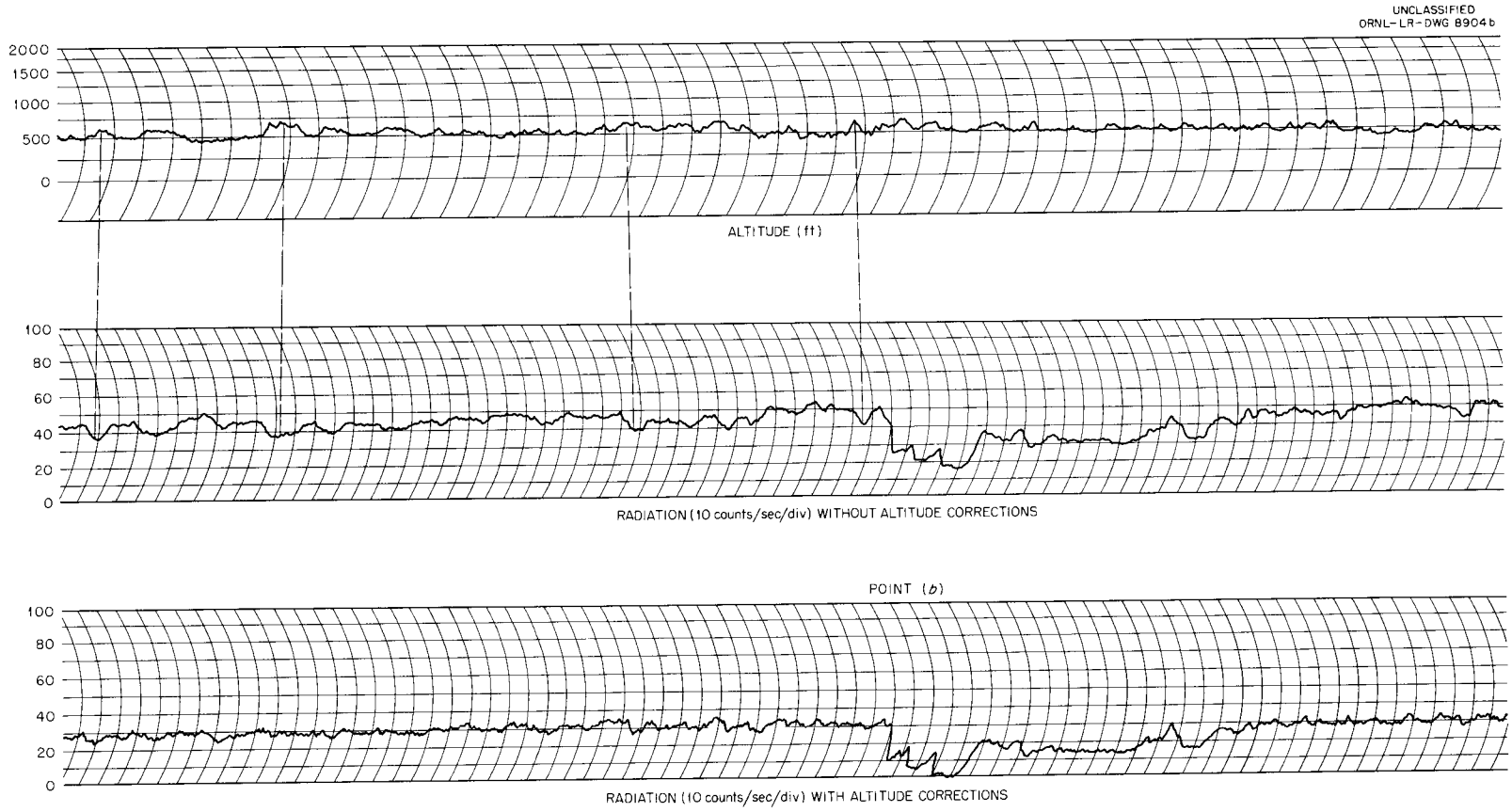

Fig. 7 (continued)

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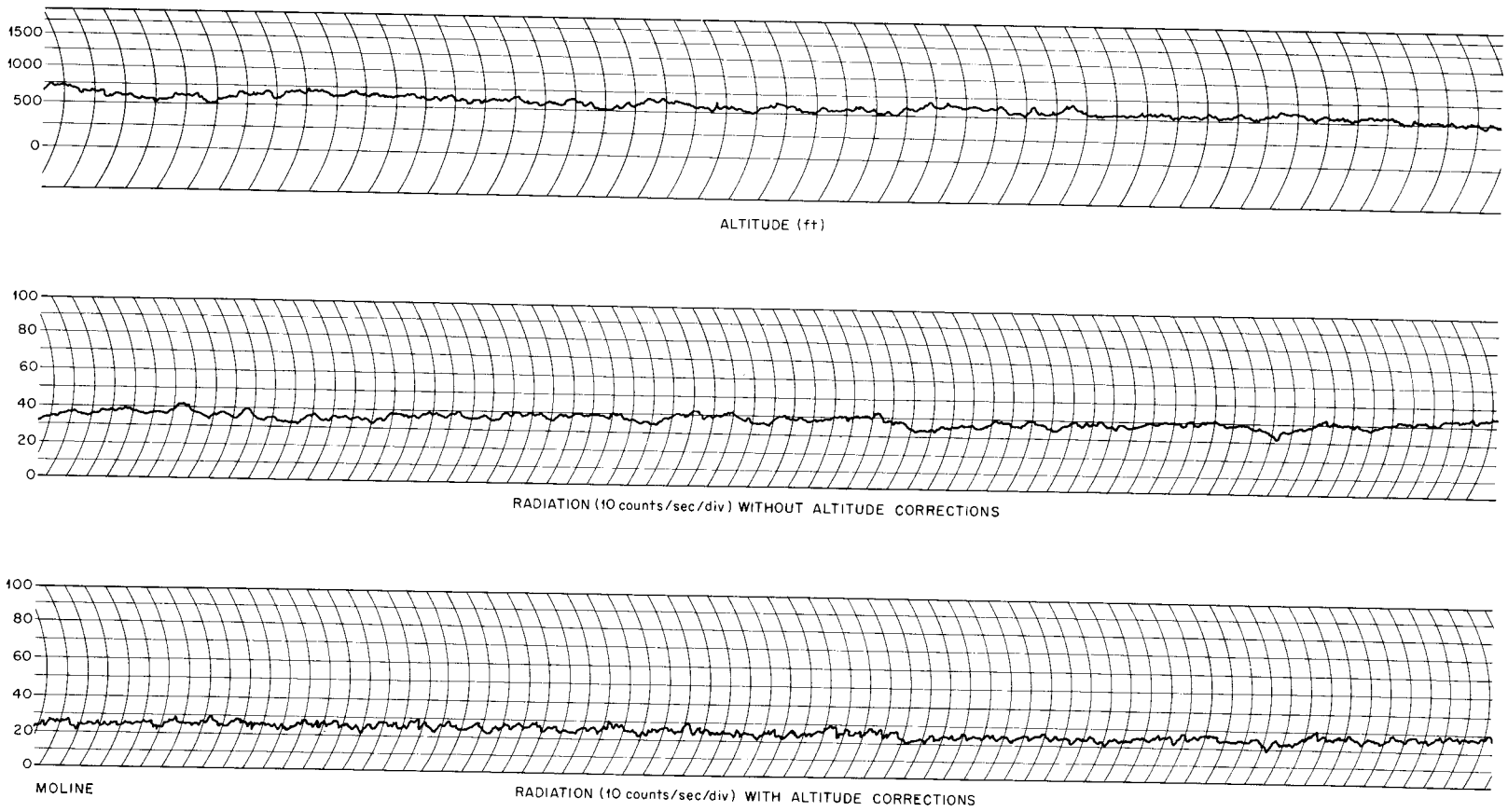

Fig. 7 (continued)

Over land the count is 300 counts/sec as shown in the bottom record. Point *b* in Fig. 7 is the crossing of the Illinois River. The radiation is seen to drop off as Moline, Illinois, is approached. This was probably due to the shielding of snow which was 4 to 6 in. deep. These records were selected because of their uniformity in radiation so that the altitude effect could be shown. This is by no means normal – records obtained on the Texas–New Mexico–Colorado flight showed variations from 10 to 100 divisions, corresponding to 100 to 1000 counts/sec. The line from St. Louis to Moline showed considerably more variation when flown in June 1955, probably because of areas of cultivation and effects of meteorology. It has been noted that after a rain the background radiation is lower.

The Use of Threshold Detectors in Operation Teapot

The technique of using threshold detectors to measure neutron spectra was further refined since the original report.[16] Before their use in Nevada in Operation Teapot, the techniques were applied to the measurements at the ORNL 86-in. cyclotron and at the Los Alamos Lady Godiva. In each case the spectrum was measured and dose was computed by means of the first-collision tissue dose curve; good agreement was found between this method of dosimetry and the direct proportional-counter method.[17]

In Operation Teapot, gold, plutonium with B^{10}, neptunium, and sulfur were used as threshold detectors, and the spectrum was measured in air and in lead hemispheres as a function of distance from the source for the five shots investigated in the Biomedical Program. From the measured spectrum the dose (rep) received by the mice being studied by the Los Alamos group was computed. The most important conclusion is that the neutron spectrum does not change with distance; hence a re-examination of data to obtain better dose estimates at previous test series is possible.

[16] G. S. Hurst *et al.*, *Neutron Flux and Tissue Dose Studies with Fission Threshold Detectors*, ORNL-1671 (March 30, 1954).

[17] G. S. Hurst, *Brit. J. Radiol.* **XXVII**, 353 (1954).

EDUCATION, TRAINING, AND CONSULTATION

E. E. Anderson

AEC FELLOWSHIP PROGRAM

E. E. Anderson M. F. Fair

The 15-week graduate course in Radiological Physics was completed at Vanderbilt University for the 20 AEC Fellows. The ORNL Field Training Program for these Fellows was organized and is now in progress. Ten of the Fellows coming to the Laboratory from Vanderbilt in June have been granted extensions of their fellowships and will work on research problems either at ORNL or at Vanderbilt University which will complete their theses in partial fulfillment of the requirements for a master's degree.

The AEC Fellows for the 1955–1956 program have been selected and will enroll at Vanderbilt University in September.

OTHER ACTIVITIES

E. E. Anderson M. F. Fair
M. R. Ford

A course in mathematics of electricity was conducted for the instrument mechanics in the Laboratory Apprentice Training Program.

A two-week advanced and concentrated course in health physics was given to one Japanese and three Egyptian scientists. Masami Izawa, the Japanese, is a member of the National Institute of Health, Tokyo, Japan, and he will have the responsibility for monitoring air, food, and water when he returns to Japan. The three Egyptians belonged to the group of four Egyptian personnel sent to this country for training in medical and biological uses of radioisotopes and will be the key scientists responsible for the establishment and operation of a radioisotope laboratory in Cairo, the first such laboratory to be established in Egypt.

A six-week advanced course in health physics was conducted for seven medical officers from the Advanced Radiobiology Course of the Armed Forces Special Weapons Project, and one officer from the Radiological Training Department, Ft. McClellan, Alabama.

The section assisted the USNRDL in the establishment of a training program and in the preparation of training material for Operation Wigwam, and also assisted Ohio State University in the planning and conducting of the first unclassified health physics conference ever to be held. The conference was held on the Ohio State University campus at Columbus, Ohio.

The section conducted special lectures and discussion periods for: (1) armed forces visitors, (2) Carbide and Carbon Chemicals Company visitors, (3) MIT Practice School, (4) ORNL Safety Department, and (5) ORNL Orientation Program.

The program of consultation with all outside agencies has continued.

One member of the section participated in calculating maximum permissible single-exposure values for the radioisotopes considered in the ICRP Handbook. Tables of values were set up and were based on five different criteria, as follows:

1. the inhalation of soluble radioactive material,
2. the injection of soluble radioactive material into the body by way of a puncture wound,
3. the injection of insoluble radioactive material by way of a puncture wound,
4. the inhalation of insoluble radioactive material with the lung as the critical organ,
5. the inhalation of radioactive material where some portion of the gastrointestinal tract is the critical body tissue.

For case 5 the dose for the various radioisotopes was calculated at the end of each of the 31 hr that the radioisotopes spent in the gastrointestinal tract. The ORNL digital computer was used for these calculations.

This material will be presented at the Geneva Conference.

Tables of values were compiled from the literature for range and specific ionization of alpha and beta particles in air and tissue. These values will appear in the *Nuclear Engineering Handbook* to be published by McGraw-Hill.

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