LBL-1799
UC-41 Health Safety
TID-4500 (59th Ed.)

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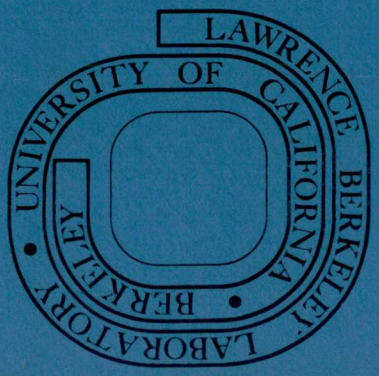
May 1973

Prepared for the U. S. Atomic Energy Commission
under Contract W-7405-ENG-48



Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $4.00; Microfiche $0.95

REPORT OF THE BIOASSAY LABORATORY*

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ABSTRACT

This report covers activities of the Bioassay Program, including whole-body counting of laboratory employees, from July 1, 1969, to June 30, 1972. Results of routine monitoring and investigation of cases of suspected exposure to radionuclides are presented. The scope and accomplishments of the research and development program are described. A number of exposure cases have been referred for investigation by outside agencies. Results of these investigations are presented.

Introduction

The last previous report of the LBL Bioassay Program was issued in 1969.[1] The present report covers the period from July 1, 1969, to June 30, 1972. Neither the routine work load nor the number of cases in which exposure was suspected has justified the preparation of more frequent reports. This fact attests to the excellent record at the Lawrence Berkeley Laboratory with respect to radiation protection. The analytical work of the Bioassay Laboratory and the Whole-Body Counting Program is summarized below. Development and improvement of analytical procedures, and a study of environmental contamination, have accounted for a large part of the laboratory effort during the reporting period. These activities are described in later sections of this report.

Routine Monitoring

Routine monitoring typically includes radiochemical analysis for gross alpha and gross beta activity, and gamma spectrometry of

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

concentrated samples from which ^{40}K and ^{137}Cs have been removed by alkaline phosphate precipitation.[2] Positive alpha determinations are followed by analysis of a second sample for identification of actinide elements. Samples from individuals at high risk of exposure to beta-emitting nuclides such as ^{90}Sr, ^{32}P, and ^{3}H are analyzed for the nuclide of interest. A number of accelerator workers and others who are potentially exposed to fission products and other gamma-emitting nuclides are referred for whole-body counts. Results of the routine program are summarized in Table I. Except for one blood sample analyzed for gross beta activity, all radiochemical determinations were made on urine. Gross alpha determinations show a higher incidence of contamination in the years 1969-70 and 1971-72 than in any other year for which we have issued a report.[1,3] We believe this increase to be more apparent than real. It is the policy of the laboratory to confirm any positive alpha determination by analysis of a second sample. Not infrequently such a check fails to confirm the original determination, indicating that the observed activity was due to contamination of clothing or collection bottles. In the period covered by this report it was impossible in a number of instances to procure a second sample, and the figures shown are therefore not confirmed. It will be noted that the upper limit of the range of gross alpha values is lower with each successive year in the reporting period. As has been pointed out in previous reports, all values that are measurably above background are reported as positive. "Positive" values found in our routine program cannot be regarded as warning levels requiring either further investigation or corrective action. A laboratory study of nonindustrially exposed individuals (see below) suggests rather that alpha activity found in routine sampling of laboratory personnel may reflect background radiation to which all residents of this area are potentially exposed. Further studies of this type may eventually establish investigation levels for individual cases in which the observed activity exceeds background significantly.

The incidence of gross beta contamination is approximately the same as that reported in 1969. With the exception of the high value of 730 pCi per 24-hour urine in 1969-70, upper limits of activity are lower than

TABLE I

Routine Radiochemical Analysis and Whole-Body Counting July 1, 1969 to June 30, 1972

Type of determinations / Period covered	Material analyzed	No. determinations 1969-70	No. determinations 1970-71	No. determinations 1971-72	No. positive 1969-70	No. positive 1970-71	No. positive 1971-72	% positive 1969-70	% positive 1970-71	% positive 1971-72	Range of values (pCi per 24 hrs. unless otherwise specified) 1969-70	1970-71	1971-72
I. Gross alpha	Urine	110	126	86	20	10	17	18.1	7.9	19.7	0.1-2.42	0.1-1.00	0.1-0.3
	Feces	0	0	0	—	—	—	—	—	—	—	—	—
	Other	0	0	0	—	—	—	—	—	—	—	—	—
II. Specific alpha													
A. Uranium	Urine	1	0	1	0	—	0	—	—	—	—	—	—
B. Plutonium	Urine	1	0	1	0	—	0	—	—	—	—	—	—
C. Trivalent actinides	Urine	1	0	1	1	—	1	100	—	100	0.24	—	0.15
D. Radium	—	0	0	0	—	—	—	—	—	—	—	—	—
III. Gross beta	Urine	102	120	79	81	108	59	79	90.0	74.7	1.0-730.0	1.0-64.0	1.0-21.2
	Feces	0	0	0	—	—	—	—	—	—	—	—	—
	Other	Blood 1	0	0	1	—	—	100	—	—	650/liter	—	—
IV. Specific beta													
A. ^{90}Sr	Urine	0	0	0	—	—	—	—	—	—	—	—	—
B. ^{32}P	Urine	13	12	7	2	1	0	15.4	8.3	—	29.4-48nCi/l	25nCi/l	—
C. Rare Earths	Urine	1	0	0	0	—	—	—	—	—	—	—	—
D. ^{14}C	Urine	12	13	6	0	1	1	—	7.7	16.7	—	24nCi/l	54nCi/l
E. ^{3}H*	Urine	9	13	6	1	2	0	11.1	15.4	—	1.8μCi/l	0.25μCi/l	—
V. Gamma spectrometry	Urine	110	119	79	1	0	0	0.9	—	—	^{137}Cs**	—	—
VI. Whole-body counting	—	—	37§	—	—	5	—	—	13.5	—	—	10nCi ^{65}Zn - ^{59}Fe(1)§§ 27-500nCi ^{99m}Tc(4)§§ 20nCi ^{125}I, 20nCi ^{129}Cs, 5nCi ^{181}W	

*Determinations made at Lawrence Livermore Laboratory.

**Spectrometry of raw urine.

§Routine whole-body counts during entire reporting period.

§§Numbers in parentheses indicate number of persons in whom activity was observed.

1969 (111 pCi/24-hour urine), and are lower for each successive year in the reporting period. During the year 1968-69 five determinations of ^{3}H were made in the routine program, and all of these were negative. Twenty-eight such determinations were made during the current reporting period, and of these three were positive (10.7%). The increasing importance of tritium as a source of human contamination at LBL is suggested by this fact, and the suggestion is further borne out by the number of suspected exposures to tritium shown in Table II below. The routine whole-body counting program included 102 counts of 37 individuals, one of whom was found to have a total of 10 nCi of ^{65}Zn and ^{59}Fe, four had burdens from 27 to 500 nCi of ^{99m}Tc, one had 5 nCi of ^{181}W, one had 20 nCi of ^{129}Cs, and one had 20 nCi of ^{125}I.

Suspected Exposures

Forty-one persons were involved in incidents in which there was release of radionuclides. Table II shows the method and result of investigation of these incidents. Case number 9 (exposure to ^{241}Am) had a high value of 1800 pCi in a fecal sample delivered the day following the incident. The body burden at this time, estimated by whole-body counting, was 13 nCi. The fecal activity was reduced to 22.4 pCi in three days, by which time the retained amount was no longer detectable by whole-body counting. Except for this single high value, activity found in fecal analysis did not exceed 5.5 pCi for any individual investigated. Table III provides a comparison of results of alpha analysis of urine and feces in cases with suspected exposure. It will be noted that both incidence and level of activity are markedly higher in feces than in urine, indicating the importance of analyzing fecal samples during the period immediately following possible exposure. With respect to incidents involving release of beta-emitting nuclides, the increasing importance of tritium will be readily appreciated (Table II). No previous report from this laboratory has contained any reference to probable exposure to this radionuclide. In each of the three years 1969-72 four persons were suspected of having had such exposure. Results of analysis for 1969-70 were all negative, in 1970-71 one determination was positive, in 1971-72 eleven determinations were made on samples from four

TABLE II

Suspected Exposure Cases

	Radiochemical Analysis					Whole Body Count		
Case No.	Isotope suspected	Material analyzed	No. determinations	No. positive	Range (pCi)	Isotope or energy (MeV)	Activity	Comments
I. Alpha								
A. 1969-70								
1.	Actinides	Urine	1	0	-	-	-	Not counted
2.	Actinides	Urine	1	0	-	-	negative	-
3.	Actinides	Urine	1	0	-	-	negative	-
4.(a)	^{244}Cm	Urine	2	0	-	-	negative	-
		Feces	1	1	1.00	-	negative	-
5.	U	Urine	1	0	-	-	negative	-
6.	^{239}Pu	Urine	1	0	-	-	negative	-
7.	^{239}Pu	Feces	1	0	-	-	negative	-
8.	U	Urine	1	0	-	-	negative	-
9.	^{241}Am	Urine	2	1	0.32/24 hrs	^{241}Am	13 nCi	No detectable activity 3 days later.
		Feces	2	2	22.4-1800.0	^{22}Na	10 nCi	Activity remained through period of observation.
10.	Actinides	Urine	2	2	1.67-2.38/24 hrs(b)	-	negative	-
		Feces	1	1	0.63(b)	-	negative	-
11.	Actinides	Urine	1	1	0.41/24 hrs	-	negative	-
		Feces	1	1	4.25(b)	-	negative	-
12.	^{239}Pu	Urine	1	0	-	-	negative	-
		Feces	3	2	0.75-5.55	-	negative	-
13.	Actinides	Urine	1	1	0.2/24 hrs	-	negative	-
B. 1970-71								
14.	^{241}Am	Urine	2	1	0.38/24 hrs	-	negative	-
15.	^{249}Cf	Urine	2	0	-	-	negative	-
		Feces	1	0	-	-	negative	-
		Nose swipe	1	1	0.12	-	negative	-
16.	^{249}Cf	Urine	2	0	-	-	negative	-
		Feces	1	1	0.725	-	negative	-
		Nose swipe	1	1	0.140	-	negative	-
C. 1971-72								
17.	^{241}Am	Urine	3	2	0.12-1.2/24 hrs	-	negative	-
18.	Actinides	Urine	2	0	-	-	negative	-
19.	Actinides	Urine	2	0	-	-	negative	-
		Feces	1	1	1.69(c)	-	negative	-
II. Beta & Gamma								
A. 1969-70								
20.	Rare Earth	Urine	1	0	-	^{160}Tb	230 nCi	Reduced to 26 nCi in 4 d. Reduced to 14 nCi in 72 d.
	Elements	Feces	1	1	68.00	^{65}Zn	30 nCi	No significant reduction during observation period.
21.	^{180m}Ta	Urine	2	2	1.72-7.8/24 hrs	^{180m}Ta	3.75 nCi	-
22.	^{182}Ta	Urine	2	1	3.4/24 hrs	^{182}Ta	20 nCi	-
23.	^{3}H	Urine	1	0	-	-	-	Not counted
24.	^{3}H	Urine	2	0	-	-	-	Not counted
25.	^{3}H	Urine	1	0	-	-	-	Not counted
26.	^{3}H	Urine	1	0	-	-	-	Not counted
27.	^{14}C	Urine	1	0	-	-	-	Not counted
28.	^{14}C	Urine	1	0	-	-	-	Not counted
B. 1970-71								
29.	^{14}C	Urine	1	0	-	-	-	Not counted
30.	^{3}H	Urine	1	1	0.21 µCi/l	-	-	Not counted
31.	^{3}H	Urine	1	0	-	-	-	Not counted
32.	^{3}H	Urine	1	0	-	-	-	Not counted
33.	^{3}H	Urine	1	0	-	-	-	Not counted
34.	^{32}P	Urine	1	1	7.6 nCi/24 hrs	^{32}P	0.4 µCi	
35.	^{32}P	Urine	1	1	26.8 nCi/24 hrs	^{32}P	1.0 µCi	
C. 1971-72								
36.	^{3}H	Urine	1	1	0.9 µCi/l	-	-	Not counted
37.	^{3}H	Urine	6	6	1.59-1.87 µCi/l	-	-	Not counted
38.	^{3}H	Urine	1	1	1.70 µCi/l	-	-	Not counted
39.	^{3}H	Urine	3	3	0.55-.68 µCi/l	-	-	Not counted
40.	^{210}Pb(d)	Urine	3	0	-	-	-	Not counted
		Feces	1	0	-	-	-	Negative
		Blood(20 ml)	1	0	-	-	-	Negative
41.	^{60}Co ?	Urine	1	0	-	^{107}Cd, ^{60}Co	-	All activity was on clothing.
III. Outside Referrals 1969-72								
1.	^{241}Am	Urine	1	1	0.28 pCi α/24 hr(e)	-	-	Negative
	^{90}Sr				1.66 pCi ^{90}Sr/24 hrs			
					negative for gamma			
	^{137}Cs	Feces	1	1	1.5 pCi α(e)			
					3.5 pCi ^{90}Sr			
					negative for gamma			
2.	Th	Urine	1	0	-			No detectable activity in 3 days.
		Feces	1	1	228 pCi	Th	9 nCi	
3.	Th	Urine	1	0	-			No detectable activity.
		Nose swipe	1	1	2.7 pCi			
4.	Th	Urine	1	0	-			No detectable activity.
		Nose swipe	1	1	0.39 pCi			
5.	Th	Nose swipe	1	1	0.227 pCi			No detectable activity.
6.	Th	Urine	1	0	-	-	-	Not counted.
		Nose swipe	1	0	-			
7.	Th	Urine	1	0	-	-	-	Not counted.
8.	^{99m}Tc	Blood(7 ml)	1	0	-	^{99m}Tc	0.5 µCi	-
9.(4)(e)	^{226}Ra	-	-	-	-	-	-	No detectable activity.
10.(2)(e)	^{182}Ta, ^{7}Be	-	-	-	-			Activity found only on clothing.
11.	^{60}Co	-	-	-	-	^{60}Co	70 nCi	Reduced to 5 nCi in 15 days; 77 nCi excreted in feces over a 4-day period.
12.(3)(e)	^{197}Hg	-	-	-	-	^{197}Hg	0.1-0.3nCi	-
13.	^{203}Hg							
	^{210}Pb	-	-	-	-			No detectable activity.
14.	Unknown	-	-	-	-	0.5 MeV	trace	-
15.	^{235}U,Th	-	-	-	-			No detectable activity.
16.	Unknown	-	-	-	-			No detectable activity.
17.(2)(e)	^{32}P	-	-	-	-	^{32}P	0.5 µCi	-
18.	^{181}W	-	-	-	-		each person	No detectable activity.

(a) Case carried over from previous year.
(b) Isotopes identified: ^{239}Pu; ^{235}U; trivalent actinides.
(c) Isotopes identified: trivalent actinides.
(d) ^{210}Pb assayed by assayed by analysis of ^{210}Po.
(e) Numbers in parenthesis indicate number of individuals

TABLE III

Summary of alpha activity found in cases of suspected exposure.

Time period	Material analyzed	No. individuals	No. determinations	No. positive	% positive	Range (pCi/24 hrs)*
1969-70	Urine	12	15	5	33.3	0.21 - 2.38
	Feces	6	9	7	78.0	0.63 - 1800.0
	Other	--	--	--	--	--
1970-71	Urine	3	6	1	16.7	0.38
	Feces	2	2	1	50.0	0.75
	Other (nose swipes)	2	2	2	100.0	0.12 - 0.14
1971-72	Urine	3	7	2	28.6	0.12 - 1.24
	Feces	1	1	1	100.0	1.69
	Other	--	--	--	--	--
Total period			(Samples)			Range µCi/l
	^{3}H	12	20	13	65.0	0.21 - 1.87

*Time covered in fecal collections not known.

individuals, and all were positive. The single positive value in 1970-71 was 0.21 μCi per liter of urine. The range in 1971-72 was 0.55 to 1.87 μCi per liter.

Outside Referrals

During the reporting period a number of public agencies referred cases of possible exposure to LBL for investigation. A resume of these investigations is given in Table II, Part III. Each referral involved one or more individuals. Cases 8 through 18 were referred for whole-body counting only. Cases 1 through 7 had whole-body counting and radiochemical analysis. Attention is directed to the higher values of both alpha and beta activity in feces as compared to urine. In case number 2 the one fecal sample submitted was collected approximately 5 days after exposure. The rapid loss of thorium from the body within 3 days of the original whole-body count, which was made on the day following exposure, indicates that fecal values had reached a peak before the sample was collected.

Identification of an Unknown Radionuclide

Two previous reports from this Laboratory have described the detection by the whole-body counter of measurable amounts of activity identified by a 60 keV γ ray in individuals working at the 88-inch cyclotron.[1,4] Analysis of urine samples from these individuals had given no evidence of alpha contamination, and it was concluded therefore that the activity observed by the whole-body counter was not due to ^{241}Am. Urine samples were also negative when submitted to gamma spectrometry, indicating a very low rate of urinary excretion, and this was borne out by evidence of prolonged retention shown by whole-body counts. Some workers from the 88-inch cyclotron had had measurable amounts of ^{181}W on their clothing at the time when accelerator workers were being studied.[4] In order to identify the activity found by the whole-body counter, swipes were made from the target and accelerator chamber of the 88-inch cyclotron. These were processed chemically and point sources were made of the isolated activity. Spectrometry with the Si(Li) semiconductor gave the values shown in Table IV, which are in excellent agreement with published values for K x-rays of tantalum, the daughter product of ^{181}W.

TABLE IV

Values for point sources made from 88 inch cyclotron (keV)	Published values for tantalum K x-rays[5] (keV)
56.7	56.3
57.8	57.5
65.0	65.2
66.9	66.9

Agreement of energy of the activity in the swipes with that observed in the workers, together with the half-life of the isolated material (about 100 days) strongly supports the conclusion that contamination of workers is due to ^{181}W.

Research and Development

1. The laboratory has collaborated with the Department of Safety Services in a study of radioactive contamination of wild deer, which will be published elsewhere.[6]

2. The laboratory participated in a study of tissue distribution and excretion of ^{253}Es in mice.[7] The study afforded a means of evaluating the capability of thin NaI crystals for counting L x-rays from ^{253}Es absorbed in bone and soft tissues. Results were compared with alpha activity determined in these tissues and in excreta.

3. Analysis of urine from nonindustrially exposed residents of the Bay Area has continued. The sample now consists of 85 individuals. The incidence of very low level alpha activity among these individuals is approximately the same as that found in routine monitoring of laboratory personnel.

4. An improved method for electrodeposition of trivalent actinide elements has been adopted. The method, first advanced by Sementsova and modified by Allaway, uses a plating medium of 0.05 $\underline{N}$ HNO_3.[8,9] Electrodeposition is carried out at 30 V, 120 mA for 50 minutes. The greatly shortened time as compared with older methods is an advantage, and preparations are cleaner, have less mass, and are less likely to flake.

5. Work has continued with the use of TIOA (triisooctyl amine) and DDCP (di-n-butyl-N-N-diethyl carbamylphosphonate) for solvent extraction of uranium and plutonium, and trivalent actinides respectively.[10] This is a rapid method suitable for routine screening of samples for alpha activity, with recoveries in the range of 75 to 85%.

6. In the course of the reporting period the laboratory acquired a Ge(Li) semiconductor. A number of point sources have been prepared and used in the calibration of this instrument.

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