Draft White Paper

SC&A REVIEW OF LAWRENCE BERKELEY NATIONAL LABORATORY SITE PROFILE MATRIX ISSUE #2

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ABBREVIATIONS AND ACRONYMS

ABRWH	Advisory Board on Radiation and Worker Health
AEC	Atomic Energy Commission
ALARA	As Low As Reasonably Achievable
DOE	U.S. Department of Energy
EH&S	Environmental Health and Safety
LANL	Los Alamos National Laboratory
LBL	Lawrence Berkeley Laboratory
LBNL	Lawrence Berkeley National Laboratory
LLNL	Lawrence Livermore National Laboratory
MAP	Mixed Activation Product
MDA	Minimum Detectable Activity
MeV	million electron volts
MPBB	Maximum Permissible Body Burden
mrem	millirem
MSD	Medical Services Department
NaI(T1)	Sodium Iodide doped with Thalium
NDA	non-detectable activity
NIOSH	National Institute for Occupational Safety and Health
pCi	picocurie
SC&A	S. Cohen and Associates (SC&A, Inc.)
SEC	Special Exposure Cohort
SRDB	Site Research Database
TBD	technical basis document
WBC	whole-body counting

1.0 BACKGROUND

In its site profile review (SC&A 2010), SC&A questioned whether there is sufficient information on Lawrence Berkeley National Laboratory (LBNL) bioassay monitoring, post-1961, particularly as it pertains to dose reconstruction based on gross alpha, beta, and gamma analyses. Workers were potentially exposed to a diversity of radionuclides at LBNL, and SC&A found the site profile (ORAUT 2007, now ORAUT 2010) to be incomplete in relation to the periods of time and quantities of radionuclides handled in the different areas. SC&A also questioned whether LBNL methods at the time for gross alpha, beta, and gamma analyses would be sufficient to detect certain radionuclides given low process recoveries and relatively high Minimum Detectable Activities (MDAs).

At the February 2012 Work Group meeting, SC&A emphasized that the issue [now Matrix Issue #2 in SC&A's Issues Matrix (SC&A 2012)] pertains to the adequacy and completeness of bioassay data, with MDA being a key question (ABRWH 2012). The National Institute for Occupational Safety and Health (NIOSH) responded by noting that the period of 1961 and earlier is covered under an existing Special Exposure Cohort (SEC) for LBNL. It was further observed that either MDA information in Table 5-4 of Revision 2 of the LBNL Site Profile or specific MDA information provided in the worker's dosimetry records has been sufficient to assess all current claims. NIOSH noted that use of gross monitoring results, though they may result in higher MDAs, still allows the dose reconstructor to provide a bounding/claimant-favorable exposure analysis. NIOSH also noted that additional information on radionuclide-specific MDAs can always be added to Table 5-4 of the technical basis document (TBD) (NIOSH 2012).

At the Work Group's request, SC&A agreed to review exposure potential from internal emitters at LBNL for the post-1961 period, and determine their significance and whether bioassay monitoring was complete and adequate for that time period. SC&A proposed to sample 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 information would then be made available to NIOSH, in advance, for its use in responding to Matrix Issue #1 (regarding historic documentation of operations and sources of radiological exposures).

In terms of approach, 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. Apparent gaps and differing assessments of bioassay program adequacy are highlighted in this review, with an emphasis on what programmatic or dosimetric shortcomings would have the most significant influence on dose reconstruction.

In terms of SC&A's review under site profile Matrix Issue #2, two central questions surface 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 the former, SC&A questions how LBNL could have adequately monitored for 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. This limitation has been acknowledged during SEC proceedings by NIOSH for another University of California laboratory, Los Alamos National Laboratory (LANL), for which an SEC has already been established in 1975 (due to the lack of WBC capability for detecting MAPs, mixed fission products, and exotic radionuclides) and for which additional years are being considered, based on difficulty bounding short-lived gaseous emissions using limited WBC data.

For the latter question, NIOSH has accepted the inauguration of the LBNL bioassay program in 1961 as the threshold of a comprehensive and reliable database 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.

Both of these questions are discussed below in more detail as issues for Work Group consideration.

2.0 ISSUES FOR WORK GROUP CONSIDERATION

2.1 Exposure Potential for which Adequate Bioassay Coverage May Be Lacking

Table 1 identifies source terms present at LBNL with an identified exposure potential, based on a review of available site monitoring records, but not addressed specifically in the TBD for their ability to be monitored with sufficient accuracy.

Table 1. Source Terms Not Adequately Addressed in TBD having Exposure Potential

Radionuclides	LBNL Building No.	Reference	Comment
Mixed Activation Products (MAPs) (includes C-11, N-13, O-15, Ar-41, Be-7, and others)	Bldgs. 6, 9, 10, 80, 51, 55, 56, 70, 70A, 71, 74, 88	ORAUT-TKBS-0049 (LBNL Site Profile) (ORAUT 2010)	MAPs present for all high energy accelerators as potential exposure source term. Primarily short-lived beta emitters.
Mo- 93 **Nb- 92,93** **Zr- 89, 93, 95**	Bldg 70A, rm 2217	Grill 1966	Source-terms not included in TBD.
Br-82	Bldg 70, rm 133	Low-Beer 1962	NDA, but not clear on MDA; not included in TBD.
Ho-160m, 161 **Nd-149**	Bldg 70, rm 143	Low-Beer 1962	NDA, but not clear on MDA; not included in TBD.
Eu-152, 154	Bldg 70	Low-Beer 1962	NDA, but not clear on MDA; not included in TBD.
Cm-244		LBL 1969	In vivo MDA for Cm-244 2-10x MPBB (1969)…missed dose?
W-181	88” cyclotron	LBL 1969 LBL 1973b	In vivo detection of exposure source w/ unknown energy signature (1969); thought to be W-181 (1973)

Table 1. Source Terms Not Adequately Addressed in TBD having Exposure Potential

Radionuclides	LBNL Building No.	Reference	Comment
S-35		Low-Beer 1963	Only fecal sampling showed activity (1 positive) (1962)
Rh-106	Bldg 70, rm 218	Thaxter 1960	Some Rh compounds gaseous – counting/collection efficiency unknown (1960)

For internal dose assessments, LBNL relied upon in vitro (urinalyses) bioassays for selected employees identified by the Safety Services Department (later Environmental Health Sciences) based on input from onsite radiological monitors, which also identified the radionuclides to which individuals had the potential to be exposed. In the 1960s, there were about 100+ individuals providing routine urinalysis samples at the laboratory, increasing to 170 by 1988 (DOE 1987). A smaller subset of workers was given routine WBCs [employing the Donner WBC using a NaI(Tl) crystal calibrated for a gamma spectrum of 0 to 2.0 MeV], e.g., 28 in 1961, 39 in 1966–1967, and 38 in 1972–1973 (Low-Beer 1962; LBL 1968; LBL 1973a). Most results were non-detectable (NDA), although some longer-lived radionuclides were identified: Y-90, Zn-65, Cs-134, Tc-99m, and Cr-51.

It was recognized early that in vitro bioassay provided for "greater sensitivity" when compared with in vivo WBC monitoring for actinide and beta-emitters. While low-level gross beta activity was typically found in most worker urine samples, the high rate of decay indicated that it was not attributable to Sr-90, a fallout constituent (LBL 1968).

A special WBC survey of 85 accelerator workers (selected from the 184-inch cyclotron, 88-inch cyclotron, Bevatron, and Hilac), performed in 1967, had 7 positive determinations of 91 workers evaluated, with Zn-65, Co-56/57, W-185, and Am-241 identified (LBL 1968). However, for urinalyses taken for the same workers at the same time, 76 of 85 (89%) proved positive for gross beta, with activities ranging from 1–42 pCi/24 hour sample. This supports the LBNL observation (cited above) that relatively low-level dose from short-lived beta emitters not attributable to fallout was being missed in the bioassay program. However, at the same time, based on a gross alpha and beta survey by LBNL of residents of the San Francisco bay area, it was concluded by LBNL that "the continuing low levels of beta activity found in a high percentage of [LBNL] personnel included in the routine bioassay program" were also prevalent in the general population and, therefore, "should probably not be regarded as due to industrial [i.e., LBNL] exposure when found in the routine bioassay program" (LBL 1969). 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.

Table 2 further illustrates this issue by comparing the half-life of radionuclides identified by routine WBC monitoring as compared with common MAPs generated by accelerator operations at LBNL. Other than an outlier radionuclide for each list (Be-7 for MAPs and Tc-99m for positive WBCs), it is clear that the shorter-lived MAPs were not detected by WBC at LBNL in the post-1961 years and made up an indeterminate amount of the "non-detectables" cited for

workers who were routinely counted. As was found at both LANL and Lawrence Livermore National Laboratory (LLNL), while MAPs are relatively short-lived, their copious production in high-energy accelerators can provide a constant exposure source term during operations. This source term is difficult to monitor and estimate because of the nature of the gaseous emissions (neither vapor nor particulate) and the relatively short half-lives involved. Furthermore, the magnitude of this exposure will obviously be a function of accelerator type and energy, operating time scales (i.e., continuous or intermittent), worker occupancy, location and height of onsite emission points, and degree of building ventilation and filtration. For LANL, NIOSH has tried to bound MAP doses by quantifying ratios of released source terms (e.g., Ar-41, O-15, N-13) to Be-7, a longer-lived MAP released with the other gases. This has proven problematic, given the need to ascertain what constitutes a "representative" set of MAP emission values, because they were heavily influenced by factors such as holdup time and facility origin.

Table 2. Comparison of MAPs Generated vs. WBC Monitored

Radionuclide	Type of Decay	Half-Life
Mixed Activation Products:		
C-11	Beta	20 minutes
N-13	Beta	10 minutes
O-15	Beta	123 seconds
Ar-41	Beta	1.8 hours
Be-7	Gamma	53 days
LBNL positive WBC (1962–1973):		
Y-90	Beta	64 hours
Zn-65	Beta	245 days
Cs-134	Beta	1.05 years
Tc-99m	Gamma	6 hours
Cr-51	Gamma	27.5 days
W-185	Beta	75 days
Am-241	Alpha	458 years

Source: Low-Beer 1962; LBL 1968; LBL 1973a; ORAUT 2010.

It should be clarified that this is not to say that the Argonne-type NaI(Tl) detector did not have the capability to discriminate and detect specific MAPs at some activity level. As with LANL and LLNL, however, no monitoring regime existed to do so on a routine basis, because they were not considered a significant enough routine exposure source for workers to warrant sufficiently frequent sampling (or modeling estimations) to be included in the personnel dose records.

Adequacy of Gross Alpha Urinalysis Procedure

A number of LBNL annual bioassay program summaries noted that individuals whose urine samples had been found to be negative were found to have positive fecal sample results (e.g., LBL 1968) and for positive urinalyses, often higher activity levels (LBL 1973b; LBL 1969). These individuals were also whole-body counted with negative results. LBNL concluded at the time (1968) that "in cases of exposure to very small amounts of alpha activity, analysis of a fecal sample is a necessary procedure for effectively ruling out the presence of alpha contamination" (LBL 1968). However, from review of bioassay monitoring records, it is clear that the laboratory continued to rely predominantly on gross alpha urinalyses to confirm exposures to alpha emitters despite this repeated finding. It is clear from these backup fecal results that many

of the negative gross alpha results may have been actually positive for alpha emitters leading to missed dose of an unknown amount.

Preliminary Conclusion:

While there are some MDA and source term-related discrepancies (as listed in Table 1), the key concern stemming from SC&A's review is LBNL's historic capability to monitor for MAPs given its reliance on gross in vivo and in vitro methods, coupled with a lengthy (monthly, yearly) sampling periodicity, that would have mitigated against routine detection of most short-lived MAPs. While the WBC had the capability to discriminate some activation products, its MDA for the weaker beta emitters was apparently too high to ascertain the chronic low-level gross beta uptakes that were being detected in worker urinalyses. In this case, this circumstance parallels that of LANL and LLNL, where concerns have been raised regarding dose reconstructing MAPs when routine WBCs were lacking or did not adequately characterize this source term.

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). The LBNL references cited (LBL 1968) above substantiate this concern through actual observations at the time, where fecal analysis confirmed uptakes that urinalyses and WBCs missed. 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.

2.2 Programmatic Issues Affecting Adequacy of Bioassay

2.2.1 Compliance with LBNL Bioassay Submission Requirements

LBNL has had a history of bioassay compliance problems dating back to the inception of the program. An audit by DOE (DOE 1987) found that while 170 employees working with radioisotopes in 1986 were required to participate in the bioassay program, 24% (or 41 workers) were non-compliant in the submittal of samples for analyses. It was noted in the audit that some of the non-compliant individuals continued to work with radioisotopes without action on the part of LBNL management. It also noted that 10 bioassayed individuals had positive bioassay results for CY1986, with the highest effective dose equivalent being 200 mrem.

Submission of required bioassay samples was a recurring problem at LBNL. A 1962 Bioassay Program report (Low-Beer 1963) indicates that at the end of the year (1962), 24 persons were delinquent in submitting samples, and 7 had not submitted any samples prior to the end of the reporting year. This led to at least one management directive (Howe 1963) warning staff to submit their samples or face disciplinary action. A Bioassay Program report for 1968–1969

(LBL 1969) indicates a compliance rate of 84.2% for 139 employees enrolled in the program for that period (i.e., 22 or 16% of workers were not compliant). For 1971–1972, the bioassay compliance rate was reported as 48%, with less than half of the 139 employees enrolled in the program submitting samples as required. For 1972–1973, the compliance rate for bioassay sample submissions was reported (LBL 1973a) as 72% for 117 workers. It was also noted that laboratory management found it "impossible in a number of instances" to obtain a second confirmatory sample in those instances where a positive alpha determination had been made (LBL 1973b).

Preliminary Conclusion:

It is not clear from available records whether delinquent bioassay samples were eventually collected (based on SC&A's site profile review, some were at least a year overdue) or not, but given the relatively high percentages of noncompliance and the apparent lack of management accountability to enforcing bioassay requirements (as late as 1986), NIOSH should review the adequacy and completeness of bioassay records from this perspective (per Matrix Issue #4) and determine the significance of missing bioassays. From the positive results cited in all of these program reports, a clear exposure potential exists for these non-compliant workers, and the question of historic non-compliance is not addressed in the TBD.

2.2.2 Program Reliability: Selection of Personnel and Radionuclides

For adequate bioassay results, it is critical that personnel are included in the bioassay program based on their work and are bioassayed for the radionuclides to which they are potentially exposed. The TBD concludes that after 1961:

> *LBNL bioassay records show that the selection of personnel for bioassay and the radionuclides for analysis have been based on the work performed by the individual. Selection of employees to be included in the bioassay program was typically made by the Laboratory's Safety Services Department through its staff of monitors. The monitors were directly aware of the radionuclides used throughout the Laboratory and were therefore best qualified to select employees at risk for potential internal exposure.* (ORAUT 2010)

However, LBNL reports and memoranda from the 1960s through the 1990s 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. At LBNL, the bioassay program was administered by the Medical Services Department (MSD), while the personnel dosimetry (external) program was administered by the Environmental Health and Safety (EH&S) Department. Given this organizational split, it was critical for EH&S to work closely with MSD to ensure that the latter had a complete listing of employees who warranted bioassay screening due to their work-related exposure potential for certain radionuclides. Otherwise, MSD, having little operational perspective, would be unable to maintain an accurate listing of who should be bioassayed for what radionuclides.

Program reviews raised repeated concerns over this organizational dichotomy and its adequate implementation of the selection process. A 1985 review of the bioassay program by DOE (DOE 1985a) found, in part:

- *…no formal mechanism for the selection of employees that are required to be on the program.*
- *…no written criteria stating which tests are to be performed of the different classes of employees on the program.*
- That… *the list of employees currently on the program had a number of inconsistencies and may not be up-to-date regarding which radionuclides are currently being used by the individual employees.*
- That… *there are LBL employees who work on Campus with unsealed radionuclides. It is not clear whether it has ever been determined if bioassay monitoring should be done on these individuals and if so, where their radiation records would be maintained.*
- *No formal mechanism currently exists for including supervisors in the loop for determining list of employees to be covered or in follow-up compliance procedure.*
- *Communications between EH&S and Bioassay* [MSD] *appears to rely primarily on informal contacts with no scheduled meetings between medical and EH&S re: Bioassay coverage.*

For corrective actions, DOE recommended that LBNL require MSD to take a stronger management role in the bioassay program, and that EH&S verify the selection and follow-up process for those in the bioassay program.

An April 1985, Functional Appraisal of the radiation safety program by DOE (DOE 1985b) found many contamination incidents were only discovered after the internal exposure was detected through the routine bioassay program or by environmental sampling. While the focus of the review was the inadequacy of the ALARA program at LBNL, the appraisal noted that a 20% staffing reduction had taken place for radiation monitors, which may have had a direct influence on the lack of response to contamination incidents. It may have also impacted the bioassay program by mitigating against updated personnel selection and follow-up.

With respect to bioassay sample compliance, a 1987 DOE review (DOE 1987) found that, "Laboratory management is not enforcing the LBL policy to submit bioassay samples." The report further found that "some of these individuals are still working with radionuclides without participating in the bioassay program."

This persistent lack of employee responsiveness and lack of accountable management systems to ensure a comprehensive bioassay program is not surprising, given the apparent laboratory management culture and thinking regarding its safety functions in the earlier years, including the 1960s–1980s. As illustrated in a memorandum (Howe 1966), some laboratory managers believed a more ad hoc, decentralized system was more conducive to a research-oriented

institution than one that entailed more formal management direction and support, as advocated by the Atomic Energy Commission (AEC), and later by DOE oversight findings. This internal memo also defends the lack of more formal communications between EH&S and MSD regarding the bioassay program and its selection process, and suggests that recommendations by the AEC were misguided and originated from its "rather bureaucratic thinking." It does not appear that there was a commitment to respond to AEC recommendations in a meaningful way and strengthen institutional accountability for more effective bioassay program implementation.

Preliminary Conclusion:

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.

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