White Paper

NIOSH Response to SC&A Comments Concerning Part of Issue 2 Regarding the Internal Monitoring Program at the Lawrence Berkeley National Laboratory

December 4, 2013

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Summary:

As a result of the Lawrence Berkeley National Laboratory (LBNL) Work Group's (LBNLWG) Meeting on February 3, 2012, SC&A reviewed exposure potential from internal emitters at LBNL for the post-1961 period to determine their significance and whether bioassay monitoring was complete and adequate for that time period. SC&A sampled available dosimetry records for pertinent information regarding what exposure potential existed in which operations and buildings, and whether monitoring was available and adequate (and addressed in the current TBD). This report is a response to those comments. The reader is referred to SC&A's original report for their full text (Draft White Paper: SC&A Review of Lawrence Berkeley National Laboratory Site Profile Matrix Issue #2, September 2012).

SC&A reviewed post-1961 records available in the Site Research Database (SRDB), evaluated dosimetry program documentation and dose information, and compared its assessment with that provided in the TBD. SC&A indicated apparent gaps and differing assessments of bioassay program adequacy in their review, with an emphasis on what programmatic or dosimetric shortcomings would have the most significant influence on dose reconstruction.

SC&A's review under site profile Matrix Issue #2 raised two central questions for Work Group consideration:

1. Exposure potential posed by radionuclide source terms for which adequate bioassay monitoring may be lacking (or which are not addressed by the TBD), making sufficiently accurate dose reconstruction problematic; and

2. Inadequate management of the bioassay program at LBNL making bioassay results less reliable for use in dose reconstruction.

For question (1), SC&A questioned how LBNL could have adequately monitored for short-lived Mixed Activation Products (MAPs) using in vitro gross alpha and beta techniques, coupled with whole-body counting (WBC), when MDA detection thresholds and extended monitoring periods would have mitigated against detection.

For question (2), SC&A indicated that NIOSH has accepted the inauguration of the LBNL bioassay program in 1961 as the threshold of a comprehensive and reliable program for internal dosimetry (and therefore the end of the SEC-covered period), while SC&A believes pertinent program documentation (e.g., DOE audits in the 1980s) clearly highlight persistent inadequacies in how the program was managed that bear directly on data reliability.

Based on information in the Discussion section below, NIOSH has determined that based on a review of pertinent literature, and LBNL accelerator air sample data , short-lived MAPs, such as C-11, N-13, and O-15, produced in LBNL accelerators were not a radiological concern.

NIOSH has determined during site research and the SEC evaluation that LBNL had an operational bioassay monitoring program in 1962. Numerous documents and memos discussing the program lead to conclusion that all individuals who worked with uncontained radioactive materials were monitored for intakes of radioactive material, and that their individual bioassay results can be used to assess their internal dose.

Discussion:

Issue 2: Insufficient information for internal dose reconstruction, especially during the early years

This issue involves SC&A comments concerning the following:

1. Mixed Activation Products;
2. Adequacy of Gross Alpha Urinalysis Procedure; and
3. Fecal Bioassay versus Urinalysis Bioassay.

I. NIOSH response to SC&A's comments regarding exposure potential for which adequate bioassay coverage may be lacking:

Mixed Activation Products (MAPs):

SC&A categorized the MAPSs above as C-11, N-13, O-15, Ar-41, Be-7, and others.

Various reports and studies have been conducted concerning airborne radioactivity at high energy accelerators. Two reports are Airborne Radioactivity Produced at High Energy Accelerators (Rindi 1967), and Aerosol and Dust Radioactivity in the Halls of High Energy Accelerators (Charalambus 1967). These reports, as well as others, have been the basis for categorizing the airborne radionuclides of inhalation concern at high energy accelerators. The findings and conclusions from these reports have been summarized in Accelerator Health Physics (Patterson 1973).

Airborne radioactivity inhalation concerns at high energy accelerators result from activated gases and activated dust and aerosols (Rindi 1967, Charalambus 1967).

During accelerator operation radioactive gases are produced by the interaction between the primary and secondary particles from the machine and the atmospheric air in the accelerator halls. Spallation reactions in solid machine parts can also lead to the formation of radioactive gases (Rindi 1967).

Radioactive dust is produced from activated machine parts by normal erosion or mechanical wear. Dust in the accelerator halls is also activated when it is in suspension in the air or when it is deposited on the machine components. However, experience shows that the principal radioisotopes found in dust are those produced from the constituents of the machine parts (Charalambus 1967).

Lists in various documents of radionuclides that can be produced in air at accelerators vary as to the number presented based on the reactions that form them. A list of radionuclides with half-lives greater than 10 minutes that can be produced in air at accelerators is shown in Table 1. This is based on a list from Thomas (1978).

Table 1. Radionuclides with Half-life > 10 Minute that can be Produced in Air at Accelerators*

Radionuclide	Half-Life	Emission	Parent Element	Production Reaction
H-3	12.2 years	β^-	C	Spallation
			N	Spallation
			O	Spallation
Be-7	53 days	γ, EC	C	Spallation
			N	Spallation
			O	Spallation
			Ar	Spallation
C-11	20.5 min	β^+	C	Spallation
			N	Spallation
			O	Spallation
			Ar	Spallation
F-18	1.85 hr	β^+, EC	Ar	Spallation
Na-22	2.6 years	β^+, γ	Ar	Spallation
Na-24	15 hr	β^-	Ar	Spallation
Mg-28	21.3 hr	β^-, γ	Ar	Spallation
Al-28	2.3 hr	β^-, γ	Ar	Spallation
Si-31	2.6 hr	β^-, γ	Ar	Spallation
P-32	14.3 days	β^-	Ar	Spallation

P-33	25 days	β^-	Ar	Spallation
S-35	87 days	β^-	Ar	Spallation
Cl-34m	32.4 min	β^-, γ	Ar	Spallation
Cl-38	37.3 min	β^-, γ	Ar	(γ, pn)
Cl-39	55 min	β^-, γ	Ar	(γ, p)
Ar-41	1.8 hr	β^-, γ	Ar	(n, γ)

* Pertinent data excerpted from Thomas 1978.

Argon-41 is an inert gas and not considered an internal hazard, but an external hazard determined via immersion in the radioactive cloud. Nuclides with half-lives less than 10 minutes are also not considered internal exposure hazards because there is insufficient time for the material to be distributed to the organs.

Accelerator Health Physics indicates that the gaseous airborne radioactivity concentrations measured are of extremely short duration after accelerator turnoff, because of the short half-lives of the important nuclides and dilution with inactive air (Patterson 1973). Also, workers are not, in general, continuously exposed to short-lived gaseous airborne radioactivity produced in accelerators for 40 hours per week.

The same reference indicates regarding accelerator-produced gaseous airborne radioactivity that, "long-lived activities, on the other hand, may be discounted because of their low production rate. In usual facilities, where complete air changes occur 2 to 3 times per hour, even normal leakages may amount to 10% of the volume of enclosed air per hour. Unless special steps are taken to prevent air from leaving the accelerator room its residence time is considerably less than 1 day. It is not possible, therefore, to produce Be-7 or H-3 at levels higher than a small fraction of saturated specific activity."

The conclusions of several studies have shown that the potential exposure from radioactive dust is negligible (Patterson 1973). The most frequently identified radionuclides in dust at a high energy proton accelerator were Mn-54, Be-7, Cr-51, Fe-59, and V-48, and are shown in Table 2 (Patterson 1973). Mn-54, Cr-51, Fe-59, and V-48 are produced in the accelerator magnets, and vacuum chambers (Charalambus 1967). Be-7 can be produced in the accelerator coils (Charalambus 1967), as well as activation of air.

Table 2. Radionuclides Identified in Dust Samples at CERN Proton Synchrotron (after Charalambus and Rindi) (Patterson, 1973)

Radionuclide	Decay Mode	Relative Quantity (%)

Mn-54	EC, γ	~50
Be-7	EC, γ	~25
Cr-51	EC, γ	~7
Fe-59	β^+, γ	~9
V-48	β^+, γ	~9

An essentially identical conclusion regarding the potential exposure from radioactive dust has been reached by studies at the Saclay electron linac operating at high power (330 to 560 MeV, 100 kW on target) (Patterson 1973).

The above list (Table 2) of radionuclides identified in dust samples at CERN (Charalambus 1967) contains only a small fraction of radionuclides that can be produced in the principal materials of the accelerator. A list of the main radionuclides that can be produced by the interaction of primary and secondary particles from a high-energy proton accelerator with the principal materials (shielding, magnets, coils and vacuum chamber) in the machine are shown in Table 3 (Charalambus 1967).

Table 3. Main Radionuclides Produced by the Interaction of Primary and Secondary Particles from a High-Energy Proton Accelerator with the Principal Materials in the Machine (Charalambus 1967)*

Radionuclide	Half-life	Decay Mode	Parent Isotope (machine part)[a]	Type of reaction
Na-22	2.6 y	β^+, γ	Na-23(S)	(n,2n)
			Na-23(S)	(p,pn)
			Na-23(S)	{p,pn)
			Mg-24(S)	(p,2pn)
			Al-27(M)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Co-60	5.2 y	β^-, γ	Co-59(M)	(n,γ)
			Ni-59(M)	(n, p)
			Cu-63(M)	(n,α)
			Cu-63(M)	(p,spallation)
			Cu-63(M)	(p,spallation)
Sc-46	84 d	β^-, γ	Ti-48(M)	(p,2pn)

Radionuclide	Half-life	Decay Mode	Parent Isotope (machine part)[a]	Type of reaction
			Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Mn-54	300 d	EC, γ	Mn-55(M)	(n,2n)
			Fe-54(M)	(n, p)
			Fe-56(M)	(p,2pn)
				(n,p2n)
Fe-59	45 d	β^-, γ	Fe-58(M)	(n,γ)
			Co-59(M)	(n, p)
			Ni-60(M)	(n,2p)
			Cu-63(M)	(p,spallation)
Co-58	71 d	EC, β^+, γ	Co-59(M)	(n,2n)
			Ni-58(M)	(n, p)
			Ni-60(M)	(n,2pn)
			Cu(M)	(p,spallation)
Zn-65	245 d	EC, β^+, γ	Cu-65(M)	(p,n)
V-48	16.1 d	EC, β^+, γ	Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Co-56	77.3 d	EC, β^+, γ	Co-59(M)	(p,spallation)
			Ni-58(M)	(p,2pn)
			Ni-60(M)	(p,spallation)
			Cu-63(M)	(p,spallation)
P-32	14.3 d	β^-	P-31(M)	(n,γ)
			S-32(S)	(n, p)
			K-39(S)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Ca-45	164 d	β^-	Ca-44(S)	(n,γ)
			Ti-48(M)	(p,spallation)
			Cr-52(M)	(p,spallation)

Radionuclide	Half-life	Decay Mode	Parent Isotope (machine part)[a]	Type of reaction
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
K-43	22.4 h	β^-, γ	Ca-44(S)	(n,pn)
			Fe-56(M)	(p,spallation)
K-42	12.4 h	β^-, γ	K-41(S)	(n,γ)
			Ca-44(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Mn-52	5.6 d	EC, β^+, γ	Mn-55(M)	(p,3np)
			Fe-56(M)	(p,spallation)
Na-24	15 h	β^-, γ	Na-23(S)	(n,γ)
			Mg-24(S)	(n,p)
			Al-27(M)	(n,α)
			Al-27(M)	(p,3pn)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Sc-48	44 h	β^-, γ	Ti-48(M)	(n,p)
			Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Co-57	270 d	EC, γ	Co-59(M)	(p,p2n)
			Ni-58(M)	(n,pn)
			Ni-58(M)	(p,2p)
			Ni-60(M)	(p,spallation)
			Cu(M)	(p,spallation)
S-32	87 d	β^-	K-30(S)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
P-33	24.4 d	β^-	K-39(S)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Sc-47	3.4 d	β^-, γ	Ti-48(M)	(n,pn)

Radionuclide	Half-life	Decay Mode	Parent Isotope (machine part)[a]	Type of reaction
			Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Ni-65	2.6 h	β^-, γ	Ni-64(M)	(n,γ)
			Cu-65(M)	(n,p)
Mn-56	2.6 h	β^-, γ	Mn-55(M)	(n,γ)
			Fe-56(M)	(n,p)
			Fe-57(M)	(n,pn)
Fe-55	2.6 y	EC	Fe-54(M)	(n,γ)
			Fe-56(M)	(n,2n)
			Mn-55(M)	(p,n)
			Co-59(M)	(p,spallation)
Si-31	2.6 h	β^-, γ	Si-30(S)	(n,γ)
			P-31(M)	(n,p)
			K-39(S)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Cu-64	12.9 h	EC, β+, β^-, γ	Cu-63(M)	(n,γ)
			Cu-65(M)	(n,2n)
			Cu-63(M)	(p,pn)
Be-7	53 d	EC, γ	C-12(S	(p,spallation)
			O-16(S,M)	(p,spallation)
			Na-23(S)	(p,fragmentation)
			Al-27(M)	(p,fragmentation)
			Si-28(S)	(p,fragmentation)
			Ca-40(S)	(p,fragmentation)
			Fe(M)	(p,fragmentation)
			Fe(M)	(p,spallation)
Cr-51	27.8 d	EC, γ	Cr-50(M)	(n,γ)
			Cr-52(M)	(n,2n)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)

Radionuclide	Half-life	Decay Mode	Parent Isotope (machine part)[a]	Type of reaction
Ni-57	36 h	β^+, γ	Ni-58(M)	(n,2n)
			Ni-60(M)	(p,p3n)
			Ni-58(M)	(p,pn)
			Cu-63(M)	(p,spallation)
Mg-28	21.3 h	β^-, γ	Si-29(S)	(n,2p)
			Si-30(S)	(n,2pn)
			P-31(M)	(p,spallation)
			Ca-40(S)	(p,spallation)
			Fe-56(M)	(p,spallation)
Sc-43	3.9 h	β^+, γ	Ti-48(M)	(p,spallation)
			Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Sc-44	2.4 d	γ	Ti-48(M)	(p,3p3n)
Sc-44	4 h	β^+, EC, γ	Cr-52(M)	(p,spallation)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Ti-45	3.1 h	EC, β+	Ti-46(M)	(n,2n)
			Cr-52(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
V-49	330 d	EC	Cr-50(M)	(n,p)
			Cr-52(M)	(p,2p2n)
			Mn-55(M)	(p,spallation)
			Fe-56(M)	(p,spallation)
Co-55	18 h	β^+, γ	Co-59(M)	(p,3p3n)
			Ni-58(M)	(p,2pn)
			Ni-60(M)	(p,spallation)
			Cu-63(M)	(p,spallation)

a - (S) = shielding, (M) = magnets, coils, and vacuum chambers.

* Pertinent data excerpted from Charalambus 1967.

LBNL recognized the issue of short-lived MAPs early on in its accelerator programs. LBNL indicated that for the 60-inch and 184-inch cyclotrons, "From the health hazard viewpoint we feel that the situation is, under existing beams and energies, marginal and readily controllable by existing exhaust ventilation" (Thaxter 1961). The same report indicated that the C-11, N-13, and O-15 combined maximum air concentration in the 184-inch cyclotron was approximately $2x10^{-7}$ $\mu Ci/cm^3$, and were below MPC. Since N-13 and O-15 have half lives less than 10 minutes, C-11 is the only measurable radionuclide of concern for internal exposures. Carbon-11 was approximately 25 percent of the total activity measured (Thaxter 1961). This results in a maximum C-11 air concentration of approximately $5.0x10^{-8}$ $\mu Ci/cm^3$, and is approximately 40 times less than its MPC_a of $2x10^{-6}$ $\mu Ci/cm^3$. The C-11 maximum air concentration of $5.0x10^{-8}$ $\mu Ci/cm^3$ yields a maximum dose of approximately 1 mrem, using the limiting ICRP 68 dose conversion factor for the pancreas of $8.89x10^{-3}$ mrem/μCi, and a breathing rate of 2400 m^3/yr (40 hours per week for 50 weeks per year). The 1 mrem dose is conservative, given that the maximum level of C-11 inhaled is assumed to be constant, and a worker would not be expected to be exposed to this level for 40 hours per week. In addition, dilution from ventilation, radioactive decay, and accelerator off time would reduce the C-11 dose to less than 1 mrem. The 60-inch cyclotron maximum air concentrations approached $1x10^{-9}$ $\mu Ci/cm^3$ for N-13, N-16 and O-15 measured, and were well below MPC_a guides. N-13, N-16 and O-15 all have half- lives less than 10 minutes.

The 88-inch cyclotron minimized the possibility of contamination by means of airborne radioactivity by forced-air ventilation at negative pressures of the cyclotron vault and pit areas. Filtered high-bay air is pulled by blower through the vault floor and roof, and is then ducted from the cyclotron pit area to a monitored exhaust in the updraft of the water cooling tower (LBNL 1979).

Bevatron safety procedures indicated that both sources of airborne radionuclides were recognized at the accelerator (gaseous isotopes produced in air and vented gases as well as airborne radioactive dust) as a result of thermal neutron capture, high-energy particle spallation, and (gamma, n) reactions. Due to relatively low average beam and methods of beam transfer, i.e., in vacuum, the yield from either mode at the Bevatron is small compared to MPC guides (Everette 1976).

A review was conducted of available LBNL accelerator air sample data for several years. The results of these air samples, along with dose estimates using the limiting ICRP 68 dose conversion factor, are shown in Table 4. The doses assume continuous occupancy for 40 hours per week and 50 weeks per year. As indicated previously, workers are not, in general, continuously exposed to short-lived gaseous airborne radioactivity produced in accelerators for 40 hours per week. As can be seen from the results, and accounting for intermittent exposure, all

the doses are less than 1 mrem. A limited review of various air sample report notations also indicate that the beta and gamma activities measured were much less than 1% of the MPC or DAC. Based on a review of available air sample data, NIOSH believes that the beta and gamma activities in the LBNL accelerators likely constitute a very small, if not negligible, internal dose potential.

The Health Chemistry Department, which later became part of the Environmental Health and Safety Department, maintained strict control of the movement of radioactive materials at LBNL. They maintained a stock of target holders, inspected and leak-tested the loaded target holder, and delivered it to the accelerator when satisfactory, operated the bombardment apparatus at the 88-inch cyclotron, and delivered the bombarded target to the experimenter and assisted with disassembly (UCRL 1964).

The use of glove boxes for the handling of dispersible radioactive material was the policy early on at LBNL. Work enclosures, including glove boxes, constituted the basic component for handling large or small quantities of the various types of radioactive materials at LBNL (Garden 1960). Environmental Health and Safety Department procedures indicated that ventilated gloved boxes were to be used for all radioactive operations that could generate aerosols, or the possible spread of contamination (EH&S 1981). The glove box use requirement for dispersible radioactive material is also indicated in various accelerator operational safety procedures, some of which are cited below.

Along with target handling control at LBNL, the accelerator target rooms and laboratories that handled radioactive materials by design were at negative pressure (UCRL 1964), thereby preventing outflow of any potential airborne activity releases into the surrounding areas. Enclosing targets in a sealed target holder and ventilation flow control was necessary in accelerator facilities to prevent contamination of the experimental cave areas, as well as the accelerator machine.

Various Operational Safety Procedures (OSPs) for the LBNL accelerators; Bevatron/Bevalac, SuperHILAC, 184 inch cyclotron, and 88 inch cyclotron, indicate that access to the cave areas where the targets were bombarded, as well as the accelerator machine were controlled by safety interlocks to prevent accidental exposure (LBL 1972, 1979, 1982, 1984).

Table 4. LBNL Accelerator Air Activity Concentration Information (LBL 1965, LBL 1967, LBL 1975, LBL 1988, EH&S 1995)

Year	Location	Radionuclide	Activity (pCi)	Activity Concentration (pCi/m^3)	Half Life	ICRP 68 Maximum Organ DCF (mrem/pCi)	Dose to Maximum Organ[2] (mrem)	ICRP 68 Maximum Organ
1965	Building 51 (Bevatron) NDT	Be-7	20000	20	53.3 d	1.56E-06	7.49E-02	ET
		Na-24	20000	10	15.0 h	4.81E-05	1.15E+00	ET
		P-32	1500	1.5	14.29 d	5.93E-05	2.13E-01	Lungs
		3 day $T_{1/2}$	1500	1.5				
1965	Building 6 Platform (184 inch Cyclotron)	Be-7	--	30	53.3 d	1.56E-06	1.12E-01	ET
		P-32	--	1.8	14.29 d	5.93E-05	2.56E-01	Lungs
1987	88 inch Cyclotron Caves							
	Cave 3	Be-7[4]	--	240	53.3 d	1.56E-06	8.99E-01	ET
	Cave 2	Be-7[4]	--	22	53.3 d	1.56E-06	8.24E-02	ET
	Cave 4C	Be-7[4]	--	16	53.3 d	1.56E-06	5.99E-02	ET
	RAMA	Be-7[4]	--	18	53.3 d	1.56E-06	6.74E-02	ET

Year	Location	Radionuclide	Activity (pCi)	Activity Concentration (pCi/m^3)	Half Life	ICRP 68 Maximum Organ DCF (mrem/pCi)	Dose to Maximum Organ[2] (mrem)	ICRP 68 Maximum Organ
1988	88 inch Cyclotron High Level Cave	Sr-89/Y-89m	1000	1	50.5 d	2.52E-05	6.05E-02	Bone Surface
		Tc-96	1000	1	4.28 d	6.67E-05	1.60E-01	ET
		Y-87/Sr-87m	600	0.6	80.3 h	2.33E-05	3.36E-02	ET
		Ru-97	700	0.7	2.9 d	9.26E-06	1.56E-02	ET
		Mn-52	40	0.04	5.591 d	8.89E-05	8.53E-03	ET
1989	88 inch Cyclotron E.AL-2	Ce-144	120	0.12	284.3 d	8.15E-04	2.35E-01	Lungs
		Eu-152	30	0.03	13.33 y	6.67E-04	4.80E-02	Liver
		Ba-133	15	0.015	10.74 y	3.56E-05	1.28E-03	Bone Surface
		Mn-54	Trace	Trace	312.5 d	2.70E-05	Trace	ET
1990	88 inch Cyclotron E.AL-2[4]	Co-57	300	0.15	270.9 d	1.37E-05	4.93E-03	Lungs
		Se-75	100	0.05	119.8 d	2.78E-05	3.34E-03	Kidneys
		Mn-54	100	0.05	312.5 d	2.70E-05	3.24E-03	ET
		Zn-65	3500	1.75	243.9 d	2.74E-05	1.15E-01	ET
		Na-22	1000	0.5	2.602 y	7.41E-05	8.89E-02	ET
		Co-60	40	0.02	5.271 y	3.56E-04	1.71E-02	Lungs

Year	Location	Radionuclide	Activity (pCi)	Activity Concentration (pCi/m^3)	Half Life	ICRP 68 Maximum Organ DCF (mrem/pCi)	Dose to Maximum Organ[2] (mrem)	ICRP 68 Maximum Organ
		Co-58	300	0.15	70.80 d	3.52E-05	1.27E-02	ET
		Co-56	Trace	Trace	78.76 d	1.07E-04	Trace	ET
		Unidentified alpha	250	0.25				
1990	88 inch Cyclotron High Level Cave	Be-7	43	0.43	53.3 d	1.56E-06	1.61E-03	ET
		Na-22	40	0.04	2.602 y	7.41E-05	7.11E-03	ET
		Co-57	17	0.017	270.9 d	1.37E-05	5.59E-04	Lungs
		Mn-54	Trace	Trace	312.5 d	2.70E-05	Trace	ET
		Co-60	Trace	Trace	5.271 y	3.56E-04	Trace	Lungs
		Se-75	Trace	Trace	119.8 d	2.78E-05	Trace	Kidneys
		Co-58	Trace	Trace	70.80 d	3.52E-05	Trace	ET
		Zn-65	370	0.370	243.9 d	2.74E-05	2.43E-02	ET
1990	88 inch Cyclotron High Level Cave	Be-7	80	0.08	53.3 d	1.56E-06	3.00E-04	ET
		Ta-182	12	0.012	115 d	1.96E-04	5.64E-03	Lungs

1 – Some air sample results were in pCi activity only. The air sample volume assumed is 1000 cubic meters based on the 1988 88-inch Cyclotron High Level Cave one week air sample. This is also found in a notation for the 1965 Bevatron one week air sample listed in the above table (LBL 1967). The 4 cubic feet per minute air sampler flow rate is also confirmed based on EH&S information on air sampling [EH&S 1981]. Four cubic feet per minute corresponds to a 1

week sample volume of 1142 cubic meters. All activities that were converted to activity concentrations in the table for 1989 and later are based on the nominal air sample volume of 1000 cubic meters.

2 – Breathing rate of 2400 cubic meter of air inhaled per work year assumed based on 40 hours per week and 50 weeks per year.

3 – Two week air sample was indicated for this result (EH&S 1995) The activity results are divided by 2000 cubic meters.

4 – Air sample report notation indicates that the activity is mostly Be-7.

Targets required cooling times and were surveyed prior to removal and transport from the cave target area to prevent external dose overexposure. In addition, the various accelerator OSPs required ventilated gloved boxes to be used for all radioactive operations that generate aerosols or cause possible spread of contamination or significant personnel exposure. Gloved box pass-outs and pass-ins were required to be done with the assistance of an EH&S Department monitor (EH&S 1981).

Health Chemistry maintained a supply of air samplers in locations where accidental releases of airborne radioactivity was likely (UCRL 1964). Health Chemistry also maintained permanently installed air samplers which operated continuously in various locations. The air samples were changed weekly in locations of moderate experimental activity, and daily in locations of intense experimental activity. Glove box manifolds and rooms that contained glove boxes were air sampled.

SC&A cited a 1968 LBNL Report of the Bioassay Program (LBL 1968) that indicated that positive gross beta urinalysis for a survey of 85 accelerator workers was in the range of 1-42 pCi/24 hour sample, and that these levels were comparable to elevated beta activity in samples of surveyed residents in the San Francisco Bay Area. The 1968 Report of the Bioassay Program indicated that urine bioassay results of San Francisco residents ranged between 5 to 20 pCi/24h of beta activity.

SC&A interpreted the 1968 Report of the Bioassay Program to mean: *"It is likely that short-lived MAPs contributed to this constant low-level beta component (particularly with the finding in LBL 1968 that was unlikely due to Sr-90), but lack of detection and discrimination due to WBC sensitivity and delay in monitoring made it unlikely that these would be picked up, and this survey "finding" apparently was the reason LBNL chose not to resolve this unknown exposure source term."*

The 1968 Report of the Bioassay Program (LBL 1968) states regarding the above, *"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."* It therefore appears that LBNL concluded that the unknown exposure was due to environmental exposures. It is not clear to NIOSH how a "*lack of detection and discrimination due to WBC sensitivity and delay in monitoring*" would have resulted in comparable gross beta results between LBNL accelerator employees and bay area residents.

A review of gross beta urinalysis bioassay reports through the mid 1970's showed notations on the results which appear to indicate that the elevated gross beta activity levels detected in urine samples are from environmental sources including fallout (LBNL 1961):

"GROSS BETA" ACTIVITIES FROM 5 TO 80 DPM PER 24 HOUR URINE COLLECTION FROM WHICH 40-K AND 137-CS HAVE BEEN REMOVED ARE COMMONLY OBSERVED DUE TO ENVIRONMENTAL SOURCES INCLUDING FALLOUT AND ARE NOT NECESSARILY INDICATIVE OF OCCUPATIONAL EXPOSURE."

The Bioassay Laboratory Manual of Procedures (Buckley 1969) included a similar statement regarding gross beta activities. Gross beta notations on bioassay reports from the later 70's and beyond contained the following similar notation:

"GROSS BETA" ACTIVITIES UP TO 20 DPM PER 24 HOUR URINE COLLECTION FROM WHICH 40-K AND 137-CS HAVE BEEN REMOVED ARE COMMONLY OBSERVED DUE TO ENVIRONMENTAL SOURCES INCLUDING FALLOUT AND ARE NOT NECESSARILY INDICATIVE OF OCCUPATIONAL EXPOSURE."

A 1969 Health Physics Journal article titled, "Whole-Body Counting and Bioassay Determinations Made on Accelerator Workers" provided a peer reviewed article regarding the study of the LBNL accelerator workers mentioned above (HPS 1969). The conclusions of the article regarding the internal assessments performed on the accelerator workers was, "*Accelerator workers who have been studied in this survey show the following incidence of radioactive contamination: alpha (confirmed) 4%, gamma (by whole-body count) 6%, beta 94%. It is believed that the high incidence of beta activity represents environmental contamination. The levels of activity found do not exceed 1 pCi per 24-hr urine for alpha emitters, and 10 nCi per whole-body count for gamma activity. Accordingly, no changes in existing procedures are contemplated as a result of this survey, since it seems evident that normal habits of cleanliness and occasionally the use of protective clothing and gloves are adequate to insure that the ordinary accelerator worker at LRL-Berkeley will not receive radiation exposures of any consequence from internal sources. Nevertheless, it is believed that periodic surveys of the type reported herein have value in helping to maintain safe working conditions and in bringing to light any unsuspected instances of possibly significant contamination of workers."*

Despite the incidence of some low-level internal beta contamination found, the conclusion was that the lower internal burden found in personnel working with the newer accelerators at LBNL indicated great improvement in operational procedures. The observation of zero or low body burdens in the people indicated adequate standards of cleanliness in working conditions and personal hygiene of accelerator personnel. The above notations and references indicate that LBNL had determined that the gross beta urinalysis results in the range discussed in the 1968 LBNL Report of the Bioassay Program, as well as those in subsequent bioassay report results for the site, were from fallout and environmental sources.

In addition, various procedures and requirements were cited earlier for target handling, accelerator cave safety interlocks, radioactive materials transport, and radioactive materials controls in using glove boxes and enclosures for dispersible radioactive material, negative ventilation flow in areas that handled dispersible radioactive material, and air sampling where accidental releases of airborne radioactivity was likely. This information indicates that LBNL had a strong operating philosophy of containing and minimizing the potential of airborne radioactivity in the work environment. Given the sealed target holder, target disassembly in a glove box and handling of dispersible radioactive material in a glove box, any short-lived radioactive material that may have been created in the accelerator target during bombardment would be contained in the ventilated glove box, making the potential for internal exposure remote. In addition, LBNL's policy was to bioassay individuals who worked with or around dispersible radioactive materials. Various accelerator airborne radioactivity studies indicate that airborne radioactivity produced at high energy accelerators from activated gases, including short-lived MAPs and activated dust and aerosols were not a radiological concern. Furthermore, it is believed based on a review of LBNL accelerator air samples activity results, that the internal dose potential from mixed activation products at LBNL's accelerators is very small, if not negligible.

***Adequacy of Gross Alpha Urinalysis Procedure**:*

SC&A Issue:
For alpha emitters, in general, SC&A had raised questions in its site profile review about how the TBD "does not discuss the fact that many radionuclides present at LBNL would not have been detected by gross measurements, or at least detected with low recoveries and resulting high MDCs [MDAs]… the potential missed dose associated with non-specific bioassay techniques should be further investigated to determine the impact on internal dose calculations" (SC&A 2010).

NIOSH Response:
The bioassay procedures manual (LowBeer 1964) gross alpha method indicates that, "The method is applicable to thorium, plutonium, actinium, americium, curium, neptunium, californium, and einsteinium." There were also separate bioassay procedures for uranium, radium, and polonium (LowBeer 1964).

***Fecal Bioassay versus Urinalysis Bioassay*:**

SC&A Issue:
SC&A cites a report (LBL 1968), where fecal analysis confirmed uptakes that urinalyses and WBCs missed. SC&A also indicates that given the laboratory's sparing use of fecal analysis confirmation (typically only used following an incident), it can be concluded that substantial

missed alpha dose may have existed at LBNL, for which NIOSH's bounding method may not be adequate. As recommended in its 2010 site profile review, SC&A believes the magnitude of this missed dose should be assessed in the context of actual monitoring results, such as those cited above, and the basis for the TBD revisited in this regard.

NIOSH Response:
The noted fecal samples were collected in response to known incidents; this is common procedure for many bioassay programs. Fecal samples can be much more sensitive than urine samples if collected shortly after an intake, but later samples (usually within a week to 10 days) will be less sensitive. They are not necessarily more reliable or superior for assessing an intake, particularly if not linked to a known intake date; urine sample results are less subject to variation. Missed dose can be based on the urine samples, with associated MDAs, from the routine bioassay program.

II. NIOSH Response to SC&A's Comments Regarding Programmatic Issues Affecting Adequacy of Bioassay:

This issue involves SC&A comments concerning the following:

1. Compliance with LBNL bioassay submission requirements; and
2. Program reliability: Selection of personnel and radionuclides.

***Compliance with LBNL Bioassay Submission Requirements*:**

SC&A Issue:
SC&A cites several LBNL Bioassay Program Laboratory reports indicating that delinquent samples were an issue. It also cited a 1987 DOE audit about delinquent samples regarding the LBNL bioassay monitoring program.

NIOSH Response:
Late bioassay submittals do not necessarily mean that the bioassay information would be missing from a worker's DOE records received for processing a worker's claim, unless the sample result is completely missing. Delinquent bioassay samples were followed up with notifications to the worker in question and generally delinquent bioassay samples were collected from individuals (Low-Beer 1960, LBNL 1985). Also, a LBNL 1983 internal audit of the bioassay lists concerning delinquent samples indicated, "*Each month 10 - 20 requests for samples are sent out by (Name Redacted). After a few weeks, if a sample is not returned, a follow up letter is sent. After about five weeks of no response, a list showing delinquent samples is sent to (Name Redacted) and the ASC's for follow up. From February to July 1983, about 96% compliance*

was achieved (Young 1983). Copies of individual bioassay results were distributed to their medical file, and Safety Services (Hartsough 1974).

***Program Reliability: Selection of Personnel and Radionuclides*:**

SC&A Issue:
SC&A cites LBNL reports and memoranda from the 1960s through the 1990s that point to a management system and culture that would have mitigated against a comprehensive personnel selection process addressing what can be a constantly changing experimental work environment involving a myriad of different radionuclide sources with exposure potential. The adequacy and completeness of LBNL's bioassay data bear directly on whether potentially exposed employees were properly identified and enrolled for bioassay sampling, along with the identity of radionuclides to which they may have been exposed. Coupled with the historic lack of compliance by employees and lack of enforcement by management, and the lack of quality assurance performance checks, the degree of adequacy and completeness of bioassay data is uncertain, at least until the early to mid-1990s, when more formal management systems were put into place at LBNL. SC&A also referred to a 1985 DOE audit of LBNL, citing a laundry list of items.

NIOSH Response:
Based on a review of the LBNL's bioassay program information, NIOSH maintains that LBNL had an operational bioassay monitoring program since 1962 and individuals who worked with uncontained radioactive materials were internally monitored, and their individual bioassay results are available and can be used to assess their internal dose. The pre-1962 era was recommended as an SEC based on the absence of an internal dose program and available records.

Generally, sources in the SRDB indicate that there was a considerable management commitment to radiation safety, as is evidenced by numerous files containing bioassay lists and discussions of who should be monitored (e.g. Grill 1966, Howe 1961). Discussions include whether or not a given worker worked with radioactivity or whether he belonged to a group who does. Also, indications are that area monitoring was relied on to pick up any unforeseen contamination incidents that could expose unmonitored workers (e.g. in 1968 an air sample was able to pick up a leaking Cm-244 source, which lead to a number of non-routine samples (Soule 1964). LBNL maintained air samplers in locations where releases of airborne radioactivity were likely. The air samples were changed weekly or daily.

The Health Chemistry Department maintained personnel in key experimental facilities. The normally staffed locations were Buildings 1, 3, 19, 8, 70, 70A, 71, 74, and 88 (UCRL 1964). When the Health Chemistry Department later became part of the Environmental Health and Safety Department (EH&S), it continued to maintain personnel in key experimental facilities.

The normally staffed locations were Buildings 1, 3, 51, 70, 70A, 71, 74, and 88 (EH&S 1981). LBNL maintained bioassay lists from the start of the bioassay program to 1996 when the bioassay program changed such that personnel were selected for operational bioassay based on the radionuclide authorization program and reviews of work performed. Bioassay lists of names of individuals were supplied by the Health Chemistry Department to the Bioassay Laboratory (Various 1961-1964). This practice continued when the Health Chemistry Department became part of EH&S, providing names of individuals who were to be monitored for intakes based on work performed and work location to the Medical Services Department (Haley 1979).

The bioassay lists included the worker name, their location, and specific bioassay analyses to be performed. These lists were updated periodically based on individuals being added and deleted from the list. Delinquents were tracked, and notifications sent. Some bioassay lists included the types of radionuclides the employee worked with (e.g. LBL 1988). In one bioassay list case there is a note that the worker refused to participate in the program. The level of detail compiled in those lists indicates that bioassay analyses were not taken lightly and that management was dedicated to the program.

LBNL also provided bioassay reports containing statistics as to how many particular bioassays were performed during the period, number of negative results, number of positive results, result ranges, and list of individuals who were not sampled (Unknown 1980).

A review of present LBNL claims at the time of this research (February 2013) indicates internal monitoring for various job descriptions and supports that LBNL monitored workers across varied job disciplines based on exposure potential. Their bioassay program selection process is described in "NIOSH Evaluation of the Internal and External Monitoring Programs at the Lawrence Berkeley National Laboratory".

Regarding the 1985 DOE audit, LBNL responded to the findings in the 1985 audit (Krebs 1985). The audit recommendations and LBNL responses are presented below:

Recommendation 1: Laboratory management assess the effectiveness of the methods used within each Division to identify and correct existing or potential safety problems. Corrective measures are to be identified, as appropriate, and remedial actions taken.

LBL Response: A copy of the appraisal report has been forwarded to all Division Heads for their information, with a request that they review the procedures within their own Division and report their findings to the Director's Office by 1st September 1985.

In addition, the Safety Review Committee has agreed to carry out a review of management implementation of Laboratory health and safety policies throughout the Laboratory and report

back to the Director, also by 1st September. It is expected that this review report will make recommendations to the Director.

Recommendation 2: An overall radiation safety training plan be written and implemented by the Laboratory.

LBL Response: The Radiation Physics Group of the Environmental Health and Safety Department are currently engaged in writing an overall radiation safety training plan. This is being done in conjunction with the Program Divisions and is expected to be completed by the 31st of May.

Major sections of this plan exist and include an Introductory Radiological Protection course; X-Ray Safety Training; Radiological Protection in Chemistry Laboratories. In addition, the Laboratory has introduced Supervisors' Training in Safety which deals in general with radiological problems; the EH&S Department has trained several technicians for certification by the National Registry of Radiation Protection Technologists.

More than 30 scientists and technicians have been trained in radioisotope laboratory radiation safety during the first four months of 1985. Radiological Safety Training for members of the Bevalac crew took place during April 1985.

The overall radiation safety training plan and a schedule for implementation will be prepared by 1st September 1985.

Recommendation 3: The Laboratory review, in approximately six months, the changes to the radiation safety program due to the EH&S staffing cuts. The Laboratory provide SAN with an assessment of the effect of the staffing cuts on the level of risk.

LBL Response: During October 1985, the Safety Review Committee will prepare for the Director an assessment of the impact of staffing cuts made in December 1984 on the radiation safety program of the Laboratory. This report will form the basis of a report to DOE/SAN and should be prepared no later than 1st November 1985.

Recommendation 4: Provide a comprehensive internal review system. Such a system should assign responsibilities for performing the reviews and for implementing, tracking, and follow up on corrective actions.

LBL Response: The program adopted by the Safety Review Committee on January 18, 1985, assigns overall responsibility for the conduct of internal reviews to the Safety Review Committee. This responsibility includes establishing and monitoring criteria for the conduct of

audit activities, providing oversight for the process and transmitting audit findings to affected Division Heads. Responsibility for carrying out audit activities and follow-up has been delegated to the SRC subcommittee.

Under this new system, the SRC has completed an internal review of the Laboratory's Personal Dosimetry Program and reviews of the operations of the Building 75 Tritium Facility, and Laboratory procurement of toxic materials are currently under way.

Recommendation 5: Officially adopt and implement a formal ALARA program and give routine feedback to line management on ALARA effectiveness.

LBL Response: The pilot Laboratory ALARA program as described by the SAN appraisal team will be incorporated in the next revision of the Laboratory Health and Safety Manual. This revision will clearly set forth duties and responsibilities for the administration of the Laboratory's ALARA program.

Recommendation 6: Prepare detailed procedures for conducting surveys.

LBL Response: More detailed procedures than those which already exist are being written. Specific information will include instrument to be used, special problems of low energy beta-emitters, frequency and techniques of surveys, bioassay etc.

These Procedures will be incorporated into Publication 3000 at the earliest possible revision.

Recommendation 7: Train Laboratory personnel on survey techniques and include' this training as part of the Radiation Safety Training Plan (See Recommendation 2).

LBL Response: Already covered in our responses to recommendations 2 & 7.

Recommendation 8: EH&S develop a system to distribute incident reports and trend analyses to line managers.

LBL Response: A system to distribute incident reports and trend analyses to line managers has been in place for several years. Use of this system has been extended in recent months.

It can be seen from the above that LBNL took audit findings seriously, and made efforts to address them.

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