RECEIVED
LAWRENCE
RADIATION LABORATORY
JAN 9 1969
LIBRARY AND
DOCUMENTS SECTION

UCRL-18588

cy 2

REPORT OF THE BIOASSAY PROGRAM
July 1967 through June 1968

Anne de G. Low-Beer and Thornton W. Sargent

November 1968

TWO-WEEK LOAN COPY

This is a Library Circulating Copy which may be borrowed for two weeks. For a personal retention copy, call Tech. Info. Division, Ext. 5545

LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

UCRL-18588

cy 2

UCRL-18588
UC-48 Biology and Medicine
TID-4500 (52nd Ed.)

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

REPORT OF THE BIOASSAY PROGRAM

July 1967 through June 1968

Anne de G. Low-Beer and Thornton W. Sargent

November 1968

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.65

REPORT OF THE BIOASSAY PROGRAM*

July 1967 through June 1968

Anne de G. Low-Beer and Thornton W. Sargent

Lawrence Radiation Laboratory
University of California
Berkeley, California

November 1968

Abstract

The report of the Bioassay Program presents results of radiochemical analysis of urine and whole-body counts on selected groups of employees of LRL. It describes on-going research and development, particularly with respect to the use of a solid-state semiconductor for wound counting. It also discusses methods of analysis and an automated system for processing and retrieval of data.

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

I. Introduction

The purpose of the Bioassay Program at LRL—Berkeley is to collect and maintain complete information concerning acquisition of body burdens of radioactive nuclides by employees of the Laboratory. This purpose is implemented by (a) a routine program of radiochemical urinalysis and whole-body counting; (b) emergency determinations for all persons involved in specific episodes through which internal contamination by radionuclides might be acquired; (c) special projects; (d) research and development.

During the past fiscal year the bioassay program has emphasized, on the one hand, increased collaboration with other departments and divisions of the Laboratory, and, on the other hand, improvement and enlargement of its own capacity to detect and identify radioactive contaminants. Increased collaboration is reflected by continuing close cooperation with the Department of Safety Services, by increased coordination of radiochemical analysis with whole-body counting, and by a new program involving cooperation with the Department of Health Physics. Efforts to strengthen internal operation have included (a) addition of one member to the bioassay staff who has responsibility for alpha and beta counting, and development of gamma spectroscopy of urine samples, and for operation of a recently acquired TMC lithium-drifted silicon solid-state semiconductor for in vivo or in vitro detection of alpha-emitting nuclides by means of their x-ray spectra, and (b) development of an automated system for processing data generated in the program of radiochemical determinations. The Bioassay Laboratory also maintains a continuing program of development and research directed toward promulgation of new and improved radiochemical procedures and more effective use of electronic equipment.

Various features of the bioassay program are more completely covered in the following sections of this report.

II. Routine Program

1. Radiochemical analysis

The list of employees designated to have annual radiochemical assays of urine samples is furnished to the Bioassay Laboratory by the Safety Services Department. The list, in addition to giving the names of the individuals, indicates the radionuclides to which these persons may have been exposed. This is a new feature of the cooperative arrangement between the Laboratory and the Safety Services Department, which enables the Laboratory to make appropriate determinations on each sample. During the past year there were 177 individuals on the routine bioassay list. Of these 117, or 66%, were listed as potentially exposed to any or all radionuclides that are encountered at LRL—Berkeley. Thus, each such sample required as least three different determinations--for alpha, beta, and gamma activity. In addition, 39 persons, or 22%, were designated by Safety Services for whole-body counts in addition to radiochemical assay of urine. Detailed procedures used in the radiochemical analysis of urine specimens are contained in the Bioassay Laboratory Manual of Procedures, revised, Summer 1968. The survey method used for screening samples for gamma-emitting nuclides and for gross beta and gross alpha activities is attached to this report as Appendix 1.

A computer program has been developed within the Bioassay Laboratory whereby results of all alpha and beta counts are processed by Data Processing Service. There has thus been added a capability for storing and retrieving data, for sorting and correlating data, and for providing summaries of data as required for analysis of statistics involved in the bioassay program. A computer program for analysis of gamma and x-ray spectra is now in the process of development.

2. Whole-Body Counting

The Donner Whole-Body Counter is of the Argonne type, with a 6-inch steel shield. The subject sits in an inclined chair and is counted by a stationary 9.75×4-inch NaI(Tl) crystal, positioned with its center 40 cm from the back and seat of the chair. The gamma pulses are analyzed by a Packard 400-channel pulse-height analyzer, calibrated at 5 keV/PHU, yielding a gamma-ray spectrum from 0 to 2.0 MeV. Data are recorded on printed paper tape, which is then keypunched for computer analysis. Calibration of the area under the photopeaks to yield microcuries of body burden is obtained from the spectra of medical patients injected with known quantities of short-lived radionuclides with various gamma-ray energies.

Results of the routine program are shown in Table I.

3. Discussion

Persons included in the routine bioassay program have a low incidence of contamination by alpha-emitting nuclides, and in no instance had the level of contamination exceeded 0.01 MPBB. Positive determinations are regarded as questionable until they have been confirmed by analysis of a second sample. This procedure also makes possible the identification of the source of contamination by alpha pulse-height analysis. The sensitivity of the gross alpha method--0.1 pCi per 24-hour urine sample--is such that levels of contamination which have no clinical significance can be demonstrated. We believe, however, that the collection of such information on a long-term basis may point out situations in which either the working environment or individual carelessness involves potential hazard.

The specific actinide determinations (Table I) were performed on four individuals who were potentially exposed to uranium, three of whom has positive urines, and on two persons whose specimens had been positive in the gross alpha determination. One of these showed contamination with ^{244}Cm and the other with ^{241}Am. The remaining two persons whose gross alpha determinations were positive had not yet submitted an additional sample at the end of the reporting period. If the positive values found in the specific actinide determinations are combined with the positive gross alpha determinations, the total number of persons showing contamination by actinide elements is 7 in a total of 84 persons whose urine was assayed. The overall percentage of positive actinide cases is therefore 8%.

During the summer of 1967, all urine samples analyzed in the Bioassay Laboratory showed gross beta contamination. In August 1967 a group of 12 persons residing in various locations in the San Francisco Bay area, and not engaged in any activity involving contact with radioactive nuclides, were asked to submit "control" urine samples. All the samples were positive for gross

Table I. Routine bioassay program.

Type of determination	Number of determinations	Determinations found positive (number)	(%)	Range of values found
I. Radiochemical				
A. Gross alpha	80	4	5.0	0.10 – 3.9 pCi/24 hr
B. Specific actinide	6	5[a]	83.5	0.15 – 1.0 pCi/24 hr
C. Gross beta	84	70	84.5	1 – 20 pCi/24 hr
D. Phosphorus-32	5	0	--	--
E. Carbon-14	3	0	--	--
F. Tritium[b]	4	0	--	--
G. Rare earth	2	0	--	--
H. Gamma spectroscopy	75	0	--	--
II. Whole-body count	39	3[c]	7.7	(i) 20 nCi (ii) 10 nCi (iii) 9 nCi

a. Three uranium, one ^{244}Cm, one ^{241}Am.

b. Processed at LRL–Livermore.

c. (i) ^{134}Cs + a mixture of unidentified material of energy 0.39 MeV;

(ii) ^{134}Cs;

(iii) $^{99}Tc^{m}$.

Detection limits: alpha, 0.1 pCi/24-hr urine; beta, 1.0 pCi/24-hr urine; gamma, 10 nCi/24-hr urine; WbC, 1 to 10 nCi.

beta activity in the range of 5 to 20 pCi per 24 hours. It has been concluded that the presence of gross beta activity in urine during this time was common to all residents of the Bay Area, and that it was due to environmental contamination. Table II shows the trend in percentage of gross beta determinations that are positive in LRL employees, and in the levels of activity found in the individuals we have observed. It is not possible at this time to identify the isotopes that have given rise to gross beta activity in the urine, since the low levels did not appear to us to justify a request for a second specimen. However, the rate of radioactive decay has been studied on some of the samples. It appears that most of the activity decays with a relatively short half-life, i.e., in the neighborhood of 55 to 60 days. It is unlikely, therefore, that any significant part of this activity is due to ^{90}Sr.

Table I illustrates the greater sensitivity of radiochemical analysis than of whole-body counting for determination of actinide and beta-emitting elements. On the other hand, the whole-body counter is a more sensitive instrument than our present method of gamma spectroscopy of urine. The data point out the complementary nature of these two methods of analysis, and suggest the desirability of including both methods in a comprehensive bioassay program.

III. Emergency and special cases

Eleven persons were involved during the year in incidents in which exposure to actinide elements had occurred. Table III presents the results of investigation of these exposures.

Four of the persons involved in these incidents are on the routine bioassay list. Seven had never before submitted samples to the Laboratory. Of the eleven persons, eight had one or more positive samples following presumed exposure. In no case did the activity exceed 0.01 MPBB. It is significant that three persons whose urine was negative later submitted fecal samples all of which were positive. A fourth person submitted only a fecal sample, which was positive. Whole-body counts made on these individuals were all negative for alpha activity. It seems evident, therefore, that in cases of exposure to very small amounts of alpha activity, or to alpha-emitting nuclides that do not have detectable gamma activity, analysis of a fecal sample is a necessary procedure for effectively ruling out the presence of alpha contamination.

One incident involved ^{90}Sr acquired through the bite of an animal. A spot urine collected immediately after the incident was negative, as was a 24-hour sample submitted 2 days later. A third determination made on a 24-hour sample collected 64 days after the incident contained 3.38 pCi of ^{90}Sr.

Table II. Average values found in gross beta determinations. (Totals include all cases from Tables I and IV--shown in parentheses)

Month		Average value (pCi/24 hr)		Positive cases (%)	Number of cases
		Positive cases	All cases		
1967	July	16.8	16.8	100	(15)
	Aug.	8.3	8.3	100	(9)
	Sept.	5.5	5.5	100	(11)
	Oct.	6.0	6.0	100	(18)
	Nov.	10.5	10.5	100	(21)
	Dec.	7.3	7.3	100	(6)
1968	Jan.	8.0	7.4	93	(15)
	Feb.	5.6	4.6	82	(22)
	March	5.7	4.1	77	(26)
	April	6.3	2.9	46	(13)
	May	2.8	1.5	55.5	(11)
	June	3.3	3.3	100	(4)

Table III. Individuals involved in alpha contamination incidents.

Individual	Urine: Number of determinations Total	Urine: Number of determinations Positive	Urine: Range of values (pCi/24 hr)	Feces: Number of determinations Total	Feces: Number of determinations Positive	Feces: Range of values (pCi/24 hr)	Isotopes identified by PHA	Whole-body count
1	2	0	--	2	2	0.31	^{235}U ^{239}Pu ^{241}Am	negative
2	0	--	--	1	1	1.00	--	count not made
3	1	0	--	2	2[a]	0.4—1.2	--	negative
4	1	1	0.5	0	--	--	--	negative
5	2	0	--	0	--	--	--	negative
6	3	2	0.25	0	--	--	^{244}Cm ^{252}Cf	count not made
7	4	3	0.39-1.0	0	--	--	^{244}Cm ^{252}Cf	negative
8	5	3	0.4—0.5	0	--	--	^{244}Cm ^{252}Cf	negative
9	2	0	--	0	--	--	--	negative
10[b]	2	0	--	1	1	4.5	--	possible trace ^{57}Co ≈ 0.01 μCi ^{187}W
11	2	0	--	0	--	--	--	negative

[a] identification of specific actinide pending

[b] sputum sample assayed--negative

IV. Special Projects

1. A survey of accelerator workers at LRL-Berkeley was initiated by the Department of Health Physics with the cooperation of the Whole-Body Counting and the Bioassay laboratories. Eighty-five individuals employed at the four accelerator sites* were whole-body-counted, and concurrently submitted urine samples for radiochemical analysis. Six additional individuals were whole-body-counted but failed to submit urine samples.

Results of this survey are presented in Table IV.

The pertinent facts and the conclusions resulting from this survey are contained in a report to be issued by the Health Physics Department.

2. During the year the Bioassay Laboratory provided consulting service to the California State Department of Public Health on several occasions. A number of urine samples were processed to provide a check on methods used by laboratories with which the Department has contracts, and several proposed procedures for determination of alpha emitters in urine were reviewed critically.

3. In October 1967 the Bioassay Laboratory was host to the 13th Annual Bioassay and Analytical Chemistry Meeting. Proceedings of this meeting have been issued as UCRL-18140, CONF.-671048.

V. Research and Development

The research effort during the past year has been directed primarily to development of the capability of the TMC lithium-drifted silicon solid-state semiconductor, for counting alpha activity deposited in tissues. The x-ray spectra of a number of alpha-emitting transuranium nuclides have been established, and others are being added to the list as they become available. See Fig. 1. Studies include determination of the sensitivity of this method of detection, quantitation of the average depth and of the amount of the activity in relation to the thickness of tissue in which it is deposited, and identification of isotopes by means of the characteristic energies of their x-ray spectra. Attention is being given to the possibility of distinguishing between the different spectra of isotopes which may be present in combination and in varying relative amounts. With the completion of these preliminary studies, it is now proposed that studies be undertaken with small animals to establish rates of clearnace of various isotopes, in order to provide a broader foundation for the use of this detector as a wound counter.

The Bioassay Laboratory has developed and perfected a method for the isolation and detection of radiophosphorus in biological materials.[1] It is believed that this method has greater sensitivity and reliability for this isotope than several other procedures in general use.

---

*184-Inch cyclotron, 88-inch cyclotron, Bevatron, Hilac.

[1] Manual of Procedures, Bioassay Laboratory, Lawrence Radiation Laboratory, Berkeley.

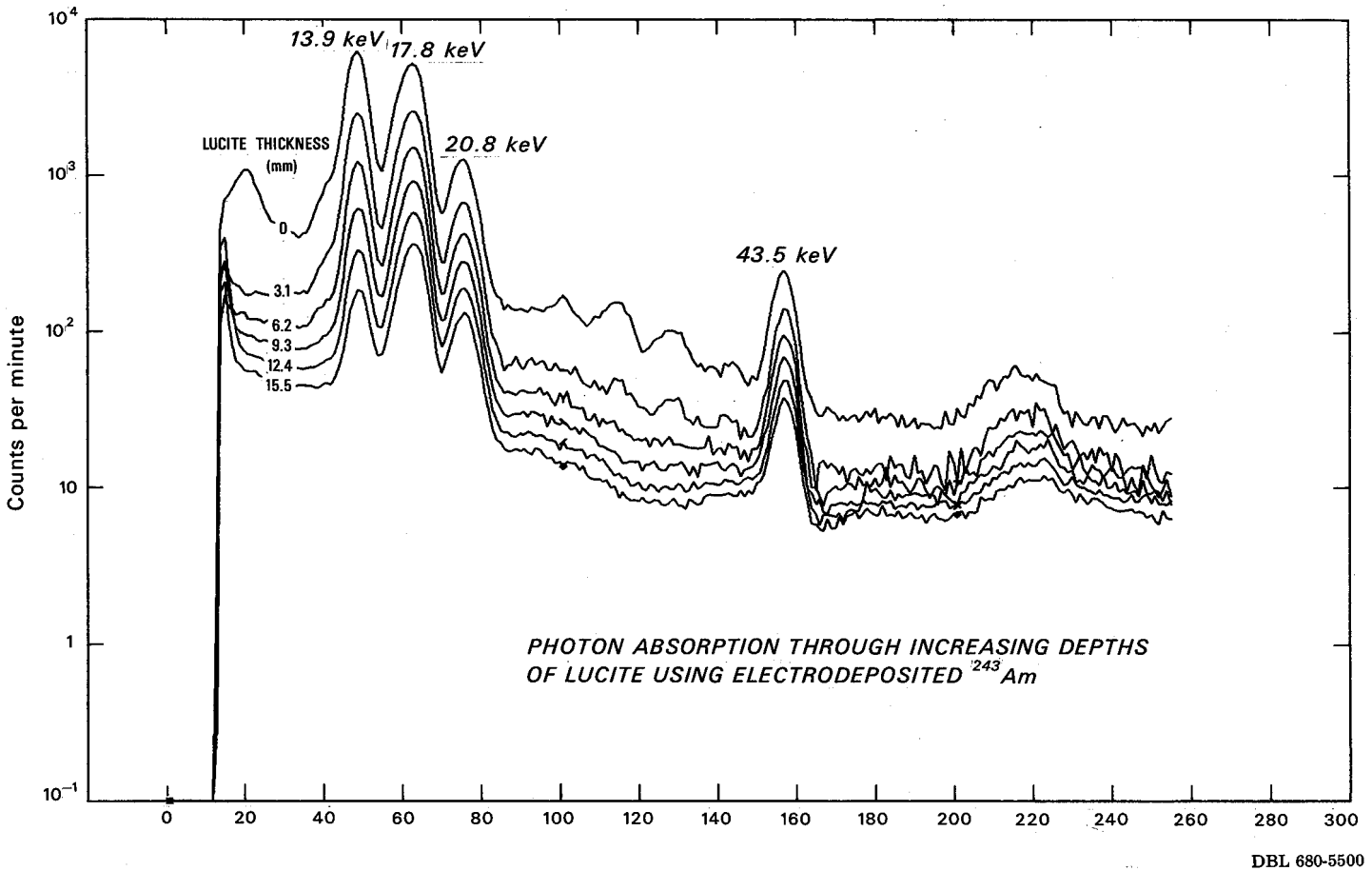

Fig. 1. Spectrum of ^{243}Am generated by computer from counts made on TMC lithium-drifted silicon semiconductor. The source was counted through varying thicknesses of Lucite. Curves illustrate attenuation, also alteration of ratios of the peaks with increasing thickness of intervening material.

Table IV. Survey of accelerator workers.

Type of determination	Number of determinations	Determinations found positive (number)	(%)	Range of values found
I. Radiochemical				
A. Gross alpha	85	6	7.0	0.15-0.5 pCi/24 hr
B. Gross beta	85	76	89.5	1 - 42 pCi/24 hr
C. Gamma spectroscopy	85	0	--	--
II. Whole-body count	91	7[a]	7.7	1 - 33 nCi

a. Contaminants identified by whole-body count were: ^{65}Zn, three persons; $^{56,57}Co$, two persons; ^{185}W, two persons; ^{241}Am, one person.

Two persons had more than one radionuclide.

VI. Conclusion

During the year the Bioassay Laboratory moved to new and more adequate quarters. The marked improvement in facilities, together with closer incorporation into the Medical Services Program, offers opportunity for expansion of activities and for development of a more comprehensive and effective program.

Appendix

Screening Method for Radiochemical Analysis of Urine

Twenty-four-hour samples (two overnight collections) are prepared for analysis by the alkaline phosphate precipitation method. All monovalent cations are removed in the supernatant. The residue is completely ashed with nitric acid and heat, and the pure white ash is suspended in 2N HNO_3 and brought to volume in a 10-ml volumetric flask. Flasks are placed on a 4×4-inch NaI crystal attached to a 400-channel pulse-height analyzer (RIDL Mod. 3412B) and counted for 90 minutes. Data are recorded on paper tape, which is keypunched for computer analysis. The energies of photopeaks are established by comparison with standards of known activity, and the results are quantitated in relation to this activity.

The flasks are then returned to the laboratory and a 2-ml (20%) aliquot is plated on aluminum planchets, which are dried and counted in a Nuclear Chicago proportional gas-flow counter for 100 minutes. The remaining 80% of the original sample is processed for gross alpha activity by bismuth phosphate and lanthanum fluoride coprecipitations. The final precipitate is slurried in water and plated on platinum planchets, and counted for 200 minutes on the proportional gas-flow counter. All actinide elements except radium and uranium are detectable by this procedure.

For details of this method and for methods for determination of specific nuclides, see Bioassay Laboratory Manual of Procedures, revised Summer 1968.