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REPORT OF THE BIOASSAY LABORATORY JULY 1972 THROUGH JUNE 1973

DONNER LABORATORY

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David J. Yaeger and Anne de G. Low-Beer

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REPORT OF THE BIOASSAY LABORATORY*
July 1972 through June 1973

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ABSTRACT

Summary results of the bioassay program for fiscal 1972-1973 are presented. These include results from the routine survey program and from special tests following known or suspected radioactive releases. Two incidents involving excretion levels of ^{14}C and ^{3}H exceeding I.C.R.P. derived investigation levels are treated in detail. Accomplishments of the research and development program are described. They are: (1) development of a portable instrument for tritium bioassay, (2) absolute efficiency calibration of the Ge(Li) gamma spectrometer, (3) development of a computer program for rigorous analysis of gamma-ray spectra from scintillation detectors.

*Work done under the auspices of the U. S. Atomic Energy Commission.

INTRODUCTION

Bioassay measurements of internal radioactive contamination are an important part of an effective radiation safety program. At the Lawrence Berkeley Laboratory both whole-body counting and radiochemical analysis of excreta are used for this purpose. This report summarizes the results of the bioassay program for fiscal year 1972-73.

At LBL, 117 employees are requested to supply 24-hour urine samples for bioassay. Requests are generally made either once or twice per year for each employee. These persons work with, or in areas containing, unsealed radioactive materials. During the period of this report, 84 employees submitted urine samples for a 72% compliance factor. In addition to this "routine" program, the bioassay laboratory performs special tests in the case of actual or suspected radioactive release.

ROUTINE PROGRAM RESULTS

Table I presents results of the routine urinalyses. The radiochemical procedures have been detailed by Buckley[1]. Briefly, the gross alpha procedure is sensitive to actinium, thorium, neptunium, plutonium, americium, curium, californium, and einsteinium with a detection limit of 0.2 dpm/sample. The gross beta procedure excludes monovalent cations, and has a detection limit of 2 dpm/sample.

The results shown in Table I for gross alpha, beta, and gamma are typical of our past experience, and do not differ significantly from data on non-laboratory residents of the Bay Area.

Table I. Routine urine sample results,
July 1, 1973 to June 30, 1973.

Determination	No. done	No. pos.	% pos.	Range of pos.
Gross alpha	104	8	7.7	0.21- 1.82 dpm
Gross beta	102	57	56.	2.9 -39. dpm
Gamma emitters	104	0	--	--
Phosphorus-32	15	0	--	--
Carbon-14	19	1	5.	0.05 μCi/liter
Tritium*	13	6	46.	0.1 - 0.8 μCi/liter

*Measured at Lawrence Livermore Laboratory.

SPECIAL TEST RESULTS

In response to actual or suspected radioactive releases, 38 urinalyses were performed. The results are presented in Table II. A total of 7 employees were involved in these tests.

Table II. Special urine sample results,
July 1, 1972 to June 30, 1973.

Determination	No. done	No. pos.	% pos.	Activity Range
Polonium-210	3	0	--	--
Phosphorus-32	1	1	100	0.12 nCi/liter
Carbon-14	14	11	79	0.4-48 nCi/liter
Tritium*	20	20	100	0.6-142 μCi/liter

*Measured at Lawrence Livermore Laboratory

Two incidents resulted in excretion rates exceeding "derived investigation levels" as recommended by the International Commission on Radiological Protection[2]. It should be emphasized that these investigation levels do not imply harm to the involved individual,

but rather that the incident should be studied in detail. No dose estimates exceeded the recommended quarterly exposure limits for radiation workers.

These two incidents are described in the appendix.

Whole-body counting results

In fiscal year 1972-73, 38 personnel were given routine whole-body counts. Of these there were 4 who had minor contamination on their clothing: 2 with ^{51}Cr, one with ^{99m}Tc, and one with ^{181}W. One subject had an internal burden of ^{65}Zn, which he had worked with about 6 months previously. From our data on normal subjects injected with this isotope, his burden at the time of exposure was calculated to have been 76 nCi. Although the amount was far below the MPB, this represents a situation of possible hazard because the personnel present were not aware of any contamination problem.

Ten subjects were counted on a nonroutine basis for suspected recent exposure. The isotopes involved were ^{210}Pb, ^{181}W, ^{109}Cd, ^{60}Co, ^{22}Na, ^{65}Zn, and ^{125}I. Of these, no detectable activity was found in any except for one subject who had 42 nCi of ^{181}W.

Research and development activities

1. Field system for tritium bioassay

We have assembled and calibrated an instrumental tritium bioassay system capable of field operation. This equipment is similar to that developed by the Hazards Control group at Lawrence Livermore Laboratory [3]. A schematic of the apparatus is shown in Fig. 1. The complete system including all accessory equipment has been contained in a 21" X 14" X 7" suitcase with a total package weight of 33 lbs.

This battery-powered instrument uses acetylene (C_2H_2) generated by the chemical reaction of calcium carbide and urine as the counting gas in a proportional counter. If tritiated water is contained in the urine, the tritium is incorporated in the acetylene and produces counts in the proportional chamber. Nominal sensitivity is 150 to 200 cpm/µCi/liter. Background readings are typically less then 1000 cpm (250 to 1000). Two solutions of tritiated water are included for field calibration. With dilutions of 1:9, these standards should produce net count rates of approximately 3000 cpm and 120,000 cpm, equivalent to 15 µCi/l and 600 µCi/l respectively.

Approximately 10 cc of the sample fluid is placed in a 20-cc syringe and allowed to drip through a 21-gauge needle onto lump calcium carbide. The generated gas passes through a CaC_2 drying tube and into the flow counter. The counter high-voltage should remain off for 2 minutes after dripping starts, since counter discharge occurs until air has been swept out of the chamber. For typical drip rates, the 10-cc sample takes about 10 minutes to empty, and equilibrium count-rates occur in 6-8 minutes. Occasionally low drip-rates are encountered, which tend to give low meter readings. This may be remedied by slow injection of the fluid using the syringe plunger. Water-vapor passage into the counter is generally avoided if the connecting tube between generator and drying tube has enough vertical run to allow refluxing of fluid into the generating flask.

In addition to its main function as a portable field-use instrument, the system has been found useful in the laboratory in providing crude but rapid (10-15 minutes) bioassays for tritium.

2. Ge(Li) photopeak efficiency calibration

Photopeak counting efficiency of our Ge(Li) gamma spectrometer has been determined for activities contained in a 10-ml volumetric flask located 5 cm from the detector. The 10-ml volumetric flask is our standard source geometry for bioassay samples; the 5-cm separation of source and detector minimizes the "summing" effects if cascade gammas are emitted from the source.

Relative detection efficiencies were determined by measuring isotopes emitting several gamma rays and taking the intensity measurements of Gunnink[4] to find relative efficiencies. The energy range 140 to 1330 keV was calibrated in a "leap frog" manner as follows. First a ^{243}Am-^{239}Np source was counted and the reported intensities used to calculate relative efficiencies at 210, 228, 278, and 334 keV. Efficiency at 284 keV was interpolated from these results. Then a ^{131}I source was counted (E_γ = 284, 364, 637, 723 keV) and the 284-keV efficiency used to calculate efficiencies for the other 3 lines. Subsequent counting of the isotopes ^{22}Na, ^{59}Fe, and ^{60}Co extended the energy range from 140 to 1330 keV.

Absolute calibration in terms of counts per emitted gamma ray was accomplished by comparison with point sources of ^{137}Cs, ^{22}Na, and ^{60}Co that had been calibrated by the International Atomic Energy Agency. Using known volumes of solutions of these 3 isotopes, we prepared point sources similar to those from IAEA. The simple ratio of observed counts from the point sources thus provided the specific activity of our isotope solutions. Transferring the solutions to 10-ml flasks then allowed an absolute-efficiency determination at 5 energy points for adjustment of the relative-efficiency curve. The final results are shown in Fig. 2.

3. Computer program for analysis of gamma-ray spectra

A FORTRAN computer program, WHOBO, has been written for analysis of gamma spectra obtained with our NaI(Tl) scintillation detector. Given a library of standard spectra for individual isotopes, the program makes a fit to an unknown spectrum with some linear combinations of the standards. The fitting criterion is minimizing the weighted squared deviation between the fitted curve and the data. The primary output data are the fractions of standards used in the fit, together with the standard deviation of these fractions.

Graphical output includes a plot of the data points, fitted curve, and individual components of the fit as shown in Fig.3 . The residuals between data and fit are also plotted. This is useful in determining whether any isotopes were present in the unknown composit that were not included in the standards library.

A preprocessor program called WHOIN has also been written. The main function of WHOIN is to convert pulse-height analyzer output on punched paper tape into a form suitable for input to WHOBO. One conversion is into a FORTRAN-compatible input file for immediate use, in which case WHOIN and WHOBO would be run in tandem. A more usual conversion is to transfer the paper tape data to cards, and later submit the card decks as input for WHOBO.

The WHOIN program also has options for producing Cal-Comp plots of the input data and calculating count rates and total counts in specified channel ranges.

Professional activities

1. David Yaeger attended the 18th Annual Conference on Bioassay, Environmental, and Analytical Chemistry, October 10-11, 1972. He presented "Radioactivity Levels in Wild Deer" by Anne de G. Low-Beer, Jensen Young, David J. Yaeger, Roscoe C. Drisdale, and Barbara C. Buckley.

2. Anne Low-Beer contributed a chapter on Bioassay of Plutonium to the Handbuch für Experimentellen Pharmakologie Vol. XXXVI (Springer-Verlag, Heidelberg, West Germany) and also completed the Subject Index of the entire volume.

3. Charles Schmidt attended the 18th Health Physics Society Meeting, June 17-21, 1973.

APPENDIX--Details of radiation incidents exceeding ICRP investigation levels

Two incidents exceeding ICRP investigation levels occurred during the period of this report. Each incident involved two individuals.

Incident 1

On February 20, 1973, two individuals were exposed to tritium following an accidental release during a tritium labeling procedure. Spot urine samples were obtained promptly and measured for tritium specific activity using the field tritium-bioassay instrument described earlier in this report. More accurate assays were determined by liquid scintillation counting at Lawrence Livermore Laboratory. The results are presented in Fig. 4.

Dose estimates were made as follows. Assuming that urine specific activity represents body-water specific activity, the initial body content, R_o, is given by

$$R_o = S_o \cdot V \qquad \mu Ci$$

where

S_o = specific activity of urine at t=0, μCi/liter

V = total body-water volume, liters.

Body content at time t is then:

$$R(t) = R_o \, e^{-\lambda t} \qquad \mu Ci$$

where

λ = clearance rate from excretion curve, day^{-1}

$$= \frac{.693}{T_{1/2}}$$

Dose is calculated from:

$$D = 51.2 * \int_0^{18000} R(t)\, dt * \frac{\varepsilon}{m} \qquad rem$$

$$= 51.2 * \frac{R_o}{\lambda} * \frac{\varepsilon}{m}$$

$$= 51.2 * \frac{S_o \cdot V \cdot T_{1/2}}{.693} * \frac{\varepsilon}{m}$$

where ε = effective energy absorbed per disintegration

= 0.01 MeV

m = body-water mass, grams

Since m = 1000 * V, we arrive at the relation

$$D = 7.4 \cdot S_o \cdot T_{1/2} \times 10^{-4} \quad \text{rem}$$

which does not involve the mass of body water. This is an important conclusion since the weights of these two subjects differed by a factor of more than 2.

The dose estimates were then 0.88 rem for subject A and 0.10 rem for subject B.

Incident 2

A routine urine sample received June 6, 1973, showed unexpected ^{14}C activity of 49 nCi/liter. Co-workers of this individual were checked and one was found to have similar urine levels. Investigation by Safety Services found a work bench top with low levels of ^{14}C activity. The bench had evidently been contaminated years previously and painted over for containment, but the cover layer was now being worn or chipped through. The area was cleaned, recovered, and was swipe-free on July 23, 1973.

Dose assessment from the excretion data of Fig. 5 is difficult since neither the chemical form of activity nor the time course of intake are known. Subject C, who submitted the first active sample, had a negative bioassay result on September 6, 1972. His co-worker, subject D, showed a negative result on April 12, 1973. We will assume that neither subject received any radioactivity prior to April 12, 1973.

From the data of Fig. 5, it appears that a reasonable excretion equation would be a single exponential with a clearance half-time of 18 days. The more rapid clearance following containment of activity suggests that some activity uptake may have occurred in the period. 0 to 40 days. To calculate a "maximum possible" dose we will, however, assume that uptake occurred as a single event immediately following subject D's last negative sample and clearance was exponential with a half-time of 18 days.

If urinary excretion $Y_u(t)$ is given by:

$$Y_u(t) = Y_o\, e^{-\lambda t} \qquad \mu Ci/day$$

then it follows that whole-body retention, R(t), is

$$R(t) = \frac{Y_o}{F_u \lambda}\, e^{-\lambda t} \qquad \mu Ci$$

where

F_u = fraction of isotope excretion via urine

= .036

Then dose is calculated from

$$D = 51.2 * \int_0^{18000} R(t)\, dt * \frac{\varepsilon}{m} \qquad \text{rems}$$

$$= 51.2\, \frac{Y_o}{F_u \lambda^2} * \frac{\varepsilon}{m}$$

Considering total body as the critical organ, ε = .054 MeV and m = 70,000 g.

Maximum possible doses are then calculated to be 0.45 rem for individual C and 0.67 rem for individual D.

REFERENCES

1. Barbara C. Buckley, Manual of Procedures, Lawrence Berkeley Laboratory Report UCRL-19355, 1969.

2. International Committee on Radiological Protection, Publication 10, (Pergamon Press, Oxford, 1966).

3. S. Block, D. Hodgekins, and O. Barlow, Recent Techniques in Tritium Monitoring by Proportional Counters, Lawrence Livermore Laboratory Report UCRL-51131, 1971.

4. R. Gunnink (Lawrence Livermore Laboratory) personal communication, 1973.

FIGURE CAPTIONS

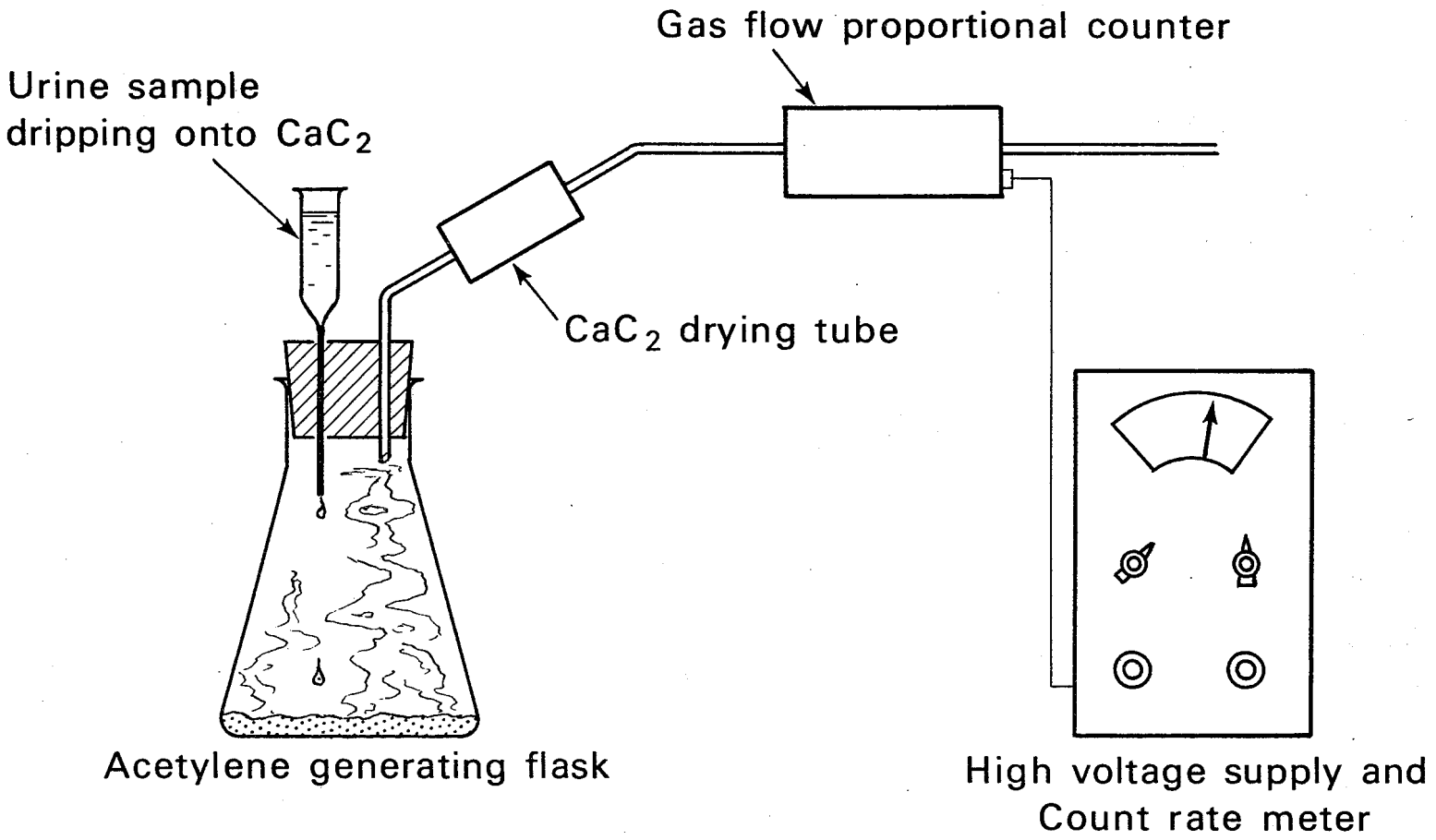
Fig. 1: Schematic of field system for tritium bioassay.

Fig. 2: Absolute photopeak counting efficiency of Ge(Li) detector.

Fig. 3: Analysis of a composit spectrum containing ^{137}Cs, ^{22}Na, and ^{60}Co.

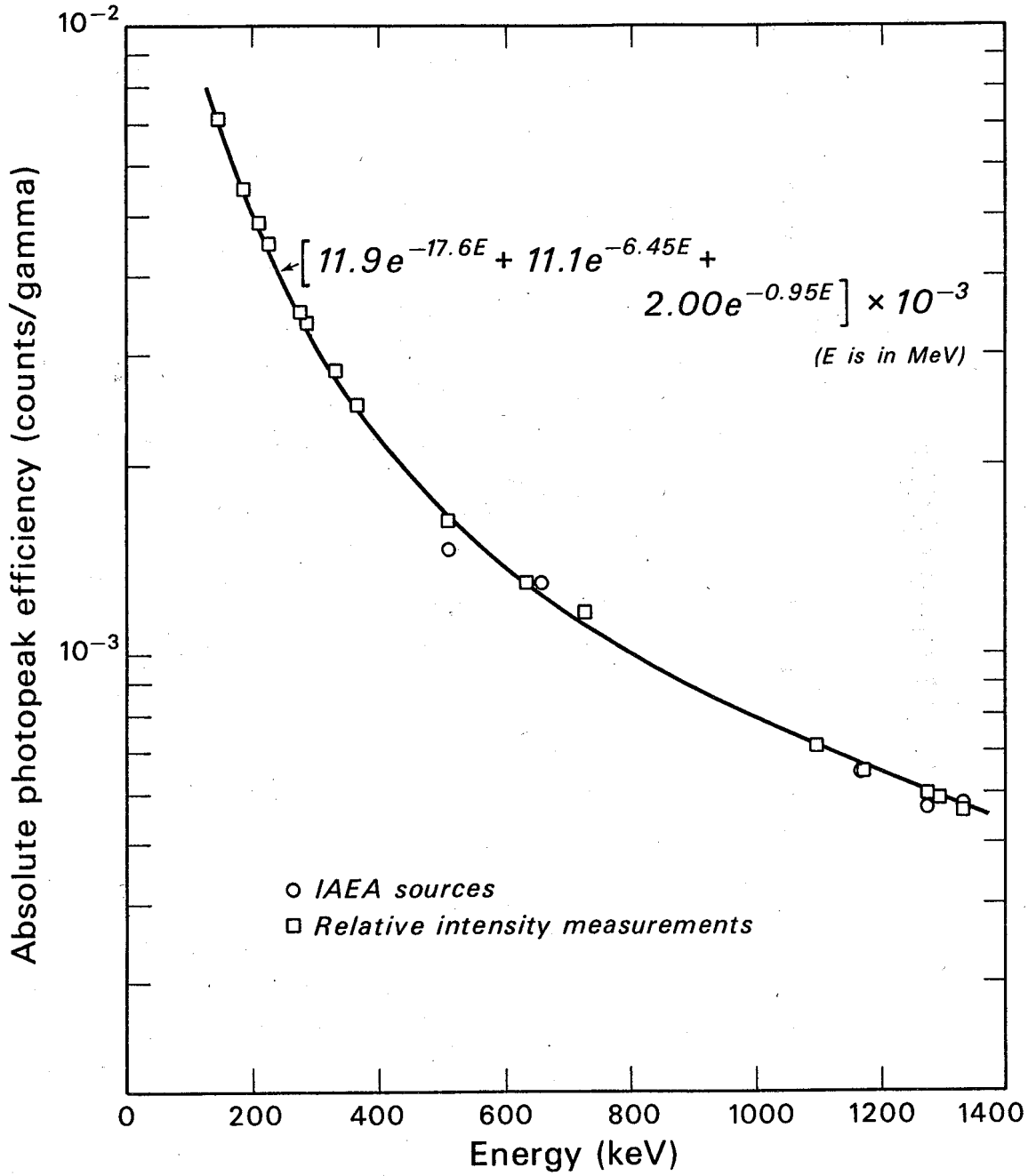
Fig. 4: Tritium excretion in two individuals accidentally exposed February 20, 1973.

Fig. 5: ^{14}C excretion in two individuals.



DBL 7311-5351

Fig. 1.



DBL 7311-5352

Fig. 2.

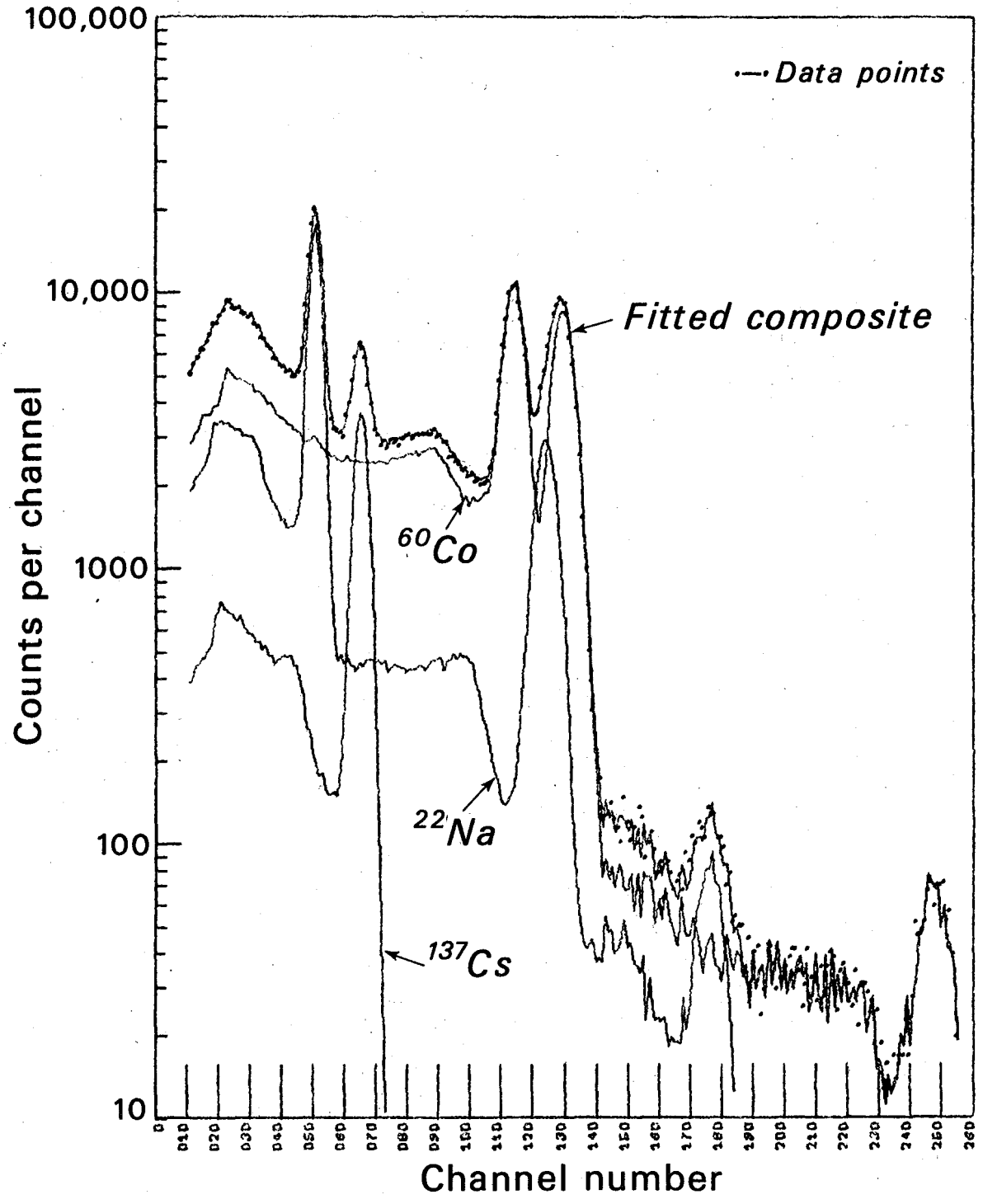


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Fig. 3.

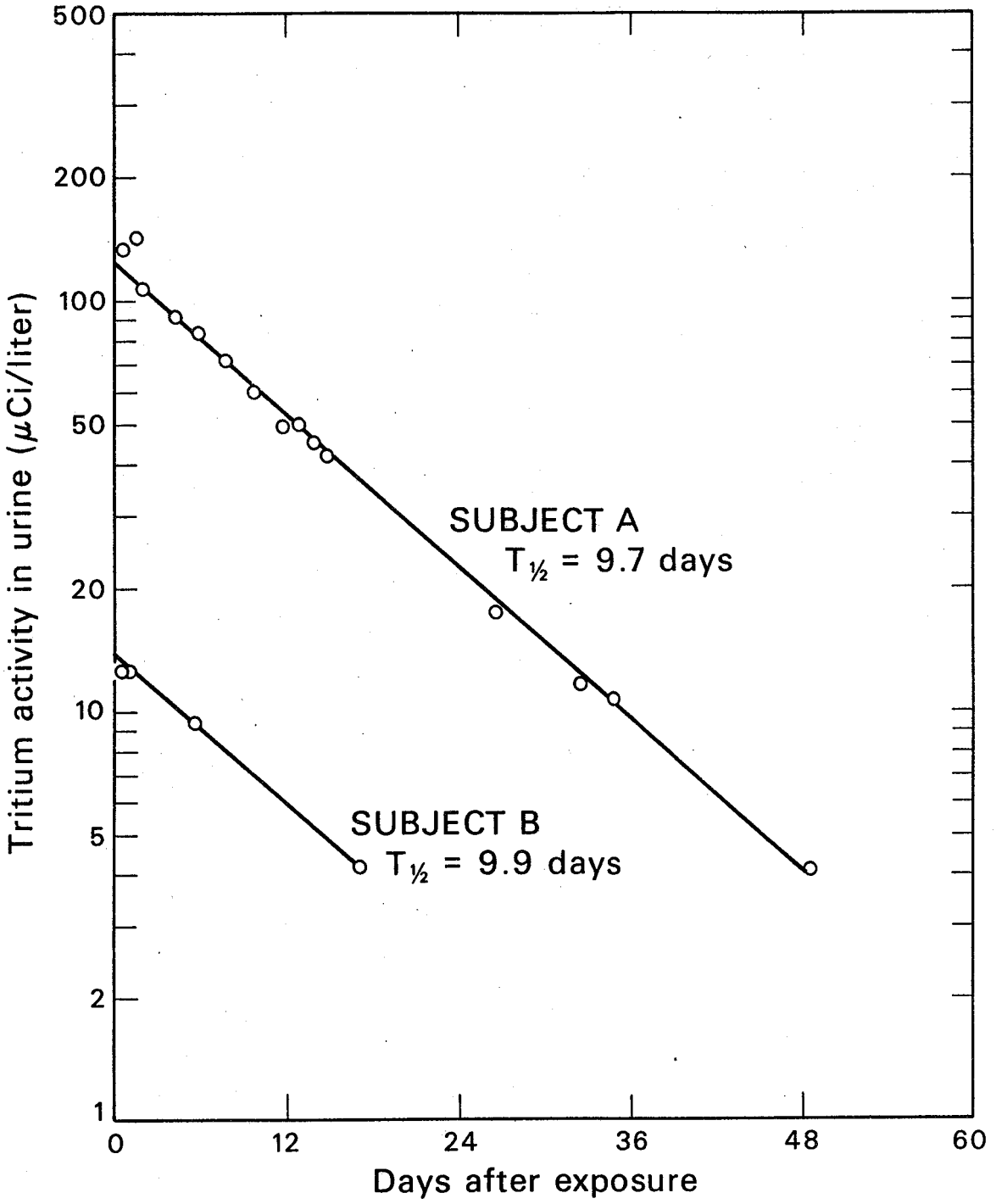

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Fig. 4.

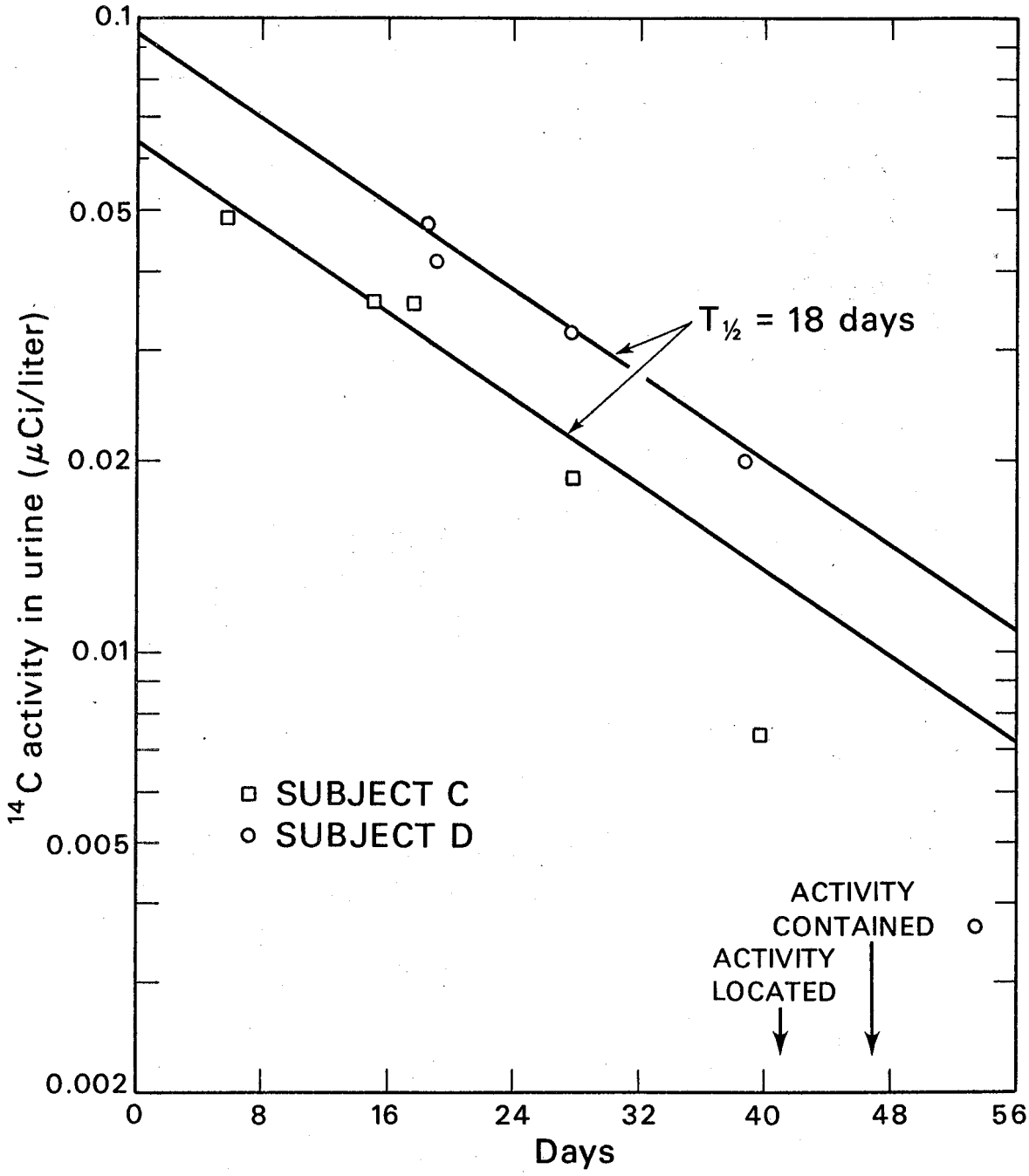

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Fig. 5.