
Draft

**ADVISORY BOARD
ON RADIATION AND WORKER HEALTH**

National Institute for Occupational Safety and Health

**SC&A's Review of NIOSH's Evaluation Report for Los Alamos National
Laboratory Special Exposure Cohort Petition SEC-00109:**

***Preliminary Issues – Availability of Bioassay Records and
Adequacy of In-vivo Dosimetry***

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 2 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

<p>S. COHEN & ASSOCIATES:</p> <p><i>Technical Support for the Advisory Board on Radiation & Worker Health Review of NIOSH Dose Reconstruction Program</i></p>	Document No. SCA-TR-SEC2010-0004
	Effective Date: Draft – April 8, 2010
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<p>SC&A’s Review of NIOSH’s Evaluation Report for Los Alamos National Laboratory Special Exposure Cohort Petition SEC-00109: Preliminary Issues – Availability of Bioassay Records and Adequacy of In-vivo Dosimetry</p>	Page 2 of 53
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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 3 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

TABLE OF CONTENTS

Abbreviations and Acronyms	5
1.0 Introduction and Summary	8
2.0 Preliminary Issues	11
2.1 LANL Capability to Monitor and Measure Mixed Fission and Activation Products by 1976.....	11
2.2 NIOSH’s Assumption Regarding Intake Values for Exotic Radionuclides at LANL	14
2.3 Completeness and Reliability of LANL In-Vitro and In-Vivo Data, and Adequacy of NIOSH Coworker Model (ORAUT-OTIB-0062 and ORAUT- OTIB-0063), Post-1975	20
2.4 Feasibility of Dose Estimation for Neutron Exposure at LANL, Post-1975	24
2.5 Feasibility of Dose Estimation for Radiological Exposure Sources at LAMPF/LANSCE.....	30
2.6 Radiation Exposure to Special Tritium Compounds	37
2.7 Unmonitored Exposure of Support Service Personnel	42
3.0 References.....	47
Attachment A: Sources and Time Periods of LANL Bioassay Repository (ORAUT-OTIB- 0063)	51
Attachment B: Timeline of Bioassay Records in LANL Bioassay Repository (ORAUT-OTIB-0063)	52
Attachment C: Timeline of LANL Bioassay Records and NIOSH Evaluation Report.....	53

NOTICE: This report has been reviewed for Privacy Act information and has been cleared for distribution. However, this report is pre-decisional and has not been reviewed by the Advisory Board on Radiation and Worker Health for factual accuracy or applicability within the requirements of 42 CFR 82.

LIST OF TABLES

Table 1.	NCF vs. LANL Facility	28
Table 2.	Approximate NCFs and Dose Fractions for Neutron Sources.....	28
Table 3.	Comparison of Measured Activity to Public Release Limits.....	36
Table 4.	Years Bioassayed Compared to Years Worked and Position Title.....	44
Table 5.	Bioassay Pattern of Six Security-related Personnel from Previous 30 Cases.....	45

LIST OF FIGURES

Figure 1.	Los Alamos Meson Physics Facility East End (Experimental Area)	34
Figure 2.	West and East Evaporation Ponds	34
Figure 3.	Number of Bioassays per Year for Six Security Workers	46

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 5 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

ABBREVIATIONS AND ACRONYMS

Advisory Board or Board	Advisory Board on Radiation and Worker Health
ALI	Allowable Limits on Intake
ALARA	As Low As Reasonably Achievable
BEST	Bioassay Enrollment Scheduling and Tracking (System)
CAM	Continuous Air Monitor
CATI	Computer-Assisted Telephone Interview
cm	Centimeter
CNC	Chemistry and Nuclear Chemistry
DAC	Derived Air Concentration
DOE	U.S. Department of Energy
DOELAP	Department of Energy Laboratory Accreditation Program
dpm	Disintegrations per minute
ER	Evaluation Report
eV	Electron volt
GeLi	Germanium-Lithium (detector)
HPGe	High Purity Germanium (detector)
HPTL	High Pressure Tritium Laboratory
HRL	Health Research Laboratory
HT or T ₂	Tritiated gaseous hydrogen
HTO	Tritiated water or tritiated water vapor
ICRP	International Commission on Radiological Protection
IRF	Intake retention factor
IVML	In-Vivo Measurements Laboratory
keV	Kilo electron volt
LAHDRA	Los Alamos Historical Document Retrieval and Assessment
LAMPF	Los Alamos Meson Physics Facility
LANL	Los Alamos National Laboratory
LANSCE	Los Alamos Neutron Science Center
LET	Linear Energy Transfer
LOD	Limit of Detection

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 6 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

MAP	Mixed Activation Products
MDA	Minimum Detectable Activity
MeV	Million electron volts
MFP	Mixed Fission Product
mrem	millirem
MT	Metal Tritide
NCF	Neutron Correction Factor
nCi	Nanocurie
NIOSH	National Institute for Occupational Safety and Health
n/p	Neutron to Photon Ratio
NTA	Neutron Track A film
NTS	Nevada Test Site
OBT	Organically Bound Tritium
ORAUT	Oak Ridge Associated Universities Team
OTIB	ORAUT Technical Information Bulletin
pCi	Picocuries
pdf	Portable Document Format
PPE	Personnel Protective Equipment
QF	Quality Factor
R&D	Research and Development
RCA	Radiation Controlled Area
RCRA	Resource Conservation and Recovery Act
RFP	Rocky Flats Plant
RIR	Radiological Incident Reports
RWP	Radiological Work Permit
SC&A	S. Cohen and Associates
SEC	Special Exposure Cohort
SMT	Stable Metal Tritide
SRDB	Site Research Database
STC	Special Tritium Compound
SWP	Special Work Permit
TA	Technical Area

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 7 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

TBD	Technical Basis Document
TED	Track-Etch Dosimeter
TIB	Technical Information Bulletin
TLD	Thermoluminescent Dosimeter
TSFF	Tritium Science and Fabrication Facility
TSTA	Tritium System Test Assembly (Facility)
TUPo	Tritium-Uranium-Plutonium
V&V	Verify and Validate
WBC	Whole-Body Count
WETF	Weapons Engineering Tritium Facility

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 8 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

1.0 INTRODUCTION AND SUMMARY

At the meeting of the Advisory Board on Radiation and Worker Health (Advisory Board or Board) held February 17–19, 2009, in Albuquerque, New Mexico, the Board voted to recommend a Special Exposure Cohort (SEC) class for all:

...employees of the DOE, its predecessor agencies, or DOE contractors or subcontractors who were monitored or should have been monitored for radiological exposures while working in operational Technical Areas with a history of radioactive material use at the Los Alamos National Laboratory (LANL) for a number of work days aggregating at least 250 work days during the period from March 15, 1943 through December 31, 1975...

The Advisory Board subsequently authorized SC&A to perform a focused review of the LANL evaluation report for petition SEC-00109, for the period January 1, 1976, through December 31, 2005. The petitioner class was identified as “Service Support Workers (which includes, but is not limited to, security guards, firefighters, laborers, custodians, carpenters, plumbers, electricians, pipefitters, sheet metal workers, ironworkers, welders, maintenance workers, truck drivers, delivery persons, rad technicians, and area work coordinators) who worked in any operational Technical Areas with a history of radioactive material use” at LANL for the petition period. This report represents a preliminary identification of potential LANL SEC issues with regard to this matter, based on a review of the National Institute for Occupation Safety and Health’s (NIOSH’s) SEC-00109 Evaluation Report (ER) (NIOSH 2009), applicable LANL site profile documents, and onsite interviews and data retrieval. SC&A focused investigations related to the LANL SEC petition and ERs are continuing, as described in this preliminary review for the Advisory Board.

As noted in the Site Description Technical Basis Document (ORAUT 2004):

The diversity of [Los Alamos National Laboratory] operations may be second-to-none when compared to other sites within the DOE complex. With a few exceptions, most man-made or natural radionuclides known to exist have, at one time or another, been present and/or processed at LANL.

As NIOSH points out in its site profile, these operations ranged from various research reactors to critical assembly facilities to nuclear materials research laboratories to the Los Alamos Neutron Source Center (LANSCE). These operations generated a diversity of radionuclide sources, including reactor fission products, short-lived accelerator radionuclides, and specific “exotic” isotopic sources from special research projects, such as Np-237.

However, as noted in SC&A’s 2006 review of the LANL Site Profile (SC&A 2006):

The SC&A review finds that inadequate consideration was given to potential exposure and missed dose from radionuclides other than the “well documented” ones cited in the TBD (e.g., plutonium, polonium, tritium, etc.).

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 9 of 53
----------------------------------	---------------------------	-------------------------------------	---------------------

In this SEC-related review, SC&A similarly finds that, while NIOSH concludes that it can perform dose reconstruction for the Mixed Fission Products (MFPs)/MAPs and so-called exotic radionuclides with sufficient accuracy, based on the advent of new measurement technology at LANL (in-vivo bioassay detectors installed in the 1970s), the actual application of that technology post-1975 to routinely monitor for MFPs/Mixed Activation Products (MAPs) and exotic radionuclides is not adequately documented or substantiated. In fact, in the ER, NIOSH proposes the use of surrogate radionuclides, i.e., the primary ones (Pu-238, Pu-239, Cs-137, and uranium), for which there may be sufficient measurement data, as a means to derive daily intake rates for the exotics, because “specific data for such measurements are very sparse and generally unavailable” for them. The basis for NIOSH’s proposed method is the assumed similarity of the historical operational handling of these primary and exotic radionuclides at the laboratory, based on a search of the LANL operational literature. NIOSH also proposes to use ORAUT-OTIB-0054 (ORAUT 2007a) for assigning MFP/MAP unmonitored intakes at LANL; however, OTIB-0054 only provides a means to assign MFP/MAP intakes for certain reactor types, and where the radionuclides are identified and ratios known, and when accelerator-generated MAPs are not included – exposure conditions that SC&A finds are not substantiated in the ER, or match those at LANL.

The use of the **site-wide averaged** surrogate data in place of **operation-specific** data for exotic radionuclides raises a series of SEC-relevant questions regarding overall data accuracy, as well as data adequacy and completeness as it pertains to the coworker model. This is particularly relevant to the support service workers at LANL, including guards and firefighters, because they were not often bioassayed, despite having site-wide access and broad potential exposure to exotic radionuclides, among other radioactive sources.

A preliminary review of this premise, i.e., that the availability of monitoring capability alone would have been sufficient to enable detection and determination of uptakes of exotics, is not necessarily supported by field evidence. For example, an internal audit of the internal dosimetry program by the U.S. Department of Energy (DOE) as late as 2001 found that thorium-232 and the short-lived radionuclides generated at the Los Alamos Neutron Science Center (LANSCC), while required procedurally for routine internal dosimetry evaluation, were not included in the in-vivo program library at that time (DOE 2001). Interviews with LANL internal dosimetrists indicated that, while they are uncertain about the degree of attention afforded the exotic radionuclides in the early part of the program (because exposures were rare), they believe that the system was capable of detecting them. However, again, no documentation was found or offered that would corroborate a LANL practice in the 1970s and 1980s to “look for” these exotics beyond an “event driven” circumstance, where they would be targeted due to suspected elevated exposure potential. And, again, few data points apparently exist to demonstrate that such attention was being given to them.

Other questions, which either are not addressed or not sufficiently addressed in the ER, include the feasibility of dose estimation for neutron exposure, given the uncertainties involved in the necessary correction factors for Neutron Track A (NTA) film threshold/fading and thermoluminescent (TLD) energy dependency, as well as the lack of treatment given dose estimation for special tritium compounds (STCs). Based on interview results, SC&A also gave attention to a special circumstance where support service workers may have been inadvertently

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 10 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

exposed to both external and internal radiation, due to their work location at the LANSCE accelerator facility. While determining that existing TLD badges would likely have been sufficient for potential external radiation near the beam stop area in question, questions remain regarding the extent to which an internal exposure source due to airborne tritium may have existed, and whether this exposure is dose reconstructible.

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 11 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

2.0 PRELIMINARY ISSUES

2.1 LANL CAPABILITY TO MONITOR AND MEASURE MIXED FISSION AND ACTIVATION PRODUCTS BY 1976

NIOSH acknowledges in its ER that, in considering Petition 00109, certain issues remained unresolved from the previous petition evaluation (for which an SEC was granted), “in particular the assessment of dose from mixed fission products.” NIOSH also observed that “...some radionuclide maximum intakes possibly could be inferred from the chest counting data. However, at the time of this report, an analysis of this technique has not been performed.” NIOSH concludes that by 1976, **in-vivo** counting methods were well-established and available for bounding intakes of MFP and MAP. On page 21 of the ER, NIOSH defines MFP and MAP as:

Mixed fission and products (MFP) and mixed activation products (MAP) are generally beta and/or gamma-emitters. MFP is common in reactor areas. MFP includes Cs-137, Sr-90, radioactive noble gases, and others. MAP is common in accelerator and reactor areas. MAP includes C-11, N-13, O-15, Ar-41, Be-7, Na-22, Na-24, Co-58, Co-57, Mn-54, Mn-52, V-48, and others.

ORAUT-OTIB-0054 (ORAUT 2007a) discusses MFP/MAP ratios and the calculation of intakes based on a single nuclide (e.g., Cs-137 coworker data). For unmonitored workers, Cs-137 coworker data are available in the draft coworker study ORAUT-OTIB-0062 (ORAUT 2007b). For bounding intakes to unmonitored workers, Cs-137 coworker data in the draft coworker study (ORAUT-OTIB-0062) may be used. NIOSH lists radionuclides with exposure potential at LANL in the ER, and provides their dose reconstruction approach to the monitoring technology involved and data available.

In 1970, an **in-vivo** counter capable of measuring four separate regions of the body began operation (Vasilik and Aikin 1983). Twin Phoswich (CsI and NaI) detectors were placed over the lungs. The two layers of the detector were capable of simultaneously, yet separately, monitoring chest burdens for 10–250-keV photons (NaI), and 250–2,000-keV photons (CsI) for a qualitative assessment of a variety of fission and activation radionuclides. A planar High Purity Germanium (HPGe) detector monitored the region between 10 keV and 250 keV with excellent energy resolution, and could be positioned over the liver or thyroid as needed. Finally, an HPGe (previously a GeLi) detector was positioned under the prone subject. This detector was primarily for whole-body assessment and has good photon energy resolution in the range of 10–2,000 keV. This system could both identify radionuclides and quantify the body burden. Plutonium-239 and Am-241 were a routine part of the **in-vivo** analysis library for all individuals receiving lung counts. In recent years, U-235 and Th-234 (as U-238) were added to the routine library. Other nuclides would be identified at their corresponding minimum detectable activity (MDA) levels if they appeared in the gamma spectrum. This typically meant that measured counts had a 95% or greater chance of being a true activity above the MDA or critical level for that region of the energy spectrum. A detailed description of how the in-vivo data was statistically evaluated is provided primarily in two LANL health physics reports (Vasilik and Aikin 1983, and Vasilik

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 12 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

et al. 1984). MDAs for whole-body counting (WBC) and lung counting by historical period are provided in Table 5-18 in ORAUT-TKBS-0010-5 (ORAUT 2009b).

SC&A Preliminary Assessment:

SC&A found the details of what capabilities were in place and what procedures were adhered to for the detection, recognition, and recording of MFP/MAP from in-vivo bioassays at LANL during the period 1976–1990s to be somewhat subjective, depending on the source of the information. Definitive documentation of MFP/MAP detection and recording as a function of time is not prevalent. Both ORAUT-TKBS-0010-5 (ORAUT 2009b) and the NIOSH’s ER (NIOSH 2009) use general terms concerning MFP/MAP capabilities and practices during this time period; there are no hard and fast rules concerning the identification and recording of these radionuclides as a function of time (such as a table listing the important in-vivo counting requirements/procedures chronologically, or referenced standard operating procedures). However, Table 5-18, page 50, of ORAUT-TKBS-0010-5 does provide MDA values for some MFP/MAP for 1971–1984, and for numerous MFP/MAP for 1985–1998, and also for 1999–present. This indicates that bioassays could have been evaluated for these particular radionuclides during these periods.

SC&A interviewed LANL staff and other experts to assist in determining further details concerning this issue. A brief summary of the results of these interviews is provided below.

- Starting in 1969, MPF/MAP radionuclides were added to the in-vivo program for quantitative analyses.
- It is fairly well recognized that most DOE facilities, such as LANL, Rocky Flats Plant (RFP), etc., had the capability to detect, identify, and quantify MFP/MAP in workers beginning in the early 1970s. However, it is not as certain (or as well documented) that it was standard practice to actually analyze and record the activity from these radionuclides in the worker’s file.
- MFP/MAP activities appear to have been investigated in certain situations, but it has not been documented that it was performed on a routine basis at LANL.
- Prior to 1998, LANL primarily relied upon the Phoswich detectors for in-vivo (whole-body or lung counting) measurements. An in-vivo count spectrum was typically not analyzed for fission or activation product radionuclides, unless a peak associated with a certain nuclide was visible in the spectrum, or LANL knew or suspected that an exposure had occurred. When that peak was identified, the nuclide was added to the radionuclide library, and the spectrum was converted to activity and reported in the record. Identification of a peak could be subjective at times and not directly correlated to MDA or critical levels, especially with the broad peaks that appeared in the photon spectra, because of the low resolution of these scintillation-type detectors.
- Phoswich detectors were unable to resolve peaks for exotic and MFP or MAP radionuclides, particularly those that emitted low-energy photons.
- To overcome the detector’s inability to resolve peaks, LANL set up regions of interest for the photon spectra, and used control groups of non-nuclear workers to estimate body and

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 13 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

room background contributions, and to statistically determine net counts above background and identify peaks of interest.

- After germanium detectors became available, if a known or suspected exposure had occurred, then measurements were repeated with high-resolution germanium detectors to verify and identify the presence of a radionuclide if the peak was determined not to be one of the primary radionuclides. If detected, the radionuclide was added to the analysis library and confirmed as a positive identification that needed a dose assessment. In most of the cases, in-vitro bioassay samples were also collected.
- There is a higher likelihood that peaks could have been missed when the germanium detectors were not in operation, or had not yet been installed (i.e., before 1998).
- It is expected that the accuracy of peak identification, including the recognition of counts above background in the region of interest, increased as a function of photon energy, and therefore, this decreased the probability of false negative results for fission and activation products that emitted higher energy photons.
- Whole-body or organ counts were repeated several times if necessary when a peak was identified, and to confirm if the counts were above the specific MDA or decision value. The worker would also be requested to submit urine samples if health physics determined or suspected that an exposure had occurred.
- For WBCs or lung counts reported in the LANL Bioassay Repository, it is not always reasonable to assume that a worker had a real exposure or was being monitored for all radionuclides simply because an MDA was determined and listed in the record. These must be addressed on an individual case basis.
- A programmatic assessment of the internal dosimetry program by DOE in 2001 found that thorium-232 and the short-lived MAP radionuclides generated at LANSCE, while required for routine internal dosimetry evaluation, were not included in the in-vivo program library. The absence of this routine monitoring capability as late as 2001 brings into question the ability of the LANL program to detect these and other exotics on a routine basis as a matter of practice (vs. technical capability). As DOE noted in its finding, “Without this information, the in-vivo laboratory cannot identify monitoring strategies or ensure adequate energy calibrations;” and that “interviews with the in-vivo staff indicated that they were not aware of the need for this capability.”
- LANL workers were selected for lung and/or WBC based on the following factors:
 - Type and chemical and/or physical form of the radionuclide(s)
 - Level of containment vs. relative hazard
 - Use of personal protective equipment (PPE)
 - Area where radioactive material was processed
 - Job responsibilities
- Generally, all workers that handled plutonium, americium, uranium, polonium, tritium, and thorium were on some form of bioassay or WBC program.
- According to NIOSH’s ER, page 55, the findings of the Tiger Team Assessment in 1991 do not significantly affect most dose reconstruction using bioassay data, because LANL primarily relied upon in-vitro bioassays for monitoring the primary radionuclides.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 14 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Summary

SC&A did not find definitive answers to the issue concerning MFP/MAP detection, identification, and recording, especially for the earlier period, 1976–1990s, of the SEC. While it appears that exposures from MFP/MAP were limited, compared to the primary radionuclides, SC&A does not find their **concentrations and/or ratios** to the primary radionuclides to be documented to allow for sufficiently accurate dose reconstruction; nor did SC&A find that the **requirements and procedures** for monitoring for these radionuclides significantly changed in the 1970s with the introduction of in-vivo counters; i.e., the presence of the technology did not ensure that monitoring and recording of the MFP/MAP radionuclides were actually performed on a routine basis. This circumstance is further corroborated by DOE audit findings as late as 2001 that determined that the LANL in-vivo program had not maintained the capability to routinely monitor secondary radionuclides, such as Th-232, and the short half-lived MAPs regularly generated at LANSCE, although that capability was required; apparently, the in-vivo staff was unaware of the need to maintain this capability.

The use of surrogate radionuclides (such as Cs-137, as proposed in the ER) is only useful if the ratio of the surrogate to unmonitored radionuclide is known and remains relatively constant. NIOSH’s ER refers to ORAUT-OTIB-0054 (ORAUT 2007a) as a method to assign MFP/MAP unmonitored intakes; however, OTIB-0054 is based on defined reactor types and known radionuclide ratios, and it does not include MAP produced by accelerators. Neither is OTIB-0054 applicable to radionuclides that have been concentrated and/or removed from the normal reactor fuel cycle, because the ratios are no longer defined. Interviews with LANL internal dosimetrists with knowledge of the in-vivo program suggest that, while they are uncertain about the degree of attention afforded the exotic radionuclides in the early part of the program (because exposures were rare), they believe that the system was capable of detecting them. However, again, no documentation was found or offered that would corroborate an LANL practice in the 1970s and 1980s to “look for” these exotics beyond an “event-driven” circumstance, where they would be targeted due to suspected elevated exposure potential.

2.2 NIOSH’S ASSUMPTION REGARDING INTAKE VALUES FOR EXOTIC RADIONUCLIDES AT LANL

NIOSH asserts in its ER that LANL health physics records indicate that exotic radionuclides were handled, controlled, and monitored in a manner equivalent to that of the primary nuclides (i.e., plutonium, americium). For example, it is indicated that many alpha-emitting exotics were handled in a manner similar to how plutonium was handled—using gloveboxes, monitoring airborne concentrations, using PPE (such as respirators), performing surface contamination surveys, and covering jobs with radiation protection technicians. NIOSH observes that, although the vast majority of the documents it found were associated with the primary radionuclides, “several” documents pertaining to exotics were located. A list of excerpts from these documents was provided in the ER to highlight operational examples of exotics-handling that resembled those for plutonium.

NIOSH goes on to conclude that because it was demonstrated that (1) from these records that exotic radionuclides were handled, controlled, and monitored in a similar manner as the primary

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 15 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

nuclides; (2) applicable guidelines for controlling airborne concentrations and surface contamination levels for all radioactive materials are defined in units of activity (e.g., $\mu\text{Ci}/\text{m}^3$ and $\text{dpm}/100 \text{ cm}^2$); and (3) the guidelines for many of the exotic alpha-emitters were the same as, or more restrictive than, the guidelines established for Pu-238 and Pu-239, then it follows that it is possible, in the absence of specific internal dosimetry data, to bound intakes of many of the exotic alpha-emitting radionuclides using coworker data for Pu-238 and/or Pu-239.

SC&A Preliminary Assessment:

Radionuclides that were not part of the mainstream work at LANL are sometimes referred to as “exotic” radionuclides; these were usually used for specific projects, or were contaminants in the predominant (primary) radionuclide material. According to NIOSH’s ER (NIOSH 2009, page 12), “The term ‘exotics’ is used to include everything other than U-234/235/238, Pu-238/239, tritium, Am-241, and Cs-137. This would include Sr-90, Th-232, Cm-244, Ac-227, Pa-231, Np-237, and others.” And on page 41 of NIOSH’s ER, it states that, “LANL clearly possessed capabilities to conduct bioassay measurements for these exotic radionuclides (LANL 2008); however, specific data for such measurements are very sparse and generally unavailable.”

NIOSH’s ER of January 22, 2009, for this petition rests partly on the premise that exotic radionuclides would have been handled, controlled, and monitored in a similar manner as the primary nuclides (Pu-238, Pu-239, U, and Cs-137). That is to say, unmonitored intakes of exotic radionuclides by workers would be assigned on the basis of monitored intakes of coworkers as follows:

- (1) The daily intake rates for Pu-239 taken from OTIB-0062 (ORAUT 2007b), in units of pCi/d, would be assigned separately for each alpha-emitting exotic radionuclide that would have required similar controls: Ac-227, Pa-231, Np-237, and Th-230.
- (2) The daily intake rates for Pu-238 taken from OTIB-0062, in units of pCi/d, would be assigned for unmonitored potential Cm-244 exposures.
- (3) The daily intake rates for uranium taken from OTIB-0062, in units of pCi/d, would be assigned for unmonitored potential of natural thorium exposures.
- (4) The daily intake rates for Cs-137 taken from OTIB-0062, in units of pCi/d, would be assigned for unmonitored potential exposures of MAPs, MFPs, and SrY-90.

The following section addresses three issues related to NIOSH’s proposed approach for assigning dose from exotic radionuclides:

- (1) **Handling and control** – Does site documentation support NIOSH’s premise that operational and radiological handling, control, and monitoring of exotic radionuclides were equivalent to those for radionuclides that were used in larger quantities at LANL?
- (2) **Use of exotics** – Did LANL continue to use processes that involved exotic radionuclides during the time period of SEC interest, 1976–2005?

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 16 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

- (3) **Assignment of Dose** – Is there sufficient representative data to support the validity of the coworker model, and does it provide a bounding dose estimate for all workers for all radionuclide internal uptakes at LANL for the time period in question?

Handling, Control, and Monitoring of Exotic Nuclides

SC&A’s assessment of NIOSH’s position is based on a review of selected radiological work control documents, searches of the Site Research Database (SRDB), and searches of the Los Alamos Historical Document Retrieval and Assessment (LAHDRA) database. Documents that were of particular interest were those that would give insight into the scale of programs involving exotic radionuclides, and those that either support or contradict NIOSH’s hypothesis that exotic radionuclides were handled in a similar manner to the primary isotopes at LANL, and that the latter could be applied as surrogates in the absence of actual monitoring data. The objective was to review additional sources of operational documentation, particularly available radiological work permits (RWPs), to determine whether they agreed with the limited results of NIOSH’s literature survey.

SC&A reviewed selected special work permits (SWPs) and RWPs; however, SC&A found this information to fall short in producing definitive results. Permits for activities involving radiation and radioactivity were only found going back to about 1983 during a limited review of the SRDB. The permits that were reviewed did not cover the entire 1976 to 2005 time period that Petition SEC-00109 covers.

RWP number 03-54-G-024 (LANL 2003) describes a contamination survey of parts at TA-54 that occurred in 2003; both uranium and thorium are identified in the document, and the PPE assigned appears to have been reasonable.

A number of work permits for radiation work were found where high- and low-toxicity radionuclides were handled at the same location, or where maintenance was required on equipment that was contaminated with co-mingled high- and low-toxicity radionuclides. This does not appear to support a notion that isotopes were generally segregated by toxicity, so that they could be managed properly according to their individual toxicities. RWP #TA-48-03-23, written in September 2003, was developed for the cleanout of hoods at TA-48. It identified an extensive list of nuclides that included depleted uranium, plutonium, curium, neptunium, and others (LANL 2004, pdf. 85). Several other RWPs covering the 2003 time period were reviewed which involved work with materials where uranium and other more toxic transuranics were apparently co-mingled; examples are RWP #TA-48-03-11 (LANL 2004, pdf. 181) and RWP#TA48-03-10 (LANL 2004, pdf. 193). Although review of these documents found evidence of co-mingling, each of the RWPs appeared to be adequate for protecting against the most restrictive radionuclides in the mixture.

RWP number OHP92-694 describes removal of a “thoria duct, stack and fan” at TA-3 in 1992 (LANL 1992, pdf. 33). The activity involved the use of a full face respirator and double anti-Cs. The controls appeared adequate for what may have been anticipated to be a dusty job.

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 17 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

An unnumbered SWP from 1986 was found on pages 56–61 of SRDB document number 51418 (LANL 1986). It was for an excavation refill task at materials disposal area “T.” Analyses of the underlying soils in the excavation had up to 1.9 nCi/gm gross alpha, and the predominant isotope disposed at materials disposal area “T” would have been Am-241. The controls were reasonable and would have been appropriate for Pu-239 contaminated soil and tuff in the same setting.

A series of SWP for activities involving MAP in 1986 at the Omega West Reactor were found on pages 110–113 of SRDB document 51418 (LANL 1986). There was not enough detail in the permits to conclude in retrospect whether the controls were appropriate or not, as no pre-job surveys or follow-up information were found with the permit.

To summarize, several collections of work permits for radiological work were reviewed that covered the time periods 1986, 1992, and 2003. Comparatively few permits involved exotic radionuclides, and no examples were found that contradict NIOSH’s hypothesis that exotic radionuclides were handled in a similar manner as the corresponding primary radionuclides. However, given the relatively narrow range of available RWPs, a definitive conclusion is not reachable.

Recent Use of Exotics

A keyword search of the SRDB did not indicate that there were ongoing programs involving thorium-230, actinium-227, or polonium-210 during the time periods of concern in SEC Petition 00109. In addition, the search did not find indications that significant work involving protactinium-231 occurred beyond the 1970s.

Evidence of use of natural thorium was found from a keyword search of the SRDB. There was a Tiger Team audit finding, number RP.7-1, in 1991, that refers to workers handling gram quantities of dispersible natural thorium powders in the Isotope and Structural Chemistry Group, the Ceramic Science and Technology Group, and the Materials Technology Metallurgy Group at TA-21 and TA-3 Bldg SM-66 (DOE 1991). Gram quantities of thorium powders suggest a research use, rather than operations on an industrial scale. The LANL *Site-Wide Environmental Impact Statement*, issued as final in 1999 (LANL 1999a), references a Thorium Storage Building at the Sigma Complex, which was a Category III nuclear facility due to its inventory of thorium as ingot and as oxide. No other details about the Thorium Storage Building were found.

A document entitled “Standard Operating Procedures for the Handling of Actinide Elements” (LANL 1973) is a statement of practice of organization CNC-4 for handling actinide elements; primarily americium, curium, berkelium, californium, and protactinium. Both the CNC-4 and H-1 organizations approved the document in October 1973. The protective measures specified in the procedure appear to be generally reasonable for handling the isotopes involved; however, the significance of the document is unclear. There is no statement of applicability as to whether it applied lab-wide, to all of the CNC division, or just to CNC-4.

The document (LANL 1973) addresses Cm-244 and “curium,” one of the exotic isotopes listed in the SEC Petition. It describes issues with handling curium in quantities of “many milligrams

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 18 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

to gram level.” It also discusses considerations for storing “grams of curium,” although it is possible that very little use of Cm-244 inside or outside of the CNC-4 organization occurred during the 1970s. The memo does not mention specific projects, or indicate whether there were ongoing activities involving curium in the 1970s.

Protactinium-231 is mentioned as “also handled” in 1973, but no other information is given in the document (LANL 1973). The ER indicates production of Pa-231 at Mound was very limited in the late 1970s and ceased in 1979. This, together with the general lack of references to Pa-231 in the SRDB, suggests that there were no activities involving use of large amounts of Pa-231 during the 1976–2005 timeframe.

In summary, a series of queries of the SRDB and review of the resulting documents produced few indications of operations at LANL that involved exotic alpha emitters for the time period 1976–2005. No evidence was found in the document search that clearly contradicts the assumptions made in the ER concerning the extent of usage of exotic isotopes at LANL.

Assignment of Dose from Exotic Radionuclides

As a result of the questions surrounding the completeness of bioassay data for exotic radionuclides, NIOSH proposes to use the intakes of the primary radionuclides to bound the intake of the exotic radionuclides for workers who had the potential for the intake of these radionuclides, as previously outlined above and as stated in Section 7 of the ER, using surrogate radionuclides that had similar radiological properties and health physics controls, e.g., use of uranium coworker intake to bound the intake of thorium.

Some of the questions concerning this approach are as follows:

- How will the dose reconstructor know that the claimant had a potential intake of radionuclides other than the primary radionuclides? While useful, the CATI report is not a definitive document concerning details of the potential intake of specific radioactive materials, and neither do the worker’s DOE records usually contain sufficient information to determine if, and for what time periods, and under what conditions, such exposures may have occurred.
- The coworker data in OTIB-0062 (ORAUT 2007b) only goes through 1988; what coworker data will be used for the years 1989–2005 to assign exotic radionuclide doses?
- Can the 50th or 84th percentile level of intake (an average obtained over the population of LANL workers who handled radioactive materials) be used to bound the intakes of claimants working on specific projects that involved exotic radionuclides? This has yet to be quantitatively demonstrated and raises questions tied to the draft guidelines on the use of surrogate data proposed by the surrogate data work group. Generally, current NIOSH dose reconstruction practice tends to use the energy employee’s primary radionuclide intake (positive or based on MDA values), if available, to derive the intakes of other radionuclides for which monitoring was not performed; this accepted methodology uses data more relevant to the claimant’s individual work environment, as

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 19 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

opposed to using the average worker's (coworker) data that was obtained from a relatively large population.

- If the dose reconstructor uses the average LANL population's bioassay results (or the individual worker's bioassay results) to determine the primary radionuclide intake, how can it be assumed that the primary radionuclide concentration is always equal to or greater than the exotic radionuclide concentration? In non-routine operations, where exotics would more likely be found, the exotic radionuclide may have been in greater concentrations than the primary radionuclide, such as in processes that involved separation and concentration of exotic radionuclides. ORAUT-OTIB-0054 (ORAUT 2007a) limits the use of surrogate radionuclides to specific applications, because the ratios for other applications may not be known; e.g., where the exotics have been concentrated and/or removed from the normal reactor fuel cycle.

Summary

SC&A's search of LANL documents indicates that exotic radionuclides were not prevalent in most daily activities at LANL, and that they were not used in industrial quantities during the SEC period of 1976–2005. Additionally, there are indications that exotic radionuclides were controlled with methods that were at least as restrictive as those for the primary radionuclides that were monitored for. However, the potential SEC issues are:

- The ratio of the exotic to the primary radionuclides would not necessarily be known, constant, or limited by the primary radionuclide.
- By definition, the use of exotic radionuclides at LANL involved operations that were not in the main stream of routine operations. The primary radionuclide intakes averaged over the population of LANL standard workers who handled radioactive materials (i.e., coworker plutonium intake data) may not be representative of, or necessarily bounding for, the intakes of claimants working on specific projects that involved exotic radionuclides, which may not have been performed under the same operating conditions as the normal work performed at LANL.
- In the majority of the cases, individual worker's records do not provide sufficient information to be able to determine the time, duration, and conditions of exposures to exotic radionuclides. Therefore, dose reconstruction in many cases will result in gross overestimates of doses assigned, or failure to assign sufficient dose.
- As was the case for MFP/MAP, considering the unknowns of who was exposed and when, and to what concentration ratios, the proposed method does not necessarily meet the regulatory requirements of assigning **reasonable**, or **reasonably bounding**, doses.

There may be an avenue of validation that the LANL work group may want to explore. The question is whether some of the bioassay results that are available for the exotic radionuclides can be used to benchmark NIOSH's proposed method. For example, a comparison could be made between the doses derived from using the primary radionuclide bioassay results as a surrogate intake and the doses derived from the actual exotic radionuclide bioassay results. If a

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 20 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

number of these comparisons could be made, then it would assist in determining if the method is valid, or not.

To summarize, in light of the uncertainties associated with both Issues 1 and 2, and to assist in determining the validity of the potential SEC concerns, SC&A finds it would be useful for NIOSH to provide examples of specific dose reconstructions for unmonitored exposures from MFP/MAP and exotic radionuclides that utilize the suggested approach in NIOSH’s ER, using actual recorded data and parameters where possible.

2.3 COMPLETENESS AND RELIABILITY OF LANL IN-VITRO AND IN-VIVO DATA, AND ADEQUACY OF NIOSH COWORKER MODEL (ORAUT-OTIB-0062 AND ORAUT-OTIB-0063), POST-1975

A draft technical information bulletin, ORAUT-OTIB-0062 (ORAUT 2007b), has been developed for dose reconstruction for unmonitored individuals, using coworker data for Pu-239, Pu-238, uranium, tritium, and Cs-137. In addition to using the coworker data in OTIB-0062 to assign unmonitored doses from the primary radionuclides, such as plutonium, daily intake rates for Pu-239 taken from OTIB-0062, in units of pCi/d, may be assigned separately for each alpha-emitting exotic radionuclide that would have required similar controls: Ac-227, Pa-231, Np-237, and Th-230. To process an individual claim, on a case-by-case basis, the nuclide that results in the highest dose to the organ of interest for the energy employee could be used as the bounding intake. As previously discussed, the ER indicates that because the properties of Cm-244 are more similar to Pu-238, daily intake rates for Pu-238 could be used for Cm-244; and because the radiological properties and health physics controls for natural thorium are comparable to uranium, coworker data for uranium could similarly be used to bound intakes of natural thorium.

SC&A Preliminary Assessment:

Before a coworker dose model can applied to unmonitored, or under-monitored, workers for assigning primary, MFP/MAP, or exotic radionuclide doses, the data that was used to construct the coworker model must be validated and verified. SC&A performed a preliminary assessment of NIOSH’s proposed coworker model and its associated data; the following presents SC&A preliminary findings.

Coworker Databases and Associated OTIBs

LANL’s bioassay records have been stored in various hardcopy (mainly LANL notebooks) and electronic databases throughout the history of the Laboratory. In order to have a unified central database, the data from these legacy systems (Historical, Exiting, Old Electronic, Electronic, and BEST databases) were reformatted and entered into the LANL Bioassay Repository. This database contains the in-vivo data in one section (1960–present), and the in-vitro data in two sections (1944–1991) and (1991–present). Most of the bioassays were for the predominant (primary) radionuclides present at LANL throughout its operating history. These primary radionuclides were tritium, plutonium, uranium, americium, polonium, and cesium. There were also occasional bioassay results for thorium, and fission/activation products in the records.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 21 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Analyzing the amount of material available and the time span it covers can be complex and involved. However, the central question concerning LANL SEC-00109 is:

Are the bioassay records that are available in the present LANL bioassay database sufficiently accurate and adequate to allow dose reconstruction for LANL claimants during the SEC-00109 period (1976–2005), and does the database contain the data necessary to construct a sufficiently accurate and adequate coworker model for LANL claimants not monitored and/or not adequately monitored for the primary, MFP/MAP, or exotic radionuclide intakes?

To address this question, SC&A analyzed LANL SEC-00109, NIOSH’s ER, and the directly related technical information bulletins (TIBs); ORAUT-OTIB-0062 (ORAUT 2007b) and ORAUT-OTIB-0063 (ORAUT 2009a). (This was not a technical review of these technical information bulletins, but instead the information they contained was analyzed as it relates to SEC-00109.) The following is a summary of SC&A’s preliminary findings.

ORAUT-OTIB-0063 – This document provides analyses of the numerous bioassay database systems used throughout the history of LANL and their associated bioassay records (radionuclides, number of records, time periods, and databases where the data was stored). It does not provide any specific intake values, such as pCi/d, for the radionuclides (other than for a few MDA values). The questions concerning OTIB-0063 are:

- The number of bioassays in certain time intervals was provided, but how many bioassay data points are in each year/interval of data that were used to derive the data for OTIB-0062? Does the information concerning the number of samples for the radionuclides listed in NIOSH’s ER Table 6-2 apply to this data? The number of Cs-137 samples each year is not listed in that table; however, the adequacy of Cs-137 data is critical because assigning of MFP/MAP intakes in ORAUT-OTIB-0054 (ORAUT 2007a) is based upon it. To what extent are these data reflective of “event driven” monitoring, versus routine monitoring, and therefore, not necessarily representative of routine operations or operations that were not routinely targeted?
- Can some of the in-vivo data be used to validate (or not) NIOSH’s proposed method to assign exotic radionuclide doses?

ORAUT-OTIB-0062 – This document used the annual bioassay records (pCi/d), from data sources described in OTIB-0063, to derive the associated annual coworker doses for tritium and the intakes (pCi/d) for Pu-238, Pu-239, uranium, and Cs-137 using the IMBA program; these data apply to the period of 1975–1988 for this SEC (except Cs-137 extends up through 1993). The questions concerning OTIB-0062 are:

- Were the data sufficiently accurate and adequate to create the coworker intake values for each year? The 50th and 84th percentile bioassays results are listed in Attachment A of the ER by year; how many data points were there for each year for each radionuclide listed (same question as listed above for OTIB-0063)? Note that no bioassay data are listed for tritium, only the yearly dose results.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 22 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

- OTIB-0062 covers the SEC period of 1976–1988 (13 years); what will be used for coworker data the rest of the SEC period; 1989–2005 (17 years)?

Accuracy and Adequacy of Bioassay Record

Accuracy

The accuracy of transferred data can be verified by comparing the data recorded in the later database to that contained in an earlier database. In practice, 100% comparison is not practical; therefore, generally, random comparison of a small percentage of the data values is performed. During the consolidation of the bioassay data, NIOSH/LANL performed some verification of the data contained in the TUPo (Tritium-Uranium-Polonium) by comparing it to the original LANL notebooks, as described in Section 6.0 of OTIB-0063 and summarized in Attachment A of this document. SC&A's review of the results indicates that the accuracy of the important data (the actual bioassay results) was reasonable, with discrepancies ranging from 0.9% to 3.2%, and that a sufficient amount of data was compared, i.e., 10% to 39%. However, the notebooks were used in the earlier years, before 1980, and therefore, can only be used to verify the early portion of the data stored on the electronic databases.

SC&A did not find details of where there had been verification of the final records in the LANL Bioassay Repository compared to the original electronic databases. However, a document that accompanies the response from LANL when a claimant's records are sent to NIOSH to be used in dose reconstruction contains the following statements:

The attached data have been generated from the Los Alamos National Laboratory (LANL) Bioassay Repository. From June 2004 to May 2005, a team of LANL and National Institute for Occupational Safety and Health (NIOSH) personnel executed the Bioassay Data Repository Project to consolidate dosimetry records and make these data available for analysis. This database application is comprised of several applications including the Bioassay (BEST), In-Vivo Measurements Laboratory (IVML), and Radiological Incident Reports (RIR) where applicable. Electronic data from many other sources were collected and uploaded as part of this project. Over 3 million records comprise this repository, and a significant effort was expended to Verify and Validate (V&V) the bioassay data and personnel (Z-Number) data.

Although many hundreds of hours of effort have been expended, not all the V&V could be accomplished where the data was traced back to the original laboratory notebooks. The V&V that was accomplished focused on larger values or other records that might have dosimetric significance. From 2000 to the present virtually all the data is V&V'd; from 1990 to 2000 approximately 85% is V&V'd; from 1944 to 1990 the V&V varies by nuclide, ranging from 90% to 25%. Virtually all records of dosimetric significance have been V&V'd. LANL management and the NIOSH project manager have determined that the level of V&V is acceptable given the financial constraints of the Bioassay Data Repository Project.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 23 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

For the early part of the relevant SEC period in question, the 1970s to 1990s, it should be clarified by NIOSH what V&V has been accomplished for the radionuclides in question, particularly for MFP/MAP and exotics.

Adequacy

Because of the large amount and complexity of the bioassay data to be considered in both OTIB-0062 and OTIB-0063 in evaluating NIOSH's ER and the SEC issues, SC&A constructed several spreadsheets to provide a condensed view of the material; this information is contained in Attachments A, B, and C of this document, as described below:

Attachment A – Summarizes the **in-vivo** and **in-vitro** databases and the bioassay data available, which consist of the primary radionuclides, the total number of records, and the time periods of the bioassay records. There was no breakdown of the number of records on a yearly basis. Most of this information was obtained from OTIB-0063.

Attachment B – Provides a layout of the bioassay records available by radionuclide and time period for both in-vivo and in-vitro monitoring, and their associated databases, compared to the time period of the SEC and NIOSH's ER. Most of this information was obtained from OTIB-0063.

Attachment C – Provides a summary of the bioassay results, calculated intakes, and time periods for the primary radionuclides as found in OTIB-0062. These data will be used for assigning doses from coworker data to unmonitored, or under-monitored, LANL workers. It also contains an outline of the in-vivo and in-vitro bioassay data available by primary radionuclides and time periods according to NIOSH's ER. Note that most of the data in OTIB-0062 stops in 1988, whereas, NIOSH's ER states on pages 46 and 50 that OTIB-0062 covers the SEC period (1976–2005). There was no breakdown of the number of records on a yearly basis in either OTIB-0062 or in the NIOSH ER.

As can be seen from these spreadsheets, there is a large amount of bioassay data available in group form for the primary radionuclides. To date, there has been no indication that the data are not sufficiently accurate for dose reconstruction purposes. However, there has been no information provided that assures that there are not gaps in some years for which an explanation is not available. An additional concern is that OTIB-0062 only covers the years 1975–1988, which is 13 years out of the 30-year (1976–2005) time period covered in LANL SEC-00109. Adequate coworker intake data for the entire period is needed not only for the unmonitored, or inadequately monitored, workers, but also to assign doses from MFP/MAP and exotic radionuclides, as proposed by NIOSH's ER. The dose reconstruction process assigns internal doses on a per radionuclide per year intake basis; therefore, it is important that the data used from the worker's records, coworker data, or data used to assign MFP/MAP and exotic radionuclide doses be representative of the intakes for each year of dose assignment.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 24 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Summary

With some qualifications, there appears to be sufficiently accurate bioassay data available for the primary radionuclides, and there may be an adequate amount of such data for individual dose reconstruction for monitored workers and to develop a coworker intake data base. However, the following issues that may have SEC implications have not been sufficiently addressed:

- It has not been validated that there is sufficient bioassay data available and that a coworker model can be developed for the **entire** SEC period of 1976–2005 for use in assigning unmonitored worker doses, MFP/MAP doses, and exotic radionuclide doses.
- It has not been validated that there is sufficient data for **each individual year** to develop the coworker model, especially for Cs-137.
- It has not been demonstrated that it is technically feasible (by benchmark comparisons, etc.) to use coworker intake data for the primary radionuclides to bound intakes of the exotic radionuclides that were not monitored.
- The worker’s files do not generally contain sufficient information that would indicate if the worker was exposed, or not exposed, to MFP/MAP and/or exotic radionuclides, and during what time period(s). By definition, if the coworker model is needed, there is a lack of exposure and monitoring information.

2.4 FEASIBILITY OF DOSE ESTIMATION FOR NEUTRON EXPOSURE AT LANL, POST-1975

NIOSH indicates that prior to 1980, the NTA personnel neutron dosimeter was used to measure and record personnel neutron dose. Because NTA film-based dosimeters do not respond to neutrons with energies less than approximately 500 keV, the use of neutron-to-photon dose ratios is necessary to bound neutron dose prior to 1980 (ORAUT-TKBS-0010-6). After 1980, recorded neutron doses are considered to be sufficiently accurate, based on a combination of albedo TLD (for low- and intermediate-energy neutrons) and NTA film (for high-energy neutrons). Characterization of workplace neutron spectra at LANL showed that the neutron energies associated with plutonium-handling operations were consistent with those from well-moderated sources. Measurements performed in plutonium processing areas in 1978 indicated an average neutron energy of 200 keV. Neutron spectrometry data collected in these areas in 1993 showed that approximately 90% of the neutron flux was from neutrons having energy of 1.2 MeV or less. Characterization of neutron spectra at TA-53 showed neutron energy spectra could vary widely, and included areas where the dominant contribution to neutron dose equivalent came from neutrons having energy greater than 10 MeV. These measurements included evaluation of NTA and TLD measured neutron doses (ORAUT-TKBS-0010-6). The External Dose TBD, ORAUT-TKBS-0010-6 (ORAUT 2009c), contains sufficient information to allow bounding of neutron dose using, as necessary, neutron-to-photon dose ratios (n/p) in LANL areas where personnel neutron exposures occurred. It also contains sufficient information to allow assessment of missed dose and uncertainties associated with reported neutron dose after 1979, as well as reported neutron dose for the entire time period under evaluation. The combination of bounding neutron-to-photon dose ratios and the photon (i.e., gamma) dose information given in Table A-2 of ORAUT-TKBS-0010-6 therefore provides a means for bounding neutron dose for all

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 25 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

members of the class for the period prior to 1980, when NTA film was still the principal means of neutron monitoring.

The petitioners in SEC-00109 state that, “NCFs can vary by more than an order of magnitude at LANL facilities.” They also stated that considering that Support Service workers could work at several facilities during a day, dose reconstruction using the data from the LANL Model 7776 TLD cannot be done. However, NIOSH states that area-specific neutron correction factors (NCFs) were used at LANL to improve the accuracy of the neutron dose based on workplace instrument measurements (ORAUT 2009c). If workers frequented multiple facilities, or if the facilities frequented are unknown, NIOSH can bound neutron doses by applying the highest NCF for any of the buildings the worker may have entered.

SC&A Preliminary Assessment:

To assist in evaluating the validity of NIOSH’s proposed neutron dose reconstruction methods for LANL workers, and because neutron dosimetry issues are contained in various sections of the ER, SC&A constructed a brief outline of NIOSH’s ER, as follows:

Major Contents of NIOSH’s Evaluation Report

In the evaluation report, NIOSH provided information and recommendations concerning neutron dose reconstruction in Section 5.2.2.3 (External Radiological Exposure Sources from LANL Operations) on page 24; in Section 6.2 (Available LANL External Monitoring Data) on pages 31–37; in Section 7.1.2 (External Monitoring Data Pedigree Review) on page 40, and in Section 7.3.1 (Evaluation of Bounding Process-Related External Doses) on pages 50–52. Extensive references were made to the then-to-be revised LANL TBD ORAUT-TKBS-0010-6 (ORAUT 2009c), which was issued June 11, 2009, as Revision 01 PC-1-A. Therefore, much of this review will be based on information contained in that revised TBD-6. Following is a brief summary of each of the sections concerned with neutron doses in NIOSH’s ER, with reference to TBD-6, where appropriate. It should be noted that the changeover from using NTA film to the TLD badge Model 7776, and later to the TLD badge Model 8823, did not take place all at once, because it took time to implement each new system. Therefore, for this review, it will be assumed that the following neutron dosimetry was used for the dose of record during the indicated time periods:

- 1976–1979 = NTA film
- 1980–1997 = Model 7776
- 1998–2005 = Model 8823

ER Section 5.2.2.3 – External Radiological Exposure Sources from LANL Operations

According to page 40 of TBD-6, the major areas for potential neutron exposure at LANL include:

- D Building (TA-1)
- DP West (TA-21)

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 26 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

- DP East (TA-21)
- Current Plutonium Facility (TA-55)
- Omega Site (TA-2)
- LAMPF (TA-53)
- Criticality Lab (TA-2, TA-18)
- CMR Building (TA-3)

Neutron energies ranged from thermal (0.025 eV) to 20 MeV.

ER Section 6.2 – Available LANL External Monitoring Data

- NTA film was used during 1976–1979 for the dose of record.
- The Model 7776 TLD badge was used during 1980–1997 for the dose of record. Its response was very energy dependent (changing over an order of magnitude) and required NCFs specific to the neutron field of exposure to assign dose from the TLD readings. A detachable holder for NTA film was included for fast neutron dose measurements, but suffered from humidity and fading problems until 1990.
- The Model 8823 TLD with Track-Etch Dosimeter (TED), which contains CR-39 foils was used during the period 1998–2005 and is DOELAP approved in all categories and continues to be used to the present time.
- The entire workforce at LANL has never been badged during any given period; however, in 1960 about 50% of the total workforce was badged, 75% of Zia employees (support) were badged, and all of the security force were badged. In the 1970s, a checklist was developed to keep a record of who should be badged, and the checklist was evaluated and updated quarterly; this checklist is still currently in use. Tables 6-4 through 6-7 of NIOSH’s ER provide a summary of the number of workers badged, including average and maximum yearly gamma, neutron, and shallow doses.

ER Section 7.1.2 – External Monitoring Data Pedigree Review

NIOSH finds that the dose of record is sufficient for dose reconstruction purposes and can be used to bound potential neutron exposures. NIOSH proposes to use an alternate method (n/p ratio) as outlined in Section 7.3.1 (apparently not Section 7.3.4 as stated on page 40) to bound neutron exposures during the period 1976–1979 when NTA film was used.

ER Section 7.3.1 – Evaluation of Bounding Process-Related External Doses

Neutron dose is addressed on page 52. NIOSH contends that n/p ratios and knowledge of gamma doses (individual records or collectively as listed in Table A-2 of ORAUT-TKBS-0010-6) will allow NIOSH to perform bounding neutron dose reconstructions for the period of 1976–1979, when NTA film was used; after 1979, the TLD badges were sufficient to correctly measure and record low- and intermediate-energy neutron doses, with an additional NTA badge used if needed to measure high-energy neutron doses.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 27 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Neutron energy field characterization at LANL shows that the average neutron energy in the plutonium areas was around 200 keV in 1978. Neutron spectrometry data showed in 1993 that 90% of the neutron flux was ≤ 1.2 MeV in the plutonium areas. However, measurement at TA-53 (LAMPF) showed that the neutron energy spectrum could vary widely, including areas where the dominant neutron dose resulted from neutrons with energies >10 MeV.

NIOSH states that, "...sufficient information is available to allow bounding of neutron dose for all members of the class for the entire time period under evaluation."

SC&A's Review of NIOSH's ER Concerning Neutron Dosimetry 1976–2005

SC&A reviewed NIOSH's ER of SEC Petition 00109 and found that, while it addresses some of the dosimetry issues, not all aspects of the neutron dosimetry problems were resolved. The following is an outline of SC&A concerns in areas that have potential SEC significance.

1976–1979 Neutron Dose Reconstruction Issues

Considering precedent set at other DOE sites, SC&A finds that NIOSH's proposed use of the n/p method to assign neutron doses during the period 1976–1979 (when NTA film was used) to be an acceptable method. However, this is true only if the following two conditions are met: (1) the derived n/p values must represent the neutron-to-photon dose exposures that took place during the time period for which they are to be applied; and (2) the neutron doses, and photon doses, used to derive the n/p values must be reasonably accurate and not subject to indeterminate uncertainties.

SC&A is concerned whether the proposed n/p values listed in the revised TBD-6 (Table 6-22, page 50) would bound the neutron doses during the period 1976–1979, for the following reasons:

1. *Representativeness of the n/p values* – These n/p values were derived using neutron and gamma dose data from Model 7776 TLD badges during 1979–1997 and Model 8823 TLD badges during 1998–2004, with ≥ 50 mrem of dose each (Figure 6-3, page 36 of TBD-6). Therefore, the proposed n/p values to be used for the period 1976–1979 (5-year period) were determined over a period of 25 years following the application period. The latter period marks an era of more stringent regulations, and when processes and facilities were much more scrutinized for ALARA dose reductions, especially for neutrons. There was no supporting basis in the ER or TBD-6 to justify the assumption that the n/p values derived from 1979–2004 data could be applied to an earlier period that may have exposed workers to different neutron and gamma fields than were present in later years.
2. *Accuracy of the measured neutron doses* – As previously stated, the proposed n/p values were derived using neutron and gamma dose data from Model 7776 TLD badges during 1979–1997 and Model 8823 TLD badges during 1998–2004. As will be outlined in the following section of this report, the Model 7776 TLD had its own problem with accurately measuring neutron doses because of its energy dependence, as illustrated in Figure 6-2, page 30, of TBD-6. The energy dependence was to the extent [three orders of magnitude over the energy range of 50 keV to 14 MeV (LANL 1994, pdf. 10)], that different NCFs

had to be used for the different facilities at LANL, as shown in the following table (LANL 1994, pdf. 9 and TBD-6). The NCF is proportional to the inverse of the sensitivity factor, and the response of the Model 7776 TLD badges was multiplied by the associated NCF to determine the neutron dose.

Table 1. NCF vs. LANL Facility

Facility	NCF
TA-53 (LAMPF)	0.2
TA-55 GB [glovebox] with shielding	0.4
TA-55 GB w/o shielding	0.7
TA-2 select personnel	0.1
TA-18	0.07
All other badges workers	0.5
Visitors	0.5

Additional information concerning the variability of the Model 7776 response to neutron energy sources is shown in Table 6-15, on page 40 of TBD-6, and reproduced below as Table 2.

Table 2. Approximate NCFs and Dose Fractions for Neutron Sources

Type of Source	NCF	Dose Fraction by Energy Category			
		<10 keV	10–100 keV	0.1–2 MeV	2–20 MeV
Bare Pu-239	~1.0				
Bare Pu-Be	~1.5	1	1	33	65
Bare Cf-252	~1.3	0	0	42	58
Cf-252 through 10.2-cm Lucite	~0.15	5	1	33	61

Because of the uncertainties in the many variable neutron energy fields and the corresponding NCFs at LANL, NTA film was sometimes used to supplement the Model 7776 neutron dose data; however, this practice had to be discontinued because it invalidated the calibration of the badge and the DOELAP accreditation, and it did not address the NTA fading problems (TBD-6, page 29). Some use of supplemental NTA film was implemented at certain areas in LAMPF starting around 1990, where the NTA film was sealed and attached separately to avoid the earlier problems (Mallett et al. 1990). Considering these problems, and LANL's desire to discontinue using the Model 7776 and switch as soon as possible to the Model 8823 (LANL 2001a, pdf. 279), it does not appear that the neutron doses derived from the Model 7776 badge during 1980–1998 would be adequate for use in determining n/p values for 1976–1979.

1980–1997 Neutron Dose Reconstruction Issues

As previously outlined, the Model 7776 badge neutron response was very dependent on neutron energy; therefore, numerous NCFs had to be applied (some details are available in the reference, LANL 1994). While theoretically, the use of NCFs can correct for this variable energy response, in practice, this is very difficult to accomplish. The neutron energy spectrum changes with changes in shielding, surroundings, sources, etc. Even in normal operations, workers were

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 29 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

exposed to different neutron energy fields, to the extent that it would be difficult to measure and assign all the different NCFs as a function of location and time appropriate to all the workers. This situation would be compounded in an accelerator facility, where the shielding and operations are constantly changing, such as at LAMPF, where 1987 measurements (Mundis and Howe 1987) show that neutrons in Experimental Room One (ER-1) of LAMPF could have relatively high energies if there was an accidental beam spill (ORAUT 2009c, page 43):

In 1987, neutron energy spectrum measurements were made at a potentially high neutron energy area at LAMPF (Mundis and Howe 1987)... When unfolding codes were applied to the measurement data, they revealed that more than 90% of the neutron DE was due to neutrons of energy greater than 1 MeV, and 70% was due to neutrons of energy greater than 10 MeV.

However, other measurements showed:

In another study, however, 9-in. to 3-in. sphere ratio measurements at 18 locations at LAMPF (not just an area of potentially high neutron energy) yielded ratios that indicated an average neutron energy of <100 keV at LAMPF (Blackstock et al. 1978).

Therefore, it can be seen that the workers' recorded neutron doses from Model 7776 badges are the results of a very generalized facility NCF and not specific to the variable neutron energy spectra workers were exposed to on a daily basis at many of the LANL facilities. Even a conservative NCF (and occasional supplemental data from NTA film) would not account for all the variables that impact the neutron energy spectra, and hence the dosimeter response, at some of the facilities. Additionally, NTA film results were not reliable (because of fading) for even high-energy neutrons until ~1990 when they were sealed against moisture (Mallett et al. 1990). The amount of track fading in NTA film depends on such factors as the energy of the neutron creating the track, exposure and storage temperatures, humidity, and time between track formation and developing the film. Track fading can result in the loss of a few percent of the tracks per week up to a third of the tracks per week, or even the loss of the majority of the tracks over a long exchange period, depending on the conditions. Therefore, this leaves a 10-year period of 1980–1989 during which reliable high-energy dosimetry of neutrons at LANL, especially at LAMPF, was questionable using the Model 7776 TLD dosimeter.

In addition to the concerns with recorded dose, there is also the fact that the use of NCFs affects the limits-of-detection (LOD) values (LANL 1994, pdf. 23) that should be used to determine missed dose during dose reconstruction. As the energy of the neutron increases, the sensitivity of the TLD dosimeter decreases; hence the need for the use of a NCF. Along with this decrease in sensitivity, there is needed a larger minimum amount of neutron dose to obtain a recognizable output signal from the TLD chip. Therefore, the use of one LOD value for the TLD Model 7776 is not sufficiently accurate considering that the NCF can range over an order of magnitude (ORAUT-TKBS-0010-6, page 29).

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 30 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

1998–2005 Neutron Dose Reconstruction

SC&A has found that the Model 8823 dosimeter, accredited by DOELAP in all categories, could have some potential site profile issues, but at this time does not present any obvious SEC issues.

Summary

SC&A finds that the uncertainties associated with the corrections (n/p method to replace NTA film results, and NCFs that were used during the TLD Model 7776 era) may be approaching the same order of magnitude as the original LANL neutron dosimetry problems (NTA threshold/fading, and TLD energy dependence). Although, theoretically bounding neutron doses could be derived by using a large n/p value and by applying the highest NCF to the recorded dose, it is very difficult to meet the regulatory requirements of assigning a **reasonable**, or **reasonably bounding**, final neutron dose value (or LOD value for missed dose) to each worker for each exposure; especially when the dose of record contains only the final adjusted/derived neutron dose and not the original signal/dose measured. Therefore, neutron dose assignment is a potential SEC issue for the period 1976–1997 during which the NTA and Model 7776 neutron dosimetry were used; especially during 1976–1979, when using n/p values to assign neutron dose, and during 1980–1990 when supplemental NTA film for high-energy neutrons were not used or were unreliable.

2.5 FEASIBILITY OF DOSE ESTIMATION FOR RADIOLOGICAL EXPOSURE SOURCES AT LAMPF/LANSCE

Former maintenance workers at LAMPF/LANSCE (in this report the term LAMPF will be used to denote the LAMPF/LANSCE complex) claim that their work location adjacent to the Target Area A beam stop and near an adjacent retention pond may have exposed them to sources of radiation for which monitoring was either inadequate or lacking. NIOSH did not find any sources of external or internal exposure that would not have been monitored at LAMPF (or any other LANL facility, for that matter) during the period in question (post-1975).

SC&A Preliminary Assessment:

Background Information

During the 1980s and 1990s, some accelerator modifications, use of different target materials, and shielding configurations took place at LAMPF (for example, see Wangler and Lisowski 2003). Depending on the changes that took place, the dosimetry system in use, and monitoring/bioassay policies, the recorded (or lack of recorded) doses and intakes may, or may not, represent the doses received by the workers. In evaluating this situation, the main facts to consider are:

- (1) *Maximum energy* – The primary proton beam of the accelerator is limited to 800 MeV; therefore, the maximum energy of the radiation created cannot be increased regardless of changes in configuration. The primary proton beam of the accelerator has always

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 31 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

operated at 800 MeV since its start up in 1972; this is not dependent on its mission as a meson physics facility (LAMPF), or as a neutron center (LANSCE).

- (2) *Increase in beam current* – Increases in beam current could increase the amount of radiation produced, but not create any new types of radiation.
- (3) *Changes in target material* – Changes in target materials could create different local types of radiation inside and near the target during the time the beam is impinging on the target and different residual radioactive materials in the target after shut down. Also, changes in target materials could change the amount/ratio of neutron and photon radiation outside the shielding (perhaps muons in some cases), but would not create any new, exotic, or unknown/unexpected types of radiation fields.
- (4) *Changes in shielding* – Changes in shield thickness or configuration could result in changes in the amount/ratio of transmitted or scattered neutron and photon radiation outside the shielding. Any reasonable amount of shielding would prevent workers from being exposed to charged particles from the primary or secondary beams, or from charged particles created in the target or surrounding materials. Additionally, the full energy of the beam, or the maximum energy of produced radiation, is not present outside any reasonable amount of shielding, because the energy of the radiation is quickly degraded by the shielding.
- (5) *Exposures in experimental caves* – In experimental caves, where the beams have been extracted from the main beamline, exposures can consist of high-energy radiation, such as protons, neutrons, heavier nuclear fragments, pi-mesons, and K-mesons, high-energy electrons, bremsstrahlung, etc., resulting from cascades created by evaporation and spallation reactions of high-energy protons on a target in the main beamline, or in the extracted beamline. Personnel will most likely not be exposed to the charged particles outside the primary or secondary beamlines because of the structural materials and shielding. If a person crosses, or looks into, a secondary beamline in an experimental area when the beam is on, then large doses consisting of both low and high LET radiation could result in a very short time (i.e., rad/sec). This situation, if it did occur at LAMPF, would most likely be encountered by experimenters.
- (6) *Exposures in general work areas* – Exposure in the general working environment of an operating accelerator with adequate shielding is composed mostly of low Linear Energy Transfer (LET) radiation (photons, bremsstrahlung, and in a few cases, perhaps muons) with a Quality factor (QF) ~ 1.0 , and low to mid-energy neutrons in the range of thermal to 20 MeV (some isolated areas of higher neutron energy up to around 50–100 MeV could be possible, but unusual), with the majority of the neutron dose equivalent from neutrons with an average energy of around 1–5 MeV; usually having a higher LET, and hence $QF \geq 1$. This would be the case for most LAMPF personnel and support workers located in work areas that surround the east end of LAMPF; such as around and outside the experimental areas shielding, beam stops, target areas, and inside buildings or trailers. This would correspond to areas in the lower part of Figure 1 shown below. This radiation exposure can generally be measured satisfactorily using photon film/TLD and neutron NTA film/TLD badges, **if properly calibrated** to the radiation fields. Sometimes track-etch detectors (TED), using CR-39, are also used if there is a potential for higher energy neutron exposure. Although, the maximum energy of the accelerator is 800 MeV, the

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 32 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

energy of the neutron fields outside of any shielding is in the range of thermal to 20 MeV; perhaps up to 50–100 MeV in unusual situations; therefore, dosimetry that normally detects neutrons in this energy range will respond to the radiation fields the workers are exposed to in this area. If a worker is wearing a dosimeter that detects photons, beta, and neutron radiation and it is properly calibrated to the radiation fields to which the worker is exposed, then the resulting dose of record will correctly reflect the doses the body received. This is true regardless of the type of targets, shielding, scattered neutrons, accelerator operating parameters, etc., as these do not produce any unusual radiation that is not detectable.

- (7) *Badging* – During accelerator operations, gamma/beta and neutron dosimetry badges should have been worn by most personnel; neutron dosimetry badges were not needed during periods that the accelerator was shut down as neutrons were not present when the beam was not operating. Exposures from activated components and shielding would consist of gamma and beta (no neutron) radiation of energies up to approximately 3 MeV, which would be detectable by standard gamma/beta dosimetry.

Therefore, in order to determine if a worker’s dose of record correctly reflects the worker’s exposure at LAMPF, the main areas to consider are:

- Was the worker badged with the appropriate photon and neutron dosimetry when working in radiation fields at LAMPF?
- Were the dosimeters correctly calibrated for the radiation fields to which the worker was exposed?
- Were the dosimeters properly read and recorded in the records?
- Are the records available to the dose reconstructor?

If the answer is “yes” to all of these questions, then the operating conditions at the accelerator, target material, shielding arrangements, location of the worker, etc., should not impair the ability to perform a reasonable dose reconstruction. Weapons- or defense-related experimental activities at the LAMPF complex would not change the basic sensitivity, capability, or results, of personnel dosimetry.

SC&A’s Evaluation of 1980s–1990s Monitoring at the LAMPF Accelerator

External Doses

SC&A reviewed ORAUT-TKBS-0010-6 and related LANL documents (such as LANL 1978–1985 and LANL 1990b) concerning the possible radiation fields and the dosimetry methods pertinent to the period 1980–1999. SC&A did not find an indication that any exotic or unknown types or amount of radiation fields were present at LAMPF during this period (see above discussion). The main concern is whether the worker was properly badged and the dosimeter was calibrated to the appropriate radiation field to which the worker was exposed. During the period 1980–1997, the Model 7776 TLD badge was used to monitor radiation workers at LANL, along with some NTA film for high-energy neutron monitoring at LAMPF during 1980–June

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 33 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

1981, and also ~1990–1994; NTA film was replaced with TEDs for high-energy neutron monitoring in ~1995 (ORAUT-TKBS-0010-6, pages 24–25).

SC&A found that external dose concerns for monitoring at LAMPF during the 1980s–1990s were not those associated with new exotic, unknown, or undetected radiation fields, but the same as those addressed in the previous SEC period for the LANL site, which is concerned with the badging, dosimetry response, and recorded dose. Specific items relative to LAMPF workers are:

- *Use of only one NCF* – The use of a single NCF of 0.2 for the TLD Model 7776 badge when it has been shown that the neutron energy spectrum at LAMPF can vary considerably as a function of location.
- *Lack of details of NTA film use* – Although TBD-6 and NIOSH’s ER state that NTA film was sometimes used in addition to the TLD Model 7776 dosimeter to monitor high-energy neutrons, the details of who, when, and where this additional monitoring was performed have not been addressed; i.e., how was it determined that a worker should receive an additional NTA badge? Was this additional monitoring only for experimenters, or did it also include craft and support workers? TBD-6, page 30, states that <40 workers used NTA film during 1981–1995; this would result in a very low percentage of workers monitored for high-energy neutrons, even if they were all located at LAMPF.
- *1980–1989 NTA film results were unreliable* – NTA film results were not reliable (because of fading) for even high-energy neutrons until ~1990 when they were sealed against moisture (Mallett et al. 1990). Therefore, this leaves a 10-year period of 1980–1989 during which high-energy dosimetry of neutrons at LAMPF was questionable.
- After the introduction of the TEDs for high-energy neutrons (~1995) and the Model 8823 TLD dosimeter (~1998), the reliability of the dosimetry was improved.

Internal Doses

Issues concerning internal intake of MFP/MAP and tritium that may have been present at the LAMPF accelerator are covered in other sections of this report.

Evaporation Ponds (Lagoons)

One item of concern expressed by the craft and support workers was the presence of tritium, and other radionuclides, in the evaporation ponds (lagoons) at the south-east corner of LAMPF. There were three wastewater lagoons at that location. Two unlined lagoons (no longer used) collected sanitary wastes prior to construction in 1994 of the LANL-wide sanitary waste treatment facility at TA-46. Traces of both radioactive and hazardous wastes have been discovered in the sludges in these lagoons, and they required closure under the Resource Conservation and Recovery Act (RCRA) regulations. The third lagoon, lined with Hypalon®, continued to receive low-level radioactive liquid wastes from floor drains in the LAMPF accelerator building and experimental halls. All three lagoons were to undergo RCRA closure prior to 2002 (LANL 2001b). The photo (LANL 2005), Figure 1, below shows the east end of LAMPF; the ponds are located in the lower left-hand corner of that photo.

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Figure 1. Los Alamos Meson Physics Facility East End (Experimental Area)

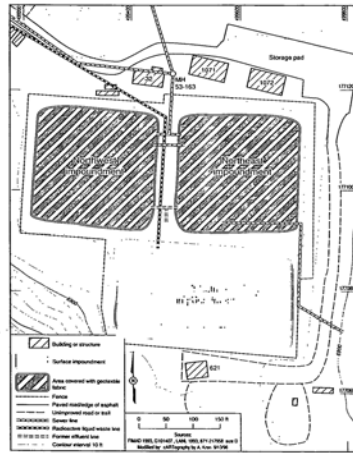


Figure 1-1. Areas covered with geotextile fabric.
September 25, 1996 -2- IA Report for TA-53

Figure 2. West and East Evaporation Ponds

Source: LANL 1996

Figure 2 shows two ponds (East and West) after they have been covered with a Geotextile® filter fabric (and no longer in use) in 1996, to prevent wildlife intrusion and the potential generation of dust. There was a third evaporation pond, the South pond, not shown in this diagram, which was still in use in 1996 (LANL 2001b). More information concerning these evaporation ponds and their cleanup can be found at the following website location:

http://www.lanl.gov/environment/cleanup/docs/factsheets/fs_ta53_lagoons_er2002-0285.pdf

Evaporation Ponds – External Doses

While the ponds were being used, the water in the ponds attenuated much of the gamma radiation that would have been emitted from the radioactive material contained in the sludge.

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However, when the ponds were no longer in use, much of the water evaporated in the arid climate and left the sludge exposed. This resulted in a measurable external dose around the areas of the ponds. For example, the 1999 LANL *Environmental Surveillance Report* (LANL 1999b) shows external annual doses (24/7) in Table 4-16 (pdf. 154) as 2,157 mrem/y at the South LANSCE Lagoon and 3,122 mrem/y at the East LANCE Lagoon, compared to a natural background of around 150 mrem/y external dose. Therefore, workers in the vicinity of the evaporation pond could have been exposed to external doses of gamma radiation (but not neutrons) from the contents of the lagoons. A worker should have been badged if located in this area, and the external exposures would have been recorded on the worker's dose of record.

Evaporation Ponds – Internal Doses

An example of the radioactive materials in the water contained in the ponds is illustrated in the following analysis:

A. Gamma analysis ($\mu\text{Ci/l}$) of East, West, and South ponds, 4/25/1989 (LANL 1990a, pdf. 2):

East Lagoon	4/25/89	0900	4/27/89	0948	Na-22	4.5E-03
					Mn-54	7.9E-03
South Lagoon	4/25/89	0900	4/27/89	1014	Co-57	3.5E-03
					Na-22	1.0E-02
West Lagoon	4/25/89	0900	4/27/89	1034	Co-60	4.5E-03
					Co-58	3.1E-03
					Co-57	1.2E-02
					Be-7	2.8E-02
					Mn-54	1.6E-02

B. Tritium analysis of East, West, and South ponds, 11/5/1990 (LANL 1990a, pdf. 79):

Sample Identification	location taken	alpha (uCi/l)	Beta (uCi/l)	Tritium (uCi/l)
1	East Lagoon			NDA
2	West Lagoon			NDA
3	South Lagoon			8.26

The south lagoon was still in use at this time.

To obtain an idea of the magnitude of these activities, Table 3 compares the results of the above analyses to the 10 CFR 20, Appendix B (1998) effluent release limits to public areas.

Table 3. Comparison of Measured Activity to Public Release Limits

Radioisotope	Location of max conc.	LANL results (μCi/l)	10 CFR 20 limits (μCi/l)	Ratio LANL/10 CFR 20
Co-57	West Lagoon	1.2E-02	6E-01	0.02
Co-58	West Lagoon	3.1E-03	2E-01	0.02
Co-60	West Lagoon	4.5E-03	3E-01	0.15
Mn-54	West Lagoon	1.6E-02	7E-1	0.02
Na-22	South Lagoon	1.0E-02	6E-2	0.17
Be-7	West Lagoon	2.8E-02	6	0.005
H-3	South Lagoon	8.26	1	8.3

This indicates that the concentration values were around the limits of the allowable effluent discharge to the environment at the time of their analysis, which was performed in the middle of the time period of concern (1980–2005). Therefore, it appears that, while there was water in the ponds, the occasional potential exposure to radioactive materials from the evaporation ponds would not constitute a significant internal dose hazard to the LAMPF workers, unless a worker was located at and/or worked with the sludge in the ponds on a continuous basis. After the water in the ponds evaporated, wind could have carried radioactive materials in the form of dust in the air creating potential internal intakes.

Summary

LAMPF

For **external** exposures at LAMPF, SC&A found no unique or unexpected radiation that would not be detected and recorded by properly calibrated and processed badges for service support or craft workers present around the accelerator areas. Issues concerning internal intake of MFP/MAP and tritium that may have been present at the LAMPF accelerator are covered in other sections of this report.

Lagoons

For **external** exposure at the lagoons, SC&A found that there are no areas of concern specific to the lagoons that were not of concern as identified in previous dosimetry issues; i.e., the main concern is the wearing of the proper dosimetry, its calibration, and dose of record. With or without water in the ponds, the external doses would have been registered and recorded if the worker wore the standard gamma/beta badge; if the worker was not badged, there may have been potential for unmonitored external doses. Neutron exposure was not present from the lagoons.

For **internal** exposure, while there was water in the evaporation ponds, the doses most likely would have been from mist and/or evaporation, and given the location of the maintenance workers, would have been due to immersion in whatever airborne tritium was present (in addition to contributions of other airborne, suspended radionuclides). While environmental monitoring measurements are available (RCRA sampling data and fence line airborne tritium data), no bioassay monitoring data are available and no air monitoring data in the immediate vicinity of the ponds were found in an initial onsite data capture. As it is plausible that an upper

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 37 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

bound estimate could be estimated based on available maximum tritium concentration data for the retention ponds in question during the relevant time period, a request was made of interviewed LANSCE personnel for such data (it was confirmed by them that it does exist and is on file at LANSCE). However, in subsequent contacts with LANL, this data could not be made available and a formal request through DOE is pending. The resolution of this issue rests with the availability of this data for the period in question (early 1990s) and whether there is a method by which it can be applied to establish an upper bound estimate of potential internal dose to nearby unmonitored workers.

2.6 RADIATION EXPOSURE TO SPECIAL TRITIUM COMPOUNDS

LANL handled special tritium compounds including stable metal tritides and organically bound tritium compounds. The NIOSH's ER provides an overview of the uses and forms of tritium at LANL (NIOSH 2009, page 20):

Tritium is a primary component of thermonuclear weapons and was used extensively at LANL throughout the time period under evaluation. Megacurie quantities of H-3 have been handled at LANL. Tritium at LANL has taken several forms, including [tritiated] water (HTO), organically-bound tritium (OBT), metal tritide (MT), and gaseous hydrogen (T₂ or HT). Gaseous tritium could have been contained as a pressurized gas or adsorbed onto various metals (e.g., uranium or palladium).

On page 47 of the SEC Evaluation Report, NIOSH describes the available data for analysis of tritium and the proposed approach for unmonitored workers:

The tritium bioassay program at LANL was extensive. Monitored worker doses were determined through urine bioassay. These data are readily available to bound tritium dose to monitored workers.

Unmonitored worker intakes may be bound using monitored co-worker data. Internal dosimetry co-worker data for LANL are presented in ORAUT-OTIB-0062 (draft).

Exposures to tritiated water and/or vapor (HTO) and gaseous tritium (HT) were the most prevalent forms of tritium exposure at LANL. Organically bound tritium (OBT) would have been encountered only in locations where biological research with labeled compounds was conducted. Exposures to the stable metal tritide (SMT) form of tritium may have been encountered as the result of the storage of tritium adsorbed on rare metals. Exposure to these compounds is a specialized circumstance [Inkret et al., 1999]. On a case-by-case basis, intakes of OBT and SMT compounds may be bounded using the methodologies outlined in ORAUT-OTIB-0066.

NIOSH has indicated that the assignment of dose from exposure to special tritium compounds can be done with sufficient adequacy.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 38 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

SC&A Preliminary Assessment:

The Department of Energy (DOE) and its predecessor agencies have undertaken missions involving work with hazardous and/or exotic materials and agents, and byproducts of their production, storage, and use. Included in these exotic materials are metal tritides (MT) and organically bound tritium (OBTs), referred to collectively as special tritium compounds (STCs). Los Alamos National Laboratory was actively involved in handling STCs. Other STCs have been introduced to sites as a byproduct of handling large quantities of elemental tritium and tritiated water, and its absorption into materials (e.g., rust, tritiated oil, tritiated dust, etc.). STCs differ from tritium oxide (HTO) and elemental tritium (HT) in their physical and chemical properties, which may make their detection, characterization, and subsequent assessment of exposure affects difficult.

The NIOSH’s ER indicates exposures to stable metal tritide (SMT) forms of tritium “may have been encountered as a result of the storage of tritium adsorbed on rare metal.” Furthermore, the NIOSH’s ER indicates that exposures to specialized compounds will be reconstructed based on the methodologies in ORAUT-OTIB-0066 (ORAUT 2007c). Page 47 of NIOSH’s ER indicates that organically bound tritium (OBTs) “would have been encountered only in locations where biological research with labeled compound was conducted.” From these brief acknowledgements, it appears that NIOSH’S ER has inadequately identified the STCs found at LANL and the potential exposure conditions that existed.

LANL has been involved in work with tritium since the 1940s. STCs identified to date at LANL include:

- Erbium tritide
- Iron oxide tritide (byproduct)
- Hafnium tritide
- Palladium tritide
- Titanium tritide
- Uranium tritide
- N(D,T)₃
- Li(D,T)
- U (D,T)
- Tritiated dust (byproduct)
- Tritiated mercury (byproduct)
- Tritiated solvents (byproduct)
- Tritiated methane (byproduct)
- Tritiated oil (byproduct)
- Tritiated siliceous materials (byproduct)
- Other tritiated carbonaceous materials

These include the more insoluble STCs, such as hafnium tritide, for which dose reconstruction methods are still challenging, given the lack of monitoring data, operational information, and worker rosters.

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 39 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Some of the major tritium facilities formerly or currently at LANL include:

- High Pressure Tritium Laboratory (HPTL) (TA-33)
- Tritium System Test Assembly (TSTA) Facility (TA-21)
- Tritium Science and Fabrication Facility (TSFF) (TA-21)
- Weapons Engineering Tritium Facility (WETF) (TA-33)

The HPTL (also called the Gas Handling Facility) was originally tasked with conducting research and development on tritium handling technology. This facility took over production work for a period of time, processing tritium gas (HT) and repackaging HT into small-volume high-pressure vessels. In 1992, WETF began receiving tritium and tritium-contaminated gases, repackaging these gases, and preparing them for shipment to other sites. TSFF provided a facility for R&D involving work with HT and metal tritides (MT). TSTA was involved in the use of tritium for development and demonstration of fusion fuel cycle technology.

Some of the other areas where STCs were handled in biological experiments and Research and Development (R & D) activities (or as a result of formation of STCs from diffusion, absorption, and reactivity of elemental tritium and HTO) include:

- Area G (Waste Management)
- LAMPF/LANSCE (TA-53)
- Plutonium Facility Site (TA-55)
- TA-48 (NTS Testing Sample Analysis)
- CMR (TA-29)
- Van de Graaff Accelerator (TA-3-16)
- Health Research Laboratory (HRL)

In addition to the use of SMTs in production processes and surveillance activities, tritiated metals can also be produced as a byproduct of handling large amounts of HT or HTO.

Tritium as HT or HTO will readily adsorb onto the surface of most metals (e.g., stainless steel, copper, and aluminum), plastics, and rubbers. The tritium will remain fairly close to the surface unless the metal is heated to high temperatures. At room temperature, permeation into these metals is extremely slow [DOE 2008].

And

Tritiated metals, metal oxides, dust, and oil can occur in nearly any tritium area. Any tritium contamination collected on a swipe survey or a particulate filter should be viewed as a potential combination of these [DOE 2008].

And

Any area where tritium is used, handled or stored may result in the formation of metal tritides [Pantex 2004].

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 40 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

If tritium is present in material containing hydrogen, the tritium will exchange to form a tritiated molecule of that material.

OBTs are produced when tritium forms a chemical bond with an organic material, typically by creating carbon-tritium bonds. Under some conditions this is done intentionally, such as radioactive labeling of biological molecules, and in other cases, it is a byproduct of organics adsorbing elemental tritium or tritium oxide. Solid particulate OBTs are largely formed by incidental contamination of environmental dust and other materials found in most tritium contamination areas. DOE-HDBK-1129-2008, *Tritium Handling and Safe Storage*, describes the interaction of tritium with hydrogenous materials (DOE 2008).

If adsorbed onto hydrogenous material, the tritium will easily permeate into the material. The HTO will move much more rapidly into the bulk material than will HT. The permeation rate varies with the type of material and is accelerated by increasing the temperature. As a result of this movement, plastics and rubbers exposed to tritium (especially HTO) are readily contaminated deep into the bulk material and are impossible to decontaminate completely.

Insoluble OBTs include tritiated pump oil, organic dust, pump oil droplets, tritiated fly ash, and tritiated nylon. Soluble OBTs include tritiated solvents and methane (DOE 2008). Particulates can be generated during dispersal mechanisms from material such as plastic, nylon, organic dust, or large molecule components of OBT oil. Insoluble OBTs present the same challenges in detection and measurement that other insoluble particulates present.

As opposed to production activities, STCs produced through diffusion, absorption, and reactivity of elemental tritium (HT or T₂) impact more operations and potentially expose a larger population of individuals. Whereas in tritium production facilities, controls for tritium exposure include containment systems, special handling procedures, routine monitoring, and special radiological controls, this is not always the case in areas where STCs are formed as a byproduct of having tritium present.

Bounding techniques proposed in ORAUT-OTIB-0066 cannot be effectively developed and applied with sufficient accuracy without some basic knowledge of the STCs handled, the quantities of material, the locations and time periods of potential exposure, and the physical behaviors of tritium compounds in the environment (e.g., conversion to HTO, formation of rust) to correctly characterize tritium exposure. The selection of parameters for maximizing doses (i.e., particle size, solubility, chemical form) is not feasible for some compounds, given the lack of biokinetic information available in the open literature on many of the compounds to which workers are potentially exposed. Where information is available, the literature seems to provide some disagreements about the solubility classifications that should be assigned to some of the SMTs (Potter 2004; Cheng et al. 1997; Cheng et al. 2002; Zhou and Cheng 2004; DOE 2008). However, several of these SMT compounds appear to be quite insoluble, such as hafnium, zirconium, and titanium, which have similar Dose Coefficients, Allowable Limits on Intake (ALI), Derived Air Concentrations (DACs), and very similar Intake Retention Fractions (IRFs). An IRF is the fraction of the intake that is retained in the body at time (t) following the intake; for these radionuclides, at 100 days post-intake, the IRF is 1.03×10^{-3} for hafnium; 1.03×10^{-3}

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 41 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

for zirconium; and 1.04×10^{-3} for titanium; and at 1,000 days post-intake, the IRF is 2.13×10^{-11} for hafnium 2.13×10^{-11} for zirconium and 2.14×10^{-11} for titanium (Potter 2004).

Caution must be observed when using available bioassay data, because factors such as particle size, temperature, shape (e.g., self absorption), humidity, and the presence of deuterium in the compound can affect the solubility of the compound and are not available in many cases. Because dissolution rates vary by orders of magnitude, an incorrect choice of a very slow dissolution rate can lead to intake (and dose) estimates that are overestimated by orders of magnitude when derived by urine data. This overestimate is confounded when intakes of HTO accompany particulate intakes.

NIOSH's ER does not provide guidance on how to distinguish between intakes of STCs, elemental tritium, tritiated water, or multiple STCs, which may occur simultaneously and overlap. Developing a model based on existing bioassay data is confounded by potential exposures to tritiated water and multiple tritides with different effective half-times in the body, making the biokinetic behavior for particular STC intakes more difficult to determine when interpreting bioassay data. Furthermore, it is unclear whether the ICRP model for OBTs referenced in OTIB-0066 reflects the organic compounds to which workers were exposed.

NIOSH has indicated that the exposures to tritides are site-specific issues. A key factor in the site-specific analysis is the identification of individuals who were potentially exposed to STCs. There has been no detailed analysis of the locations where STCs were handled and the individuals who worked in these areas. Personnel and field monitoring data focused on potential exposure from tritiated water and other soluble forms of tritium, which resulted in a failure to adequately monitor for insoluble forms of tritium. Results from traditional bioassay method, as proposed by NIOSH, cannot be used to determine plausible tritium doses from insoluble tritium compounds. OTIB-0066 acknowledges this with the following statement (ORAUT 2007c).

If the metal substrate of the SMT is not known, type S solubility should be assumed. However, fairly modest tritium urine concentrations can imply extremely large type S SMT exposures that might be quite implausible. For example, 1 $\mu\text{Ci/L}$ of tritium in the urine that is assumed to be the result of an intake of Type S SMT 30 d earlier implies an unencapsulated source term in excess of 300 mCi. This assumes that the fraction of an accidental release inhaled is 1×10^{-6} .

This is reaffirmed in the Mound Technical Basis Document for Stable Tritiated Particulate and Organically Bound Tritium (Mound 2001). Furthermore, questions were also raised regarding assessment of dose from tritium labeled organic compounds (Taylor 1986):

It is concluded that although the ICRP OBT model may underestimate doses for specific compounds by up to an order of magnitude, it can still be applied with caution for prospective radiological protection purposes, but it should not be applied for the interpretation of bioassay data.

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 42 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

Summary

No consideration has been given to the diffusion, absorption, and reactivity of elemental tritium and tritiated water that produce STCs as a byproduct of normal operation. As opposed to production activities, STCs produced through diffusion, absorption and reactivity of elemental tritium (HT or T₂) impact a larger population of individuals. Furthermore, many of the situations where this becomes an issue do not have the same level of engineering and administrative controls as those seen in tritium processing areas. From brief acknowledgments provided in the ER and SC&A's review of operational history, it appears that potential exposure pathways to the more insoluble STCs likely existed at LANL; therefore, some characterization is needed of what compounds were significant, what workers may have been exposed (or whether they can be identified), and whether historic exposure pathways were significant enough to warrant dose reconstruction.

In the final analysis, these issues may best be resolved through a “proof of principle” demonstration that dose reconstruction is feasible with available site-specific data.

2.7 UNMONITORED EXPOSURE OF SUPPORT SERVICE PERSONNEL

The Zia Company was the principal subcontractor to LANL from 1946 to June 1986. During this time frame, as many as 15,000 workers were employed to provide a broad range of site-wide maintenance, construction, janitorial, waste management, and facility support activities; much of it involving potential radiation exposure. Security guards and fire fighters also figured prominently as support service workers who had free access to almost the entire LANL site and therefore, had potential exposure to most radiation sources. In addition to raising issues regarding the adequacy of data needed for dose reconstruction after 1975, the petitioner explicitly questioned NIOSH's ability to reconstruct internal doses, given what was indicated as the inadequacy of PPE and the lack of routine bioassay monitoring. As noted in the ER:

The petitioner also provided ten affidavits (listed in Section 4.7). A number of these affidavits asserted that Service Support Workers with inadequate or no PPE were routinely assigned to areas in which workers were using full PPE. They also asserted that these Service Support Workers had little or no participation in the LANL urine sampling or whole-body counting program. Because this petition qualified for further evaluation based on the issues discussed in the preceding paragraph, these affidavit statements will be considered during the evaluation.

While the internal dose TBD, ORAUT-TKBS-0010-5 (ORAUT 2009b) provides cursory information regarding Zia employees and other support service employees, little detailed information is provided regarding what is characterized in the TBD as a “*separate*” monitoring program. This is of particular concern because Zia workers were involved in almost all of the radiological operations at the laboratory during most of its history, and were frequently called upon to conduct jobs involving potentially significant internal and external radiation exposure potential including decontamination, radioactive waste disposal, and “hot” maintenance. While the Zia monitoring program later had a computer program, initiated in 1976 (ORAUT 2009c, page 97), that “locked out” (i.e., administratively restricted) access to plutonium areas for

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 43 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

workers not bioassayed within 425 days, it is not clear from the TBD how this measure would have precluded workers from receiving uptakes and then discontinuing employment, or moving to other radiological areas, and therefore not being bioassayed. The site profile and the ER do not provide the requisite basis for determining what the potential missed and unmonitored dose may have been for the Zia workforce, given the spectrum of radiation sources involved. Similar concerns apply to how dose estimation will be handled for unmonitored or inadequately monitored LANL security guards who had site-wide access and were frequently in locations where potential and external radiation exposures were likely.

SC&A Preliminary Assessment:

SC&A analyzed the available information concerning internal and external monitoring of the LANL support service workforce. The following represents SC&A’s initial findings.

Analysis of claims – To assist in determining the **adequacy** of bioassay data available for the dose reconstructor to use for the dose reconstruction process for service support personnel, SC&A sampled 30 LANL dose reconstruction claims for workers employed between 1976 and 2005, with job titles that would indicate that their work could have exposed them to intakes of radionuclides, but with most having been service support or craft workers who may not have been monitored (which was of concern in LANL SEC-00109). SC&A selected the claims on the basis of position title and years worked during the SEC period (1976–2005) before viewing the bioassay records themselves. Therefore, the amount of bioassay records available did not bias the selection of the workers to be analyzed.

This very limited sample indicates that, in general, bioassay monitoring was based on the individual work situation rather than by worker category, i.e., two chem techs had 100% monitoring and another chem tech had 27% monitoring. A heavy equipment operator, welder, plumber, and machinist had over 50% monitoring, while a research physicist and several lab associates had 0% monitoring. Custodians/janitor monitoring ranged from zero to 100%.

Most in-vivo bioassays were chest counts taken once, sometimes twice, in a year; a few were whole body counts (WBC) using gamma-detector arrays. Many of the in-vitro bioassays (urinalyses) were taken numerous times during a given year, indicating routine bioassays. While the results of this analysis are not necessarily definitive, they do indicate that a “reasonable” (but not necessarily sufficient) amount of bioassays were routinely taken and recorded for various security/service/craft personnel for some periods. As noted under preceding SC&A issues 1 and 2, it may not be feasible to apply a coworker model to estimate an upper bound dose for potential exposures to exotic radionuclides to which workers, monitored or unmonitored, may have been exposed for the early part of the SEC period.

In summary, SC&A sampled and analyzed 30 LANL claimant files, consisting mostly of security, service, and craft personnel, who may have worked with radioactivity materials between 1976 and 2005. From this limited analysis, it appears that, in general, all categories of workers (security, service, craft, techs, staff, etc.) were bioassayed according to duties performed and not according to job title or employer, and that a reasonable amount of bioassay data may be

available for the 1976–1995 time period for these 30 employees. There are indications that the number of bioassays generally declined in the mid-1990s for many LANL workers.

Table 4. Years Bioassayed Compared to Years Worked and Position Title

Case*	Position title	# of Years Worked 1976–2005	# of Years Bioassayed [†] 1976–2005	# Yrs bio/ Yrs Worked
A	Chem tech	18	18	1.00
B	Chem tech	13	13	1.00
C	Janitor	11	11	1.00
D	Health tech	8	8	1.00
E	Heavy equipment operator	20	17	0.85
F	Tech I	27	22	0.81
G	Laborer	26	21	0.81
H	Security inspector, Firefighter	14	11	0.79
I	Welder	22	15	0.68
J	Guard	12	8	0.67
K	Plumber/Steamfitter	9	6	0.67
L	Plutonium recovery	18	11	0.61
M	Machinist	16	9	0.56
N	Uranium custodian	16	8	0.50
O	Security guard	18	9	0.50
P	Maintenance machinist	15	7	0.47
Q	Custodian	22	10	0.45
R	Electrician	21	8	0.38
S	Equip operator, security inspector	18	6	0.33
T	Carpenter	18	6	0.33
U	Test measurement tech	22	7	0.32
V	Chem tech	15	4	0.27
W	Iron worker	15	3	0.20
X	Electronic tech	15	2	0.13
Y	Custodian, Security escort	25	1	0.04
Z	Custodian, Laborer	20	0	0.00
AA	Sheetmetal worker	12	0	0.00
BB	Research physicist	15	0	0.00
CC	Lab associate	16	0	0.00
DD	Lab associate	19	0	0.00

* These are randomly assigned case designations.

[†] Urinalysis and/or WBC and/or chest ct. Many had multiply bioassays per year.

Monitoring of Security Guards

From interviews with current and former guards, fire fighters and support service workers at LANL, a number of them apparently were not routinely bioassayed over most of their employment at LANL. Consistent with the affidavits submitted with the petition, they provided many examples of instances where they were stationed or frequented locations where sources of known radiation exposure existed, where operators wore PPE or had routine bioassay and they did not, and where former practices were superseded by more stringent ones requiring PPE and bioassay as policies shifted (e.g., after the Tiger Team review). Occurrences such as the Cerro

Grande fire were highlighted to illustrate their view that guards and firefighters were, in particular, required to risk unmonitored exposure to unknown sources of radiation exposure where the dose received was not recorded and would be difficult, if not impossible, to estimate.

From interviews with LANL health physics and operating personnel, the guards generally were required to patrol in the “clean” radiological areas; e.g., for TA-55, this meant staying out of radiation controlled areas (RCA) and remaining in hallway corridors. They were stationed in and patrolled through hallways, stood near shipments, and were stationed at entryways to sensitive facility locations. External radiation exposure was routinely monitored and was found to be generally minimal; all incoming nuclear package shipments were swiped for contamination before they left their point of origin and were swiped again at LANL upon receipt. If there was an event or contamination, CAMS would alarm and everyone, including the guards, would need to evacuate. Over the past 20 years, the health physics staff observed that there had been no history of guards being significantly contaminated or having elevated radiation dose. However, it was pointed out that the guards were often nervous about being in proximity of certain radiological operations and were sometimes given “real-time” dosimeters; this was done when criticality experiments were run at TA-18.

Until two years ago, the LANL health physics department had indicated that there was no need to bioassay the guard forces in TA-55, because they did not routinely access radiological control areas. However, beginning 2 years ago (approximate 2007), guards were newly assigned to TA-55 security areas that actually involved radiological control areas; therefore, since then, they have been routinely bioassayed.

To further investigate the bioassay pattern for personnel that may have had security-related duties, SC&A analyzed the bioassay records for Cases H, J, O, S, U, and Y (as listed in Table 4 above) in more detail. The results are summarized in Table 5.

Table 5. Bioassay Pattern of Six Security-related Personnel from Previous 30 Cases

Case*	Position Title	# of Years Worked 1976–2005	# of Years Bioassayed†	Routine/Yearly Bioassays Indicated?	Special Bioassays Indicated?	In-vitro?	In-vivo?
H	Security inspector, Firefighter	14	11	Yes	No	Yes	No
J	Guard	12	8	Yes	No	Yes	Yes
N	Uranium custodian	16	8	Yes	No	Yes	Yes
O	Security guard	18	9	Yes	No	Yes	Yes
S	Equip operator, security inspector	18	6	Yes	Yes	Yes	Yes
Y	Custodian, Security escort	25	1	No	Yes	Yes	No

* These correspond to the randomly assigned case designations used in the previous table.

† Urinalysis and/or WBC and/or chest ct. Some had multiply bioassays per year. Major radionuclides analyzed for include Pu-238/239, Am-241, H-3, & U; most results <MDA, a few were around the MDA values or positive.

The results of the analysis of these six cases show that for this sampling, most security-related personnel were routinely bioassayed for extended periods. The majority of the bioassay results were below the minimum detection activity (MDA), indicating that precautionary routine bioassays were taken when exposure potential may have been present. However, SC&A further

analyzed this bioassay data on a yearly basis and found that the majority of the bioassays were performed prior to 1995, as illustrated in Figure 3 below.

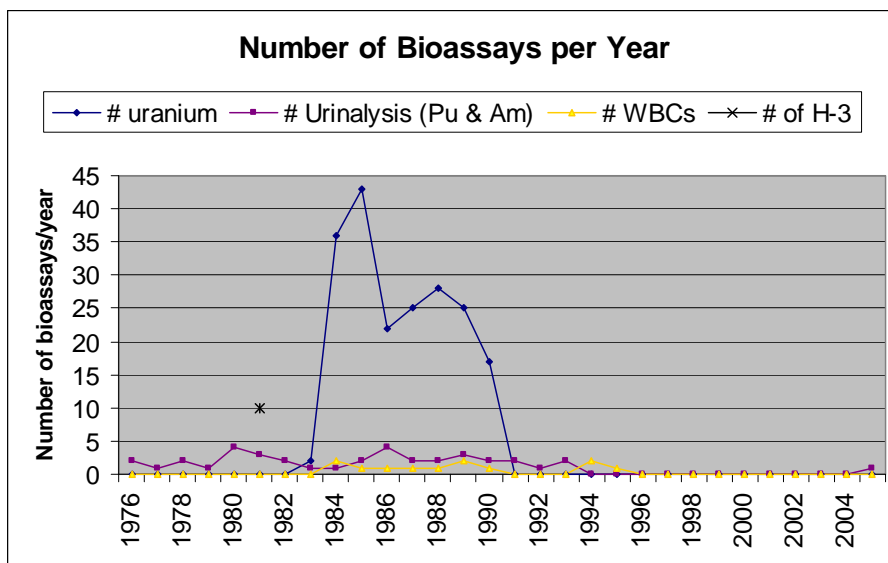


Figure 3. Number of Bioassays per Year for Six Security Workers

SC&A also observed this same trend of a decrease in the number of bioassays after the mid-1990s for the other LANL workers in the 30-case study as well.

Summary

From the information SC&A analyzed, security, service support, craft, technical, and staff workers were monitored using similar procedures that were determined by the potential for radiation exposure or intake of radionuclides, as opposed to job title or employer. In general, the number of bioassays started to decrease in the mid-1990s at LANL, which could create dose reconstruction problems for non-bioassayed workers; especially for workers that frequented many different areas within the lab complex, but were not routinely bioassayed.

Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 47 of 53
----------------------------------	---------------------------	-------------------------------------	----------------------

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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 48 of 53
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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 49 of 53
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Effective Date: April 8, 2010	Revision No. 0 – DRAFT	Document No. SCA-TR-SEC2010-0004	Page No. 50 of 53
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ATTACHMENT A: SOURCES AND TIME PERIODS OF LANL BIOASSAY REPOSITORY (ORAUT-OTIB-0063)

In-vivo (IVML) = <i>Historical DB</i> (pos only, no MDA) and <i>Existing DB</i> (some MDAs) = 1960s-2003 & 2003-present => LANL Bioassay Repository												
Table 4-1: Radionuclide total records and time period in both DBs; main analyte: (1960s-present)												
		AM-241	44.6k	1969-2009								
		Pu-239	43.6k	1969-2009								
		Th-234	5.7k	2004-2009								
		U-235	5.7k	1975-2009								
		MF&AP*	~9k	1969-2009								
		*Mix fission & activation products.										
In-vitro = <i>Old Elec DB</i> (H-3, U, & Po) + <i>Lab NB</i> + <i>Elec DB</i> (1944-2000 Am & Pu) + <i>BEST DB</i> (~1991-present; Pu, Am, H-3, & U) => LANL Bioassay Repository												
Old Electronic DB = TUPo = DEC-VAX => Interim DB => LANL Bioassay Repository (contains the info listed divided into 1944-1991 & 1991-present)												
Table 5-2 Total records, Time period, and Validation:												
		H-3	49.2k	1950-1990	70% in Lab NBs, 10% validated => 45 data changes (0.9%).							
		U-235	43.5k	1955-1990	93% in Lab NBs, 10% validated => 32 data changes (0.7%).							
		U-238	46.7k	1949-1990	93% in Lab NBs, 10% validated => 60 data changes (1.2%).							
		Po	5.2k	1947-1965	99% in Lab NBs, 39% validated => 72 data changes (3.2%).							
		Lab NBs = LANL notebooks										
Electronic DB contained Total records and Time period (pg 11):												
		Am-241	58	1975-2000								
		Pu-238	47.3k	1967-2000								
		Pu-239	91.4k	1944-2000								
BEST DB Table 5-20 contained Total records and Time period:												
		Am-241	2.6k	1973-2009								
		H-3	28.2k	1991-2009								
		Po-210	69	2007-2008								
		Pu-238	57.2k	1973-2009								
		Pu-239	77.6k	1973-2009								
		Pu-239+240	13.5k	1996-2009								
		Pu-240	12.5k	1997-2009								
		Th	13	2005-2008								
		U-234	17.6k	1991-2009								
		U-235	17.6k	1991-2009								
		U-238	17.6k	1991-2009								
		No Sr-90 monitoring program										
1944								1990	1991	2000	2001	present
<-----	25% to 90% verified & validated (V&V), varies by nuclide, in LANL Bioassay Repository*							----->	85% V&V*		~100% V&V*	
*As per HSR-4 Bioassay Data Repository Project document accompanying claimant's DOE files.												

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