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ADVISORY BOARD ON
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National Institute for Occupational Safety and Health

SC&A'S Evaluation of RPRT-0090, "Monitoring Feasibility Evaluation for Exotic Radionuclides Produced by the Oak Ridge National Laboratory Isotopes Division," Revision 00

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ABBREVIATIONS AND ACRONYMS

Advisory Board or ABRWH	Advisory Board on Radiation and Worker Health
Am	americium
Au	gold
Be	beryllium
Bk	berkelium
BNWL	Battelle Northwest Laboratory
C	confinement factor
Cd	cadmium
Cf	californium
CFR	*Code of Federal Regulations*
Ci	curie
Cm	curium
Cr	chromium
Cs	cesium
CTW	construction trade workers
Cu	copper
D	dispersibility
DCF	dose conversion factor
D&D	decontamination and decommissioning
DOE	U.S. Department of Energy
dpm	disintegrations per minute.
DR	dose reconstruction
ER	petition evaluation report
Fe	iron
FPPP	Fission Product Pilot Plant
GA0 or GU0	gross alpha in urine sample
GB0	gross beta in urine sample
GF0	gross alpha in fecal sample
HFIR	High Flux Isotope Facility
Hg	mercury

HP	health physics
HTO	tritiated water
I	iodine
Ir	iridium
LANL	Los Alamos National Laboratory
LBNL	Lawrence Berkeley National Laboratory
MAP	mixed activation product
MDA	minimum detectable activity
MFP	mixed fission product
mCi	millicurie
µCi	microcurie
MMES	Martin Marietta Energy Systems
Mo	molybdenum
mrem	millirem
NaI(Tl)	sodium iodide (thallium)
NIOSH	National Institute for Occupational Safety and Health
NOCTS	NIOSH OCAS Claims Tracking System
OCAS	Office of Compensation Analysis and Support
ORAUT	Oak Ridge Associated Universities Team
ORNL	Oak Ridge National Laboratory
Os	osmium
pCi/d	picocurie per day
Pd	palladium
Pm	promethium
PNL	Pacific Northwest Laboratory
POC	probability of causation
Pt	platinum
Pu	plutonium
R	release fraction
RaLa	radioactive lanthanum
Ru	ruthenium
SEC	Special Exposure Cohort

Sr	strontium
SRDB	Site Research Database
TBD	technical basis document
Tc	technetium
Th	thorium
TP0	gross trans-plutonium alpha in urine sample
TF0	gross trans-plutonium alpha in fecal sample
U	uranium
WBC	whole body counter
yr	year

1 INTRODUCTION AND BACKGROUND

The National Institute for Occupational Safety and Health (NIOSH) evaluated the internal monitoring capability of Oak Ridge National Laboratory (ORNL) for radionuclides that were produced by the Isotopes Division (termed "exotic radionuclides") and its predecessors from 1955 to 1988 in ORAUT-RPRT-0090, Revision 00, *Monitoring Feasibility Evaluation for Exotic Radionuclides Produced by the Oak Ridge National Laboratory Isotopes Division* (2018; hereafter "RPRT-0090"). In RPRT-0090, NIOSH listed 213 radionuclides in Table 6-3, which was presented as the final inventory for the Isotopes Division for the period 1955–1988. Table 7-2 provided a detailed list of each of the 213 radionuclides and the years they were in inventory (representing potential exposure), along with monitoring capability and bioassay data availability. NIOSH found that ORNL had adequate monitoring capabilities for 179 of these 213 radionuclides. Attachment B of RPRT-0090 provided a brief summary of the decay characteristics and bioassay methods for each of these 179 radionuclides. Table 7-4 of RPRT-0090 summarized the 34 remaining radionuclides that needed additional evaluation. Five of these 34 radionuclides were addressed in Attachment C of RPRT-0090 concerning radioiodine. Plutonium-241 was removed from the list of consideration because it was located at the Y-12 Plant (now Y-12 National Security Complex). In April 2018, the Advisory Board on Radiation and Worker Health (Advisory Board) tasked SC&A to evaluate RPRT-0090.

2 RPRT-0090 FEASIBLITY EVALUATION: BASIS FOR REVIEW

Demonstrating "feasibility" is central to petition evaluation under the Special Exposure Cohort (SEC) rule and process and requires that NIOSH determine whether it is "feasible to estimate the level of radiation doses of individual members of the class with sufficient accuracy" (42 CFR Part 83). To accomplish this determination, NIOSH would need "access to reliable information on the identity or set of possible identities and maximum quantity of each radionuclide (the radioactive source material) to which members of the class were potentially exposed without adequate protection" (42 CFR Part 83).

In RPRT-0090, NIOSH has identified each nuclide produced by the ORNL Isotopes Division within the time period in question (1955–1988) and has derived an estimated maximum annual inventory for each radionuclide. A "feasibility evaluation" was performed for each of the 213 radionuclides identified in the inventory for 1955–1988, as reflected in Table 7-2, by matching an "adequate monitoring method" for each radionuclide for each year for which an inventory for it was established (and assuming that the method was available thereafter) (NIOSH 2018a). For the 28 radionuclides without an applicable monitoring method, NIOSH conducted a dosimetric analysis combining the maximum annual inventory for each radionuclide with a maximum organ dose conversion factor (DCF) to estimate the "committed dose to the maximally exposed organ from inhalation of 1×10^{-5} of the total inventory"[1] (NIOSH 2018a, p. 40). NIOSH's analysis showed doses (50-year committed dose equivalent) from inhalation ranging from 0.3 millirem (mrem) to 1,464 mrem to the maximally exposed organ, from which it concluded that "the relatively low radiotoxicity of these same nuclides [the 28 without evidence of monitoring] in comparison with a bounding potential intake (Table 7-6) lends credence to the position that a significant intake of one of these nuclides would not be credible" (NIOSH 2018a, p. 43).

SC&A has reviewed RPRT-0090 for the general premise and scope of its "feasibility" evaluation, as described above, as well as in terms of the adequacy and completeness of its review of monitoring capabilities and operational inventories.

The following findings and observations need clarification or further substantiation in terms of actual dosimetric practice and how that translates to demonstrated feasibility to monitor the wide range of exotic radionuclides historically present in ORNL operations.

[1] SC&A's evaluation of the use of the 1×10^{-5} factor is provided in Appendix 4 of this report.

3 SC&A'S REVIEW OF SCOPE OF EXOTIC RADIONUCLIDE SOURCE TERMS

The petition evaluation report (ER) for ORNL (X-10), SEC-00189, reserved "the radionuclides created by cyclotrons, accelerators, and reactors for this evaluation report due to the complexity and interdependency involved in transferring materials between two DOE-covered facilities (X-10 and Y-12)" (NIOSH 2012, p. 40). It is not clear from RPRT-0090 whether its scope represents part or all of the reserved radionuclides that are to be evaluated. This needed clarification has three components, as follows.

First, the residual and the decontamination and decommissioning (D&D) periods for specific facilities at ORNL involved handling the cleanup of exotic radioisotopes, as well any post-operational waste management associated with them. The extensive accelerator and reactor operations at ORNL would have involved a broad spectrum of mixed activation products (MAPs) and mixed fission products (MFPs) (according to the ER: "Many facilities have exhibited fission and activation product exposure potential" [NIOSH 2012, p. 46]), not all of which would have been part of the Isotopes Division inventory that NIOSH addressed. While RPRT-0090 addresses what has been identified as being handled by the Isotopes Division, this inventory would likely need to be expanded to accommodate operational source terms, including D&D and waste management, for these other activities.

This is not an insignificant distinction. Of the 11 cases where internal exposure guides were exceeded at ORNL in 1964–1967, six cases involved iodine-131 (I-131) uptakes from decontamination work, four cases involved tritium uptakes due to target preparation and decontamination work, and one case involved a strontium-90 (Sr-90) uptake due to work at a solid waste disposal burial ground (MacPherson 1968). It can be assumed that the number of intakes that did not happen to exceed exposure guides would have been higher (that information was not readily available) and exotic radionuclides would likely have been associated with some of these activities.

Second, the processing of radioisotopes from beginning to end involved workplace emissions and waste streams, as well as byproduct materials, that may have included quantities of the exotic radioisotopes in question. Are these also reflected in the inventory NIOSH used in RPRT-0090, or is that inventory simply the "finished product" inventory of isotopes produced for transfer? That former amount would obviously be larger than the latter, and such a discrepancy would directly influence the basis for determining the "maximum annual inventory" in Table 7-6, from which a justification is made for 28 radionuclides not having a "bounding potential intake" of concern.

Third, construction trade workers (CTWs), maintenance personnel, and other support personnel (such as health physics [HP] technicians) moved around the ORNL site and were potentially exposed to a broad range of the exotic radionuclides in question, even beyond those generated by the Isotopes Division. This fluidity of work assignment is captured in an interview with a maintenance engineer:

> *Please understand that we had central maintenance shops that were responsible for maintenance all over ORNL. It would not be correct to assume that because a*

> *worker was not assigned to a certain building that he or she did not actually do work in that particular building. The issue of overtime must also be considered. Workers from all over the plant site are asked to work overtime on any particular job. A worker that would normally work in a clean area could in fact receive an exposure on an overtime job.* [Bancord 2004]

This raises three questions. First, was bioassay monitoring for CTWs, maintenance workers, and other personnel implemented in the same manner as operating personnel? In other words, did area health physicists direct nonroutine bioassays based on the same subjective exposure criteria and were such exposures (e.g., due to contamination) identified in the same manner as with operational staff? Second, do records exist to document how they were actually monitored during the 1955–1988 time period? Third, what radionuclides would these worker categories have been exposed to (given their laboratory-wide work locations), and how does that compare with Table 7-2 in terms of the adequacy of bioassay monitoring?

Finding 1: Scope of RPRT-0090 needs to be clearly defined.
SC&A finds that the scope of RPRT-0090 needs to be clarified in terms of whether (and how) it is meant to encompass the "reserved" portion of the ER for "cyclotrons, accelerators, and reactors" and whether NIOSH intends to address the full scope of radionuclides involved in waste management (including D&D), site-wide construction, and maintenance.

4 SC&A'S EVALUATION OF RADIONUCLIDE INVENTORY

SC&A compared several of the radionuclide initial dates listed in Table 7-2 of RPRT-0090 to the NIOSH-supplied Microsoft Excel *X-10 Exotics Workbook_022015 kwv* under the tab "Master Summary Data" (NIOSH 2015; hereafter "X-10 Inventory"). While some of the early dates matched, some did not. For example, Table 7-2, page 31, of RPRT-0090 shows that curium-244 (Cm-244) was present beginning in1962; however, the NIOSH X-10 Inventory spreadsheet, Column AA, Row 72, lists the first inventory of Cm-244 as 1964. Similar discrepancies were found for berkelium-249 (Bk-249), Cm-242, plutonium-238 (Pu-238), Pu-239, and uranium-232 (U-232) in the early years.

SC&A compared Table 7-2 of RPRT-0090 with the contents of Table 5-15 of the ORNL technical basis document (TBD), ORAUT-TKBS-0012-5, Revision 02, *Oak Ridge National – Occupational Internal Dose* (NOISH 2013a) and found several radionuclides listed in Table 5-15 for years that were listed as "N" (meaning no radionuclide present in inventory in the specified year) in Table 7-2. For example, protactinium-233 is listed with a 1961 maximum measured in vivo activity of 2,800 nanocuries in ORAUT-TKBS-0012-5, Revision 02, but has a note of "N" in Table 7-2 of RPRT-0090.

Observation 1: Inventory discrepancy.
A sampling of some of the inventory of the radionuclides for the early years indicated some discrepancies in inventory between Table 7-2 in RPRT-0090 and NIOSH's X-10 Inventory spreadsheet.

SC&A also compared the radionuclide listings in Table 7-2 of RPRT-0090 with various ORNL records, including a sampling of customer shipment listings (ORNL 1957, ORNL 1965, BNWL 1977, PNL 1984) and an in-house isotope inventory (Kohring 1990).[2] Table 7-2 of RPR-0090 was derived using source documents from the Site Research Database (SRDB), including isotope shipping and sales reports and operational and technical reports, as well as target rupture records (for sealed shipping containers for which leakage of radionuclides may have presented an exposure source). SC&A's comparison, based on similar source documents, identified the following radioisotopes as generated at ORNL but not included in the NIOSH inventory in RPRT-0090.

Table 1. Examples of Radionuclides Not Included in RPRT-0090

ORNL 1957	ORNL 1965	BNWL 1977	PNL 1984	Kohring 1990
Europium-154 Iron-55, -59	Lutetium-174 Iodine-128 Europium-149 Europium-154 Thulium-168	Europium-154	Aluminum-26	Clorine-34 Manganese-57 Iodine-128 Europium-154

[2] Kohring 1990 provided a listing for 1989. While it was dated shortly after the Dec 1988 cutoff, SC&A considered it illustrative of radionuclides that may have been shipped in the late 1980s (no other inventories were found for fiscal year 1987–1988).

As these were radioisotopes for offsite shipment, it is unclear whether these particular radionuclides were shipped directly off site for processing and were, therefore, not handled in an unsealed form at ORNL (and accordingly, excluded from the inventory). If they were shipped in a sealed container off site, the only remaining question would be whether any record of target rupture exists (which would lead to inclusion in Table 6-2 of RPRT-0090). Clearly, isotopic variants of certain radionuclides, such as euriopium-154, would likely have the same radiochemical properties as other isotopes of the same radionuclide; this listing is provided as a means of verifying completeness.

In reviewing the completeness of the Table 7-2 radioisotope listing, SC&A also reviewed the corresponding ORNL facilities involved with their generation. One ORNL report, *Oak Ridge National Laboratory Isotopes Facilities Shutdown Program Management Plan* (ORNL 1992, p. 2), noted that:

> *The goal of the shutdown program* [was] *to place 16 formerly utilized isotopes facilities at ORNL…in a radiologically and industrially safe condition for routine, long-term maintenance and surveillance prior to eventual decommissioning.*

The additional facilities identified by SC&A in this review that handled such radioisotopes, but were not listed among the 10 buildings cited by RPRT-0090 (and not including subparts of those facilities, e.g., 3038-M and 3038-E), were:

- Krypton-85 Enrichment Facility — Building 3026-C
- Radioisotope Production Laboratory-H — Building 3118
- Tritium Target Preparation Facility — Building 7025

Building 3026-C was the Krypton-85 Enrichment Facility, and handled a mixture of krypton gases of different isotope masses, with receipt of tritium-filled tubes from Building 3033 (Patton 1988). Building 3118 was constructed in the early 1960s to enclose access to the rear entry area for the hot cells in Buildings 3030 and 3031 and provided a storage area for "drums and containers of hazardous and radiological waste, radioactive shielding materials, and casks" (ORNL 1997a, p. 1). Building 7025 housed a tritium target fabrication system for preparing thin oxide targets for domestic and international customers, as well as tritium tritide targets and various metallurgical samples (ORNL 1997b).

Another ORNL facility handling exotic radioisotopes was Building 3515, the "Fission Product Pilot Plant" (FPPP)," one of the first facilities at ORNL to extract radioisotopes from liquid radioactive wastes. The pilot plant was initially constructed in 1948 and modified in the early 1950s with additional shielding and a hot cell. FPPP operations included "extracting radioactive isotopes from waste liquids from off-site and ORNL activities," and the "radionuclides removed included ruthenium, strontium, cesium, cerium, and others" (Mandry and Snedaker 1994, p. 2).

The basis for defining the ORNL radioisotope complex as the "10 buildings designed for processing, packaging, and shipping radioisotopes" is not clear in RPRT-0090 (p. 6). While these 10 are clearly key facilities that handled radioisotopes at ORNL, others had similar operations or were even co-located and associated with them. If the scope of RPRT-0090 is intended to evaluate "the monitoring capability of the ORNL HP program to have monitored all materials

that were produced and handled by the Isotopes Division regardless of production location" (RPRT-0090, p. 6), then other such facilities and associated radionuclides should be reviewed for inclusion.

Finding 2: Incomplete radionuclide and radioisotope facility inventory.
A sampling of the radionuclides listed in Table 7-2 found a few missing when compared with operational and customer records. Likewise, a few ORNL facilities that historically handled radioisotopes are also not included in those cited and addressed in RPRT-0090. Given the operational diversity of ORNL accelerator and reactor operations, consideration should be given to an inventory scope that encompasses isotopic source terms broader than that of the Isotope Division.

5 SC&A'S EVALUATION OF ORAUT-RPRT-0090 WITH RESPECT TO *IN VITRO* METHODS

5.1 SC&A'S EVALUATION OF MONITORING CAPABILITY AND BIOASSAY DATA FOR 179 RADIONUCLIDES

SC&A selected 12 of the 179 radionuclides to evaluate monitoring capabilities and bioassay data. These 12 radionuclides were selected because of the relatively large quantities of the radionuclides in inventory for a number of years and their radiotoxicity, which could result in potentially significant dosimetric intakes. This mixture included both routine and exotic radionuclides. The 12 radionuclides selected were americium-241 (Am-241), Bk-249, californium-252 (Cf-252), Cm-242, Cm-244, promethium-147 (Pm-147), Pu-238, Pu-239, ruthenium-106 (Ru-106), Sr-90, thorium-230 (Th-230), and U-232.

For each of the 12 radionuclides SC&A evaluated, the available bioassay data for the years the radionuclides were listed in the inventory. SC&A selected bioassay data (as opposed to other search criteria) for evaluation because if bioassay data are available, the procedures, bioassays, and records were available and functional for dose reconstruction (DR) purposes. For this evaluation, SC&A used the following sources of information:

- **Copies of original bioassay records:** NIOSH Office of Compensation Analysis and Support (OCAS) Claims Tracking System (NOCTS).
- **List of Code 000 data in NOCTS:** NIOSH's Excel spreadsheet *Type0_archiveR1* (NIOSH 2018b).
- **X-10 bioassay database:** NIOSH's Microsoft Access database, *niosh_04282013*, tab "VPA ornl_invitro" (NIOSH 2013b), which contains 94,988 urine and fecal bioassay records for the period 1955–1988 (hereafter "X-10 Database").
- **Radionuclide inventory:** SC&A considered the radionuclide inventory information in NIOSH's X-10 Inventory spreadsheet to be the correct inventory list when performing comparisons for this evaluation.

For the 12 radionuclides under evaluation, SC&A compared the information in Table 7-2 of RPRT-0090 (using the bioassay codes listed in Table 7-2 for each of the radionuclides) to the bioassay data in the NIOSH's X-10 Database and NIOSH's X-10 Inventory. Examples of SC&A's summary of the evaluation for Cm-244 and Ru-106 are shown in Appendix A of this report. Table 2 summarizes the results for the 12 radionuclides evaluated. Table 7-2 in RPRT-0090 used a green cell color and "G" to indicate that a radionuclide was present in inventory, a bioassay method was available, and sample results were available for that year. It used a yellow cell color and "Y" to indicate that a radionuclide was present in inventory and a bioassay method was available, but no sample results were available for that year.

Table 2. SC&A's Evaluation of 12 Radionuclides from Table 7-2 of RPRT-0090

Radioisotope	Major Radiation Emitted	Table 7-2 Bioassay Codes for Specific Radioisotope	Years Table 7-2 Listed Green (G) when Bioassay Data Were Not Present but Radioisotope Was in X-10 Inventory [a]	Years Table 7-2 Listed Yellow (Y) or Green (G) when the Radioisotope Was Not in X-10 Inventory	Contents of Table 7-2 Compared to Supporting Data
Am-241	Alpha	000(Am-241), AM0, GU0, TP0	None	None	Complete
Bk-249	Alpha	000(Bk-249), TP0	None	1964	Mostly complete
Cf-252	Alpha	TP0	None	1962	Mostly complete
Cm-242	Alpha	000(Cm-242), TP0	None	1961, 1962 & 1964	Fairly complete
Cm-244	Alpha	000(Cm-244), TP0, CM0	None	1962 & 1963	Fairly complete
Pm-147	Beta	000(Pm-147), 013, PM7, FU0, Pm-147	[a](1956 & 1957)	None	Fairly complete
Pu-238	Alpha	GU0, PU0	None	1961 & 1962	Fairly complete
Pu-239	Alpha	GU0, PU9, PU0	None	1961 & 1962	Fairly complete
Ru-106	Beta	000(Ru-106), 013, GB0, RU6	(1956, 1957, 1959) [a], 1975, 1978, 1986, 1987, & 1988	None	Incomplete
Sr-90	Beta	SR0	None	None	Complete
Th-230	Alpha	GF0, TF0, Th-230	None	1962 & 1963	Fairly complete
U-232	Alpha	UR0	None	None	Complete

[a] Lack of beta records in the X-10 Database for 1955–1959 may be because they were not entered into the X-10 Database; however, they could be contained in the NOCTS files, as discussed on page 15 of RPRT-0090.

5.1.1 Gross Alpha Counting

SC&A found that, in general, the pre-1965 records for the most prevalent alpha-emitting radionuclides (such as plutonium, thorium, and uranium) indicated the specific element assayed (e.g., plutonium). For the trans-plutonium elements, such as Am-241, Cf-252, etc., gross alpha in urine or fecal samples were used (bioassay codes GA0, GU0, TP0, GF0, or TF0). In the 1950s, some specific radionuclides were listed on the bioassay card; however, it became more prevalent in approximately 1965 for the specific radionuclide analyzed (such as Am-241) to be specified in the bioassay records. Most of the ORNL original bioassay cards available in NOCTS provide detailed analytical information, including chemical process yield, counter efficiency, etc., with specific radionuclides listed as far back as 1955. Some of these are illustrated in the examples in Appendix B of this report. SC&A found that there were a large number of bioassay records for each year for the period 1955–1988 containing alpha counting results (coded GA0, GU0, TP0, GF0, or TF0) in the X-10 Database from the 94,988 urine and fecal bioassay records. An example from this database for Am-241 for a small part of 1955 and 1956 is shown in Appendix C of this report.

As can be seen from Table 2, the major alpha emitters of the 12 radionuclides evaluated appear to have been monitored as was listed in Table 7-2 of RPRT-0090, with the exception that in several instances Table 7-2 indicates that inventory was present when the inventory list does not indicate that the radionuclides were present during that year. Additionally, it has to be assumed that the dose reconstructor has sufficient information in the claimant's files to assign the correct radionuclide the worker was exposed to from gross alpha counts, such as gross alpha in urine (GA0, GU0, and TP0 bioassay codes), or gross alpha in fecal samples (GF0 and TF0 bioassay codes). The X-10 Database records do not always list the specific radionuclide analyzed, as the original ORNL bioassay cards often do.

Observation 2: Specific alpha-emitting radionuclide needs to Be identified for DR.
The specific radioisotope monitored is not always presented in NIOSH's X-10 Database as it generally is in the NOCTS files. Gross alpha results could be applied to many radionuclides. Is the information on the original bioassay cards available in the X-10 Database, and will the X-10 Database be used in DR or coworker model development?

Trans-plutonium Radionuclides

According to pages 19 and 20 of ORAUT-TKBS-0012-5, Revision 02, bioassay results for plutonium are to be assigned as Pu-239 intakes, and bioassay results for trans-plutonium radionuclides (using such methods as gross alpha counting) are to be assigned as Am-241 intakes for DR purposes. In general, this is an acceptable DR method; however, with the increased potential for concentrating exotic radionuclides at the ORNL Isotopes Division, which may contain such radionuclides as Bk-249, Cf-252, etc., this method may, or may not, be applicable.

Observation 3: Trans-plutonium radionuclides may need further analyses.
SC&A is concerned that assigning trans-plutonium gross alpha counting results as Am-241 intakes without consideration of other potential trivalent alpha-emitting actinides (such as Bk-249, Cf-252, Cm-242, Cm-244, etc.) and their individual radiotoxicity could result in underestimating the internal dose. It could be beneficial to determine if assigning the intake as Am-241 is claimant favorable, considering the exotic trans-plutonium radionuclides at ORNL.

5.1.2 Gross Beta and Gamma Counting

Gross beta counting was sometimes used for strontium, but the bioassay code SR0 or SR9 was generally used to specify Sr-90 because of the prevalence of Sr-90, as far back as the 1950s. In addition to beta counting specifically for strontium, Table 4-1 and Table 4-2 of RPRT-0090 indicate that ORNL used gross beta counting (153 samples with bioassay code 013 and 26 with bioassay code GBO), gross gamma counting (9 with bioassay code GGO), fission products (17 with bioassay code FP0), and rare earths (1,333 with bioassay code FU0 and 70 with RF0). While these are small numbers of bioassays compared to the total of 91,867 coded bioassays in Table 4-1, some of these gross counting codes are listed as bioassay codes for many radionuclides in Table 7-2 of RPRT-0090. These gross counting methods are briefly discussed in Attachment A of RPRT-0090, but no specific counting efficiencies, correction factors, etc., are provided. Several SRBD references provide detailed procedures for the radiochemistry and gross beta counting of bioassay samples, but they do not include any specific radionuclide-efficiency

calibration information (e.g., Henley 1968, PDF page 39, and Henley 1978, PDF page 23). Henley 1968 states on PDF page 38:

ANALYSIS FOR BETA EMITTERS

> *The object of a mixed beta count is to estimate the beta activity in the urine exclusive of the naturally occurring potassium-40. This is largely accomplished by using two group separations and counting the product of each.*
>
> *Group #1 includes the ions precipitated by or absorbed in a calcium phosphate precipitate. These include the alkaline earths, the lanthanides, the actinides and most other ions with strong tendencies for hydrolysis.*
>
> *Group #2 includes the elements which can be precipitated from a dilute acid solution as the sulfide. Cu, Mo, Tc, Ru, Pd, Cd, Os, Ir, Pt, Au and Hg are precipitated directly from urine in this manner. Alkali metals and anion activities are not carried by this precipitation.*

As can be seen from this description, there could be a variety of combinations of radionuclides, or a single radionuclide, contributing to the gross beta counts. Attachment B of RPRT-0090 provides a summary of the decay emissions from the radionuclides at ORNL, which indicate a wide range of beta energies and percent of emission (abundance) for the many radionuclides that could be analyzed by beta counting. This can present problems when assigning gross beta count data to a specific radionuclide. The use of data obtained from gross beta counting of bioassay samples to project specific radionuclide intakes (from an assortment of potential radionuclides) presents potential issues as outlined in SCA's recent evaluation of a NIOSH white paper on Lawrence Berkeley National Laboratory (SC&A 2018). In that report, SC&A also analyzed issues with applying gross alpha counting data, such as from air samples, to project biological intakes. However, the issues for alpha emitters are not as prevalent for ORNL bioassays because of the use of defined urine and fecal counting and the fact the most dosimetric-important alpha emitters listed in Attachment B of RPRT-0090 are similar in energy and abundance. This is not true for beta emitters listed in Attachment B of RPRT-0090, which are not all similar in energy and abundance. For example, Attachment B of RPRT-0090, page 68, shows that copper-67 (Cu-67) beta energy and abundance is quite different than those of Ru-97 on page 94; however, both could be present in the Group #2 radionuclides as stated in the Henley 1968 quote above. Additionally, among radionuclides of the same element, such as silver-110m and silver-111 as shown on pages 96 and 97 of RPRT-0090, the beta energies and abundance can be very different. A similar analogy applies to gross gamma count data.

SC&A's evaluation of the three beta-emitting radioisotopes Pm-147, Ru-106, and Sr-90 is summarized in Table 2 of this report. Table 2 indicates the following:

- **Pm-147** – The data in Table 7-2 of RPRT-0090 for Pm-147 are in good agreement with the supporting data in the X-10 Database and the X-10 Inventory, considering that the 1956 and 1957 beta results may not have been recorded in the X-10 Database.
- **Ru-106** – The data in Table 7-2 of RPRT-0090 for Ru-106 are not in good agreement with the supporting data in the X-10 Database and the X-10 Inventory, even when

considering that the 1956, 1957, and 1959 beta results may not have been recorded in the X-10 Database. SC&A could not locate any appropriate beta counting data for the years 1975, 1978, 1986, 1987, and 1988, although Table 7-2 of RPRT-0090 has those years colored in green indicating that bioassay data were available for those years.

- **Sr-90** – The data in Table 7-2 of RPRT-0090 for Sr-90 are in good agreement with the supporting data in the X-10 Database and the X-10 Inventory.

Observation 4: Use of gross beta or gamma count data could result in underestimate of assigned dose.
Using gross beta or gamma count data without knowledge of the radionuclide the counter was calibrated with and the radionuclides in the bioassay sample could result in assigning the incorrect radionuclide and radioactivity content because of different counting efficiencies for the different energy of beta particles and gamma photons. Has this issue been addressed for DR for ORNL claimants? Additionally, bioassay data for at least one beta-emitting radionuclide (Ru-106) could not be located for several years that Table 7-2 indicated it was available.

5.1.3 SC&A's Evaluation of RPRT-0090 Attachment A

The Attachment A references upon which the *in vitro* radionuclide monitoring capability cited in Table 7-2 are based come down to essentially two key documents: Henley 1968 and Henley 1978. Both of these comprise radiochemical "recipes" for processing gross alpha, gross beta, rare earths, etc. Whether and how they (and the other methods cited) were actually used onsite based on field procedures, available dose records, and interviews, is not discussed in RPRT-0090.

SC&A believes that how the identification and quantification of "associated" radionuclides was performed by ORNL, in practice, should be validated. The isotope-specific monitoring capability analysis in RPRT-0090 carries with it certain inherent assumptions not addressed in the report, including necessary identification of associated radionuclides through process knowledge and quantification by radiochemical assays.

The TBD for ORNL occupational internal dose, ORAUT-TKBS-0012-5, Revision 02 (NIOSH 2013a), observes the following (p. 10):

> *Before that time* [1989], *chemical methods were used to separate radioelements as well as practicable, and the materials were assayed in terms of total activity. The measured activity would later be assigned to a predominant nuclide. Therefore, a result from the early years might indicate* ^{90}Sr, *when in reality it includes* ^{89}Sr. *The same is true for early plutonium results and results for transuranic materials. Therefore, "associated" radionuclides are inherently included in such results. Process knowledge of radionuclides present in various work areas was used to assign nuclides to sample results.*

What is clear here is that the detailed matrix covering 213 specific radionuclides is dependent on a much smaller set of gross alpha and beta/gamma analytic procedures, and an uncertain set (see later comments) of *in vivo* monitoring protocols, all of which are, in turn, dependent on the extent to which area health physicists were able to identify the scope of radionuclides involved based on process knowledge, and whether and how radiochemical assays were performed to

"separate radioelements" and to ascertain how much intake activity to assign a "predominant" nuclide.

In reviewing RPRT-0090, as well as the ER and internal dose TBD, SC&A has not found any validation of how ORNL actually implemented the above assays and process reviews such that assurance can be given that dose-contributing nuclides were identified and nuclide-specific dose contributions were adequately estimated within the constraints of the technological capability of the time. If "associated" radionuclides were not adequately addressed in bioassay sample results, the credit taken for that capability in Table 7-2, and the results themselves, may not represent internal dose in an adequate and sufficiently accurate manner.

In broader terms, a "bridge" between ORNL written procedures, monitoring requirements, and capabilities for radionuclide identification, and the recorded bioassay data available for DR, is not clear at this time. As SC&A has found in the past, program directives and monitoring capabilities do not necessarily equate to what was actually put into practice and how complete the data are for DR. For example, a 1988 safety appraisal of ORNL's High Flux Isotope Facility (HFIR) found that "implementing or operating procedures have not been developed for internal dosimetry. These procedures should be developed to address the day-to-day activities of the internal dosimetry function" (MMES 1988, PDF p. 41). In terms of carrying out internal dosimetry procedures, this same 1988 safety appraisal found that "frequency schedules have been established and are documented in Appendix 12 of the Health Physics Manual; however, the established frequencies for whole-body counting of HFIR personnel are not followed" (MMES 1988, PDF p. 39).

It is also not clear whether some of the 28 radionuclides for which matching monitoring capability has not been verified would have required whole-body counting coverage for adequate detection and dose assessment. If so, technological and programmatic limitations (including higher minimum detectable activity [MDA] benchmarks) in the earlier years may have precluded such coverage, bringing into question ORNL's "ability to develop specialized bioassay methods as needed" (RPRT-0090, page 43). Actual radionuclide-specific *in vivo* monitoring capabilities need to be verified to establish feasibility.

If, as indicated in the ORNL site profile (NIOSH 2013a), there were no routine radionuclide-specific *in vitro* or *in vivo* bioassays before 1989, how does NIOSH intend to bound routine doses when (1) a significant portion of the ORNL workforce was mobile, (2) radiological source terms varied widely from operation to operation (and included exotics, MAPs, and MFPs), and (3) various area HP technicians made individual judgments on who would be bioassayed for cause? Under such circumstances, the "availability" of monitoring technology or procedure would almost be a secondary consideration for reviewing feasibility.

Finding 3: Attachment A *in vitro* bioassay methods lack information about actual implementation.

In vitro *bioassay methods are outlined in Attachment A, but it does not include any discussion or references regarding their actual field implementation. The exclusion of comparable* in vivo *monitoring methods makes a review of ORNL monitoring capability incomplete.*

5.2 SC&A's Evaluation of Feasibility of DR for 28 Radionuclides

Section 7.2 of RPRT-0090 discussed the 28 radionuclides that were identified as needing additional consideration because of the lack of defined bioassay methods, and some of their characteristics are listed in Tables 7-4, 7-5, and 7-6 of RPRT-0090. To evaluate the significance of the 28 radionuclides listed in Table 7-6, SC&A ran the Integrated Modules for Bioassay Analysis program using a factor of 1E-5 times the maximum annual inventory as listed in column 2 of the table for several of the radionuclides and found that the resulting organ doses were similar to the doses listed in column 5 of the table, for solubility Type M; other solubility types could produce different doses. SC&A's detailed analyses of using a factor of 1E-5 is in Appendix D of this report.

SC&A then compared the maximum annual inventory value for each radionuclide, as listed in column 2 of Table 7-6, to the inventory value in NIOSH's X-10 Inventory for each year there was a red symbol in Table 7-2 for that radionuclide. SC&A found a few discrepancies in the annual inventory values, but, in general, there was reasonable agreement. However, the dose values still indicate that some of radionuclides may be dosimetrically significant. Table 3 provides a summary of Table 7-6 of RPRT-0090 to include the SC&A radionuclide inventory analyses.

Table 3. Dosimetric Analysis of 28 Radionuclides

Radionuclide	RPRT-0090 Maximum Annual Inventory (mCi)	RPRT-0090 Year of Maximum Inventory	RPRT-0090 Maximum Organ DCF (mrem/mCi)	RPRT-0090 Organ of Maximum Dose	RPRT-0090 Committed Dose Equivalent (mrem)	X-10 Inventory Maximum Annual Inventory (mCi)	X-10 Inventory Year of Maximum Inventory	Revised Committed Dose Equivalent (mrem)
Beryllium-7	340	1957	1,554	Extrathoracic	5	340	1957	5
Calcium-41	501	1986	9,250	Bone	46	501	1986	46
Chromium-51	46,225	1961	925	Extrathoracic	428	46,225	1961	428
Manganese-54	115	1957	27,010	Extrathoracic	31	115	1957	31
Iron-55	620	1955	27,750	Spleen	172	620	1955	172
Cobalt-57	175	1957	13,690	Lungs	24	175	1957	24
Gallium-67	120	1959	6,290	Extrathoracic	8	120	1959	8
Selenium-75	2,160	1955	27,750	Kidneys	599	2,160	1955	599
Strontium-85	142	VNL [a]	20,720	Extrathoracic	29	105	1957	21
Strontium-87m	84	1960	2,220	Extrathoracic	2	84	1960	2
Molybdenum-93	(d)	NA	103,600	Bone	(d)	39	1962	40
Ruthenium-103	1,020	1955	55,500	Lungs	566	1,020	1955	566
Palladium-103	(d)	NA	6,290	Extrathoracic	(d)	47.5	1962	3
Cadmium-109	64	1964	851,000	Kidneys	545	64	1964	545
Tin-113	610	1957	48,100	Lungs	293	610	1957	293
Tin-119m	3,598	1986	40,700	Lungs	1464	3,598	1986	1464
Tellurium-121	13	No data [b]	19,980	Extrathoracic	3	NA	No data	NA
Cesium-131	25	No data	2,775	Extrathoracic	1	NA	No data	NA

Radionuclide	RPRT-0090 Maximum Annual Inventory (mCi)	RPRT-0090 Year of Maximum Inventory	RPRT-0090 Maximum Organ DCF (mrem/mCi)	RPRT-0090 Organ of Maximum Dose	RPRT-0090 Committed Dose Equivalent (mrem)	X-10 Inventory Maximum Annual Inventory (mCi)	X-10 Inventory Year of Maximum Inventory	Revised Committed Dose Equivalent (mrem)
Barium-133	80	1961	35,520	Bone	28	80	1961	28
Cerium-139	(d)	NA [(c)]	37,000	Lungs	(d)	(d)	NA	(d)
Promethium-145	32	Not listed	151,700	Bone	49	NA	NA	NA
Terbium-156	(d)	NA	51,800	Extrathoracic	(d)	(d)	NA	(d)
Dysprosium-159	60	1961	9,620	Bone	6	60	1961	6
Tungsten-181	18	No data	1,628	Extrathoracic	0.3	NA	No data	NA
Osmium-185	(d)	NA	26,270	Extrathoracic	(d)	(d)	NA	(d)
Gold-195	5.6	1961	30,710	Lungs	2	5.6	1961	2
Mercury-197	542	1957	133,200	Lungs	722	542	1957	722
Bismuth-206	(d)	NA	88,800	Extrathoracic	(d)	(d)	NA	(d)

[(a)] VNL denotes that this value was not listed in the X-10 Inventory.
[(b)] No data for this radionuclide were listed in the X-10 Inventory.
[(c)] NA denotes not applicable to this radionuclide.
[(d)] No inventory data were available for this radionuclide, per RPRT-0090.

Observation 5: The results in Table 7-6 depend on inventory used.
As outlined in Observation 1, there appear to be some discrepancies in the inventory used by NIOSH compared to those provided to SC&A for evaluation of RPRT-0090. These discrepancies change a few of the results of Table 7-6, as illustrated in Table 3 of this report.

The feasibility of monitoring for the 28 radionuclide intakes for DR purposes was not definitively addressed in RPRT-0090 Section 7.2, or in the Summary in Section 8.0, which states (p. 43):

> *Evaluation of the ORNL bioassay program indicates the ability to develop specialized bioassay methods as needed to adapt to changing conditions and emergent events. While evidence has not been found for all of the 28 identified nuclides, it is clear that ORNL had the capability to develop methods as needed. In addition, the relatively low radiotoxicity of these same nuclides in comparison with a bounding potential intake (Table 7-6) lends credence to the position that a significant intake of one of these nuclides would not be credible.*

The presence of the capability to develop monitoring does not necessarily mean that it was implemented, especially since bioassay records do not exist to show that it was used. The dosimetric significance of the 28 radionuclide depends on the inventory quantity and the inhalation factor used, as illustrated in Table 3 of this report.

5.2.1 Implications of Lack of Routine Bioassay Program

Moreover, SC&A questions whether the lack of routine bioassays until the late 1980s may take precedence over "monitoring capability" as addressed in RPRT-0090. ORNL did not routinely bioassay its workers for day-to-day potential intakes "in the early years," and isotope-specific

analyses for *in vitro* samples did not become routine until 1989. As noted in the TBD for occupational internal dose, ORAUT-TKBS-0012-5, Revision 02 (NIOSH 2013a, p. 12):

> *Urine samples were collected in the early years of the bioassay program based on the area health physicist's knowledge of field conditions (e.g., known spills or incidents, air and contamination sample results, etc.). This practice of scheduling did not use a specified sampling frequency…. A 1961 procedure manual…references procedures and practices governing the health physics program at that time, including internal and external exposure monitoring. Although referenced, the procedure detailing internal dosimetry was not among the documents available for review.*

Regarding whole body counting frequencies, this same reference noted that:

> *Historical information of the* in vivo *monitoring program…indicates that the whole-body counting frequencies in Table 5-3 were not consistently followed. Discussions with previous site personnel indicate that no formal counting frequency was used at ORNL until the later 1980s.*

In other words, no routine whole-body counting was performed at ORNL until after the time period in question (1955–1988).

Although, an argument can be made for the availability of bioassay monitoring capability, as well as the ability to use nonroutine bioassay data to bound routine exposures, several concerns need to be addressed. First is the inventory completeness question previously raised; i.e., did the nonroutine bioassays performed encompass the full scope of radionuclides (including associated "exotic" radionuclides) for which an exposure potential existed? Second, did the area health physicists making judgments about which workers received bioassays treat CTWs and maintenance personnel the same as operations personnel for internal monitoring purposes? Third, how representative were the nonroutine bioassays in terms of potential exposures, as compared with other exposures experienced by the full range of workers at ORNL (i.e., including CTWs and maintenance personnel)? If intakes could have been missed at ORNL with this approach, as indicated by the then-lead ORNL internal dosimetrist (Berger 2004), what are the implications from an adequacy and completeness standpoint?

5.2.2 Implications of Significant vs. Non-negligible Intakes

The only apparent evidence for implementation of analytic procedures are actual bioassay results and dose records. For NIOSH to claim that "ORNL had the capability to develop methods as needed" (e.g., for the 28 identified nuclides lacking analytic protocols) is questionable in light of actual whole-body counting system limitations that existed in the 1960s–1980s. The qualifying comment in RPRT-0090, that "the relatively low radiotoxicity of these same nuclides in comparison with a bounding potential intake (Table 7-6) lends credence to the position that a significant intake of one of these nuclides would not be credible," appears to acknowledge the lack of information on such monitoring.

From this concluding statement of RPRT-0090, a key question is whether the lack of a "significant intake" is an adequate basis for not considering a potential internal exposure in DR, as opposed to a determination that the potential for such an uptake could be considered "negligible." The question of whether a source term is negligible from an exposure potential standpoint, and the corresponding implications for DR under the Energy Employees Occupational Illness Compensation Program Act have been previously addressed by the Advisory Board; that precedent should provide guidance here.

Finding 4: Feasibility of monitoring 28 radionuclides not adequately addressed. *While the 28 radionuclides were discussed in Section 7.2 and some of their characteristics were listed in Tables 7-4, 7-5, and 7-6 of RPRT-0090, the feasibility of monitoring for intakes for DR purposes was not completely addressed, particularly given the lack of routine bioassays in the earlier years. Methods for accounting for the lack of monitoring of these radionuclides need to be addressed in more detail, and an acceptable resolution derived. SC&A finds that it is not possible at this time to validate implementation without further onsite review, including document review and interviews with health physicists of the time period involved.*

5.3 SC&A's Evaluation of Feasibility of DR for Five Iodine Radionuclides

Five of the 34 radionuclides that needed additional evaluation were radioiodine, as listed in Table 7-4 of RPRT-0090. NIOSH provided further evaluation of these five radionuclides in Attachment C of RPRT-0090. In that attachment, NIOSH postulates that radioiodine monitoring data from the period 1944–1954 can be used to formulate a coworker model to cover unmonitored radioiodine exposures during the period from 1955 to 1962, at which point it is presumed *in vivo* monitoring can be used to reconstruct doses. Specifically, NIOSH's methodology utilized data from 1947–1949 to develop a chronic intake model and states the following conclusions to justify it as bounding (NIOSH 2018a, p. 119):

1. *The projected urinary excretion (1.7 × 105 pCi/d) is more than an order of magnitude greater than the highest measured urinary excretion for routine sampling of 4.5 × 10^3 pCi/d…*

2. *The projected whole-body accumulation (1.2 µCi) is a factor of 4 larger than the highest measured whole-body accumulation of 0.28 µCi….*

3. *The projected air concentration (1.8[3] µCi/cm^3) is nearly a factor of 2 greater than the maximum operating level used to control facility air concentrations.*

SC&A has the following concerns with NIOSH's conclusions:

- Of the 168 bioassay samples evaluated in RPRT-0090, only 8 were taken prior to 1963 and only 2 were taken prior to the first use of the whole body counter (WBC) in 1961.

- Although RPRT-0090 notes that the projected urinary excretion rate is more than an order of magnitude higher than the maximum observed routine sample, no information or

[3] SC&A assumes that this is a typo and should actually read 1.8×10^{-8} µCi/cm^3 rather than 1.8 µCi/cm^3.

references are provided to indicate when that routine sample was taken. The analysis in Section C.7 of RPRT-0090 indicates that the evaluated urinalysis results spanned all the way to 1988.

- Per Table C-8 of RPRT-0090, the highest observed radioiodine urinalysis sample was 2.2×10^{7} picocuries per day (pCi/d), which is a factor of 130 higher than the projected urinary excretion rate using the chronic coworker model. NIOSH indicates this sample was categorized as "incident/follow-up/resample" but does not elaborate on the timeframe or conditions.

- Conclusion 2 notes that the projected whole-body accumulation is a factor of 4 larger than the highest whole-body accumulation recorded (0.28 microcuries [µCi]). However, this whole-body measurement was made in 1962, and no whole-body measurements were made until 1961. It has not been established that these data can be back-extrapolated to represent prior exposure conditions.

- Conclusion 3 notes that the projected chronic air concentration (1.8×10^{-8} µCi/cm^3) was nearly a factor of 2 higher than the maximum operating level used to control facility air concentrations. However, the air sampling data are only available in summary form, and neither the quantitative results nor the locations of these air samples are currently known.

- The ORNL site profile (NIOSH 2007, p. 34) notes that the tolerance-level air concentration during 1954 (the year just prior to the unmonitored period of interest) was actually 3×10^{-8} µCi/cm^3, 50% higher than the projected air concentration calculated in RPRT-0090 (1.8×10^{-8} µCi/cm^3).

In addition to the above commentary, the fundamental question is whether the monitoring data and exposure potential occurring during the period 1947–1949 is representative of the period when exposures to radioiodine were not adequately monitored. To establish that the exposure potential during the proposed coworker development years is representative and/or bounding, RPRT-0090 notes that the relative quantity of radioiodine processed was much greater than during the unmonitored period. Specifically, RPRT-0090 states (p. 119):

> *The quantity of iodine in process from 1955 to 1961 (1,000 to 3,600 Ci/yr) is bounded by the amount in process from 1947 to 1949 (8,800 to 42,000 Ci/yr).*

However, this comparison is between the commercially *produced* radioiodine (1955–1962) and the estimated annual radioactive lanthanum (RaLa) iodine *releases* for the period from 1944 to 1956 as provided in ATSDR 2008. This is an "apples to oranges" comparison for at least 1955 and 1956, when RaLa production at X-10 was still occurring. Unfortunately, the radioiodine releases from RaLa production are not necessarily provided on a year-by-year basis in the underlying documentation (ATSDR 2008). However, ATSDR 2008 does indicate that the highest radioiodine releases occurred during 1956 as a result of the processing of Hanford slugs. Figure 1 shows an excerpt from ATDSR 2008 that includes estimates of the highest annual releases by operation.

Figure 1. Excerpt from ATSDR 2008 Showing that the Largest Radioiodine Releases Occurred in 1956

Radioactive iodine source area (years of operation)		*Estimated maximum annual radioactive iodine release**	
		Curies	*Year*
Radioactive lanthanum processing	X-10 graphite reactor slugs (1944–1951)	64,200	1947
	Hanford slugs (1952–1956)	66,700	1956
Chemical separation of plutonium from Clinton Pile fuel (November 1943–January 1945)		23,600	1944
Thorex processing of short-decay irradiated thorium (July 1956–November 1957)		11,700	1957
Graphite reactor fuel slug ruptures (1944–1948)		96	1947

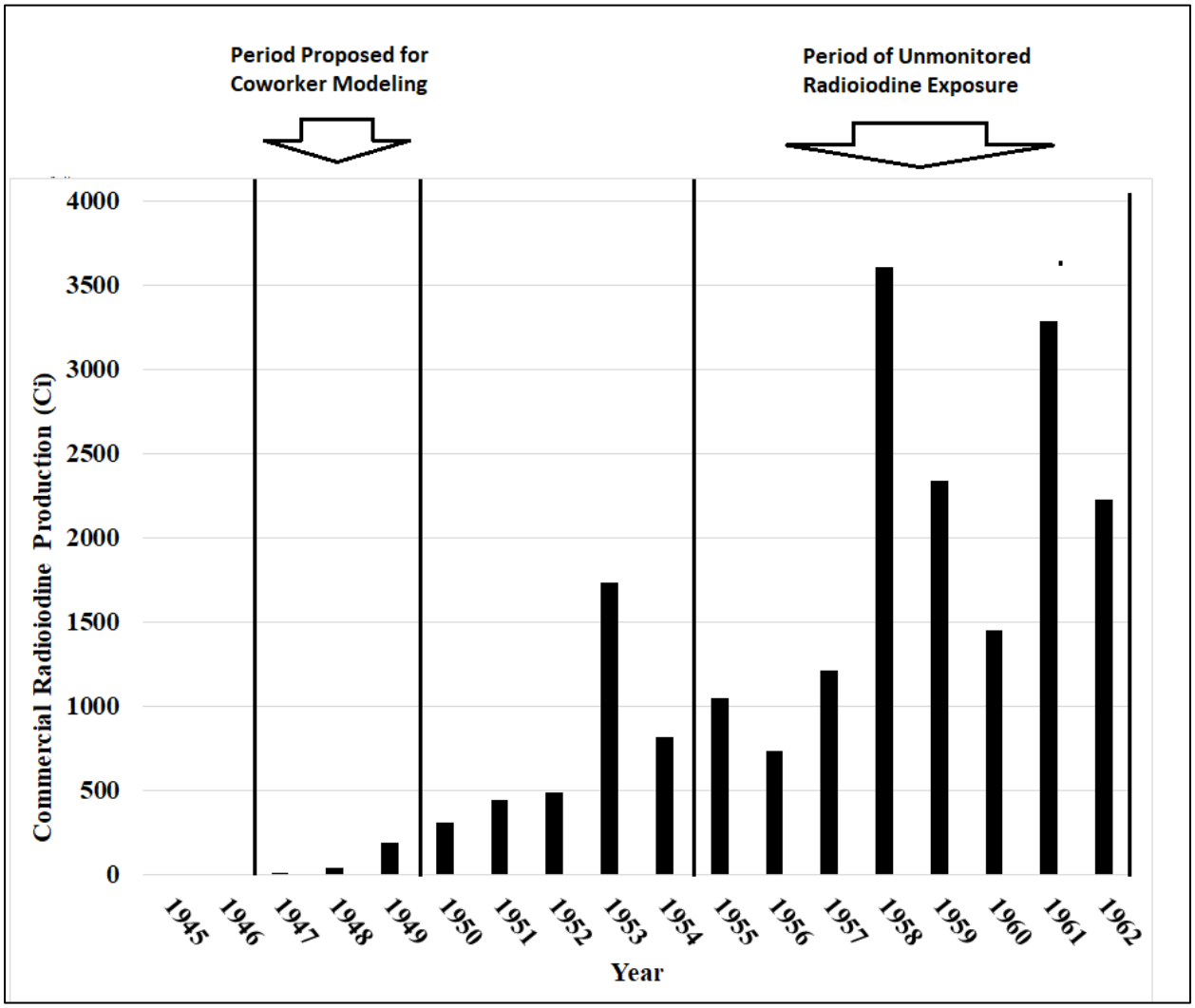
* Composition is presumed to be I-131.

Finding 5: 1955 and 1956 intakes may not be bound by earlier coworker data.
Assessment of RaLa radioiodine releases at X-10 indicates the highest annual releases occurred during the campaign to process Hanford slugs during 1956. Therefore, the radioiodine production and releases during the years used for coworker development (1947–1949) do not appear to bound the production throughput, at least during 1956 and possibly 1955.

When strictly comparing commercially produced radioiodine by year, the proposed coworker years (1947–1949) do not bound the unmonitored period (1955–1962), as shown in Figure 2. While commercial radioiodine production was far lower in quantity than the radioiodine produced via the RaLa program, the relative exposure potential between the two operations has not been discussed. In general, the radioiodine produced during the RaLa campaigns was considered byproduct waste material and vented to the atmosphere via either a 200-foot central pilot plant stack or the local 30-foot stack. Conversely, commercial production of radioiodine would logically involve more direct handling of the material; thus, exposure potential might be larger even though the actual production amounts are lower than the RaLa operation. The relative exposure potential between the two operations should be evaluated and discussed.

Observation 6: Additional RaLa production information should be provided.
NIOSH should provide an evaluation and discussion of any potential differences in exposure potential between commercial radioiodine production and the radioiodine produced via the RaLa operation to justify the extrapolation of exposures occurring during the years 1947–1949 to the unmonitored period (1955–1962).

Figure 2. Commercial Radioiodine Production by Year



6 SC&A'S EVALUATION OF ORAUT-RPRT-0090 WITH RESPECT TO *IN VIVO* BIOASSAY METHODS

SC&A finds that the adequacy and completeness of *in vivo* monitoring is unclear in RPRT-0090. Notwithstanding the *in vitro* methods highlighted in Attachment A, the only treatment of available *in vivo* technology and methods is found in Section 5.0 of RPRT-0090,[4] which provides a brief history of whole-body counting at ORNL accompanied by Table 5-1, containing a tabulation of *in vivo* counts by year (1961–1988) from the ORNL database versus the number of *in vivo* counts from annual reports. It is not clear from this treatment to what extent whole-body counting was relied upon for specific radionuclide monitoring capability, particularly MAPs and MFPs, both of which figured prominently at ORNL. It is also not clear for what radionuclides *in vivo* monitoring would have been the only bioassay capability for adequate estimation of internal intakes and dose.

ORNL operated several major accelerators along with several smaller accelerators, such as the Van de Graaff accelerator. These included the Oak Ridge Electron Linear Accelerator, the Oak Ridge Isochronous Cyclotron, and the Holifield Heavy Ion Facility. These accelerators produced or analyzed radionuclides of all types and would have generated localized emissions of both short-lived and longer-lived MAPs.

In terms of reactor operations, ORNL has had a long history of operating both research and test reactors, including the Graphite Reactor, Low-Intensity Test Reactor, Bulk Shield Reactor/Pool Critical Assembly, Oak Ridge Research Reactor, Tower Shielding Facility, Health Physics Research Reactor, and the High Flux Isotope Reactor. While NIOSH finds it has insufficient *in vitro* bioassay data for MFPs prior to 1950, this changed with the availability of cesium and strontium bioassay data starting in 1950. NIOSH's qualified conclusion about DR after that date is that it "may be feasible" (Taulbee 2012), which seems to suggest that more evaluation of available data and monitoring capabilities remains to be done. This raises two questions: (1) how were *in vitro* and *in vivo* monitoring capabilities together historically applied to MAPs and MFPs at ORNL? and (2) are internal dose data adequate between the availability of both capabilities (particularly in light of the aforementioned limitations in whole-body counting)?

For some of the longer-lived activation source terms (e.g., beryllium-7 [Be-7], chromium-51 [Cr-51], iron-55 [Fe-55] by way of monitoring for Fe-59) for which whole-body counting would have been a primary means of monitoring, whole-body counting was not routinely used until 1989, and a full *in vivo* program was not in place until as late as 1994 (Berger 2004). Before that time, *in vivo* counting was prescribed by individual area health physicists, who ordered it when an intake was suspected, leading to what was described as "inconsistent" selection of workers and the likelihood that intakes were missed for this reason (Berger 2004).

As with other national laboratories, the capability to detect and monitor for MAPs and MFPs was a function of technological advancement of WBCs and to what extent monitoring extended beyond the "core" radionuclides of concern (e.g., plutonium, uranium, tritium, cesium-137 [Cs-137], Sr-90). The timeframe for when such *in vivo* capabilities were realized for what

[4] A more detailed description of the history and capabilities of the ORNL *in vivo* monitoring program is provided in the internal dose TBD, ORAUT-TKBS-0012-5, Revision 02 (NIOSH 2013a).

radionuclides varied from site to site, but that would need to be established in order to ascertain whether those radionuclides requiring WBC monitoring were, in fact, being monitored adequately. This is not clear from RPRT-0090.

Moreover, SC&A finds that monitoring feasibility is not adequately defined by technological capability or procedures alone, which has been borne out in past reviews of *in vivo* monitoring at DOE sites. RPRT-0090 provides references for radiochemical analysis protocols available at ORNL to process actinides, rare earths, tritium, etc., from *in vitro* gross alpha and gross beta monitoring, but does not provide details from the *in vivo* (whole-body counting) program to support capabilities to monitor for the broad scope of MAPs and MFPs at ORNL.

For example, evolving WBC detector sensitivity over the time period in question (1955–1988) may determine the feasibility and MDA for applying *in vivo* monitoring for a number of the nuclides in question. As noted in RPRT-0090, the WBC system went through a series of upgrades since installation in 1959, beginning with a 4×4-inch sodium iodide (thallium) [NaI(Tl)] crystal, moving to a 512-channel analyzer and two 8×4-inch NaI(Tl) detectors in 1961. A phoswich detector was installed sometime between 1971 and 1974 (Auxier et. al. 1975) followed by an 80-centimeter squared HPGe counting array in 1980 (Berger and Lane 1981). It is not clear from RPRT-0090 what detection sensitivities were being achieved over time and for what radionuclides the WBC would be necessary for adequate monitoring (albeit, for nonroutine purposes, as routine monitoring did not begin until 1989, according to [redacted] 2004).

A WBC "library" distinguishing specific energy spectrums for nuclides of interest is a critical tool for the internal dosimetrist to ensure that the relevant radioisotopes can be identified, a yield for the particular energies can be calculated, and a deposition estimated. Calibration of the WBC for specific target spectrums is also necessary, typically through use of a phantom or similar standard. If the former is incomplete or the latter deficient, monitoring capability would be hampered and intakes may be missed.

The 1995 WBC nuclide library for ORNL (ORNL 1995) includes 9 of the 28 radionuclides listed in Table 7-6 (Cr-51, manganese-54, cobalt-57, Ru-103, cadmium-109 (Cd-109), tin-113, Cs-131, barium-133, cerium-139) as not having corresponding proof of monitoring capability, and three of the radioiodines addressed in Attachment C (I-125, I-131, I-133).[5] These WBC library citations are from the ORNL 1995 internal dosimetry technical basis document. Given the lack of formality for the ORNL *in vivo* program before 1989 (as noted in ORAUT-TKBS-0012-5, Revision 02), has NIOSH verified what WBC monitoring was actually conducted, in practice, by establishing either such radionuclide-specific library listings or *in vivo* monitoring procedures for these earlier time periods? If operational presence for and exposure potential to these radionuclides can be demonstrated in the 1955–1988 timeframe, how can ORNL capabilities to develop feasible monitoring be demonstrated in the absence of WBC libraries, calibrations, and

[5] It is clear that for some of these radionuclides, e.g., the radioiodines, an *in vitro* method was also available as noted in RPRT-0090. However, RPRT-0090 does not address the WBC monitoring capability at ORNL in a manner that enables a complete assessment of monitoring feasibility across the broad spectrum of radionuclides for which exposure potential existed.

procedures for them? Are there any records of actual monitoring and worker exposure for radionuclides that would have been monitored via the WBC (e.g., NOCTS, bioassay records)?

For five other radionuclides subject to WBC monitoring at ORNL in the Table 7-6 compilation (Be-7, gallium-67, selenium-75, osmium-185 [Os-185], mercury-197 [Hg-197]),[6] the 1995 nuclide library would not likely have included them due to lack of operational prevalence by that later time. It would be useful to confirm this by reviewing earlier WBC libraries and likewise ascertaining whether actual monitoring was taking place in the 1955–1988 timeframe. All told, half (14) of the 28 radionuclides listed in Table 7-6 as lacking evidence of monitoring appear to be radionuclides for which WBC monitoring would have been necessary.

The technological ability to identify, and the capability needed to analyze, specific radionuclide intakes at the national laboratories (e.g., Los Alamos National Laboratory [LANL], Lawrence Berkeley National Laboratory [LBNL], Brookhaven National Laboratory, and Lawrence Livermore National Laboratory), particularly for MAPs and MFPs, was still in its infancy in the 1950s and 1960s. In general, these methods were not yet applied to bioassays on a routine basis (and the results recorded and rendered useful for DR) until later in the 1970s and 1980s because they required considerable development time to improve detector sensitivity, calibration, and stability. To illustrate, like ORNL, LANL developed WBC technology in the late 1950s and began performing routine *in vivo* counts in June 1969 (unlike ORNL, which did not perform routine WBCs until 1989). However, even when the technological capability was available (e.g., in the 1970s and 1980s), some laboratories, such as LANL, did not maintain energy spectra information for exotics in their WBC libraries and did not routinely monitor for them.

As SC&A pointed out in its review of NIOSH's ER for LANL (SC&A 2010, p. 9):

> *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 (LANSCE), while required procedurally for routine internal dosimetry evaluation, were not included in the in-vivo program library at that time.… 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.*

An obvious question would be whether ORNL was much more advanced and ahead of its time than LANL and these other laboratories in this respect (or not), and was the WBC fully

[6] For validation, SC&A cross-referenced these with WBC monitoring results at LANL for the post-1975 period.

operational and capable of monitoring for these exotics for which *in vitro* methods would not have been available? If field evidence indicates it was not, as with LANL, the premise that available capability translates to adequate monitoring does not hold true.

This exact premise was also tested in an SC&A review of the bioassay program at LBNL in terms of capability to monitor for specific radionuclides associated with the laboratory's extensive accelerator operations (SC&A 2014). This review addressed the claim in a 2013 NIOSH white paper supporting the LBNL site profile that "LBNL's internal bioassay program, which was fully operational by 1962, was capable of determining specific alpha, beta, and gamma emitting radionuclides. Urinalysis and fecal sampling were performed, as well as whole body counting" (NIOSH 2013c, p. 3). SC&A sampled the NOCTS claimant file to validate the presence of bioassay data, particularly radionuclide-specific information, for job titles for which exposure potential for intakes would have been likely. The following is an excerpt of results of that review.

> *While LBNL was a forerunner in accelerator health physics, it appears from an SC&A preliminary evaluation of NIOSH claimant files for LBNL that the capability to analyze specific radionuclides may have remained in a laboratory development stage, as opposed to being applied to routine bioassays, especially for WBCs. LBNL claim files were searched for POCs <50% (to ensure a complete DR) and if there was a DR report on file (to see how the internal doses were determined). This resulted in 195 claims. Claims with job titles that indicated potential exposure were selected for investigation. This included Physicist, Nuclear Physicist, Chemist, Lab Tech, Technician, Researcher, Accelerator Operator, Maintenance, HP, Machinist, and Magnet Tester. A total of 25 claimants that worked some time during the period 1960s–1980s were analyzed by reviewing their DOE Response files and DR reports to determine if bioassays were recorded, and if so, what bioassay information is available; i.e., frequency, urinalyses, WBCs, and radionuclide identification. From this review, there did not appear to be many bioassay results recorded, and very few routine bioassays; only 4 claimants had any bioassay records out of the 25 reviewed. What bioassays were recorded generally did not contain nuclide-specific information (mainly gross gamma, beta, and alpha counts) and did not appear to be used in the DR process, except for a 1971 P-32 measurement for a potential acute intake. It would seem reasonable to expect that some of the personnel that worked at the facilities on a routine basis, such as operators and technicians, would have some records of routine, or at least periodic, bioassays in their records if the HP program was firmly in place and operational by 1962.* [SC&A 2014, pp. 1–2].

Again, as the LBNL assessment demonstrated, the existence of monitoring capabilities and procedures belied the scope and completeness of the bioassay records themselves and required a reexamination of available LBNL bioassay data to determine alternate means to estimate internal dose beyond reliance on specific bioassay results.

For ORNL, a basis for a similar concern can be found in internal self-audits or appraisals of the laboratory's internal dosimetry program. In one such self-appraisal of the radiation protection program for ORNL's internal dosimetry program, as implemented at HFIR, the following

findings and recommendations, made in 1988, raise questions about the formality, accuracy, and effectiveness of the ORNL internal dosimetry program:

- "There is no technical basis documented for whole-body counting or bioassay" (MMES 1988, PDF p. 38).
- "Procedures for performing internal dose assessments should be developed. The current practice of not performing dose assessments until activity levels indicate greater than 50 percent maximum permissible organ burden (MPOB) appears to be too high" (MMES 1988, PDF p. 42).
- "Frequency schedules have been established and are documented in Appendix 12 of the Health Physics Manual; however, the established frequencies for whole-body counting of HFIR personnel are not followed" (MMES 1988, PDF p. 39).
- "Procedures should be developed for establishing Minimum Detectable Activities for the whole-body counter" (MMES 1988, PDF p. 43).

Another illustration of capability and procedures not being a sufficient basis alone to determine bioassay feasibility was a significant Price-Anderson Amendments Act violation and penalty levied by DOE on ORNL in 1998. This was for lack of broad compliance with bioassay requirements under 10 CFR Part 835 regulatory requirements by ORNL construction contractor MK-Ferguson, as well as by Martin Marietta, the operating contractor, which included this finding (DOE 1998, p. 2):

> *Further, after identifying problems with the bioassay program in October 1996, i.e., that approximately 100 positive bioassay results had been identified as positive that had previously been considered negative, results for these two workers* [the specific subject of the enforcement action] *were administratively invalidated without further evidence that uptakes had not occurred. These repeated failures resulted in additional 10 CFR 835 deficiencies in the areas of record keeping and issuance of accurate worker annual exposure reports. Other deficiencies identified during the investigation included (1) missed bioassay sampling, (2) failure to initiate special follow-up bioassay monitoring as required, (3) failures to notify workers of their exposures in a timely manner, and (4) failures to implement work restrictions in accordance with written procedures.*

If fundamental problems existed with ORNL bioassay program compliance and implementation well into the 1990s, it underscores the importance to go beyond the written procedures and methods to investigate whether actual practice corresponded to these established capabilities and procedures in the earlier time period.

In summary, the stated purpose of RPRT-0090 to evaluate monitoring capability by matching identified radionuclides with corresponding bioassay methodologies as a gap analysis for demonstrating feasibility seemingly overlooks more significant program shortfalls in the 1950s into the 1980s. Notably, the lack of routine bioassay monitoring would lead to missed intakes, and limitations on whole-body counting capability for MAPs and MFPs could preclude reliable

monitoring of such intakes. Reliance on inventory-based gauges of "radiotoxicity" or ORNL's capability to overcome these and other technological or programmatic limitations by being able to develop needed monitoring is questionable and would need some level of corroboration.

Finding 6: Adequacy and implementation of *in vivo* bioassay program not addressed. *Information is lacking for the actual implementation of the ORNL* in vivo *program, including what and how radionuclides were monitored in practice, what and how workers were identified and included for counting, and how capability to monitor for MAPs, MFPs, and exotic radionuclides paced both technology developments and onsite monitoring practice (e.g., routine vs. nonroutine monitoring). SC&A recommends that the Work Group request a review of available records, particularly internal dosimetry program records and WBC nuclide libraries, and scheduling of interviews with appropriate ORNL dosimetry staff.*

7 POST-1988 INVENTORY AND CLEANUP

RPRT-0090, page 6, states:

> *The period under evaluation in this report is August 1, 1955, through December 31, 1988. The start date was selected to coincide with the end of the SEC period (NIOSH 2012). The end date coincides with the end of large-scale isotope production at ORNL.*

Although large-scale production of some of the radionuclides may have ended at ORNL in 1988, the remaining inventory was still present and required storage/disposal, and many facilities remained intact awaiting future activities. It is clear that after that date, the facilities involved were in various phases of continued operation, abandonment, deactivation, and D&D. During this period after 1988, workers, consisting primarily of surveillance, maintenance, and cleanup workers (e.g., CTWs), entered these buildings and should have been monitored for the radionuclides to which they would have been potentially exposed. However, for various reasons, including lack of institutional memory regarding historic facility use of isotopes, state of transferrable contamination, and surveillance capabilities, workers may have been exposed to facility contamination in the isotope production facilities in question during the postproduction period.

For example, in Building 3026-C (Krypton Enrichment Facility), workers were contaminated in 1988 with tritium that went initially undetected because routine tritium surveillance had been discontinued because of nonfunctioning tritium air monitors, lack of adequate survey instruments, and no HP surveillance prior to entry for work activities (Ramey 1988).

In 1990, for Building 3517 (Fission Product Development Laboratory), “craftspersons” were directed to begin wearing special protective clothing upon entry to due to “recent contamination incidents” involving transferrable radioactive contamination in “relatively inaccessible areas” (Patton 1990).

In the mid-1990s, ORNL proceeded to conduct facility characterization studies and, ultimately, D&D for Building 3515, the FPPP, one of the first facilities at ORNL to extract radioisotopes from liquid radioactive wastes (Mandry and Snedaker 1994). While this longstanding ORNL operation was not administratively part of Isotope Production (as noted earlier), it clearly involved a wide range of exotic radionuclides and extensive contamination, and it would have entailed corresponding bioassay coverage for workers during D&D activities.

With the uncertainty about what radionuclides were present in abandoned or deactivated facilities, how were surveillance and maintenance, cleanup, and other workers monitored for internal emitters that may have been present in these facilities? What facility operational and contamination characterization was performed, and what bioassay methods were used?

Finding 7: Unclear treatment of post-1988 monitoring capability during abandonment, deactivation, and decontamination and decommissioning phases.

After radionuclide production ended, the adequacy of monitoring and feasibility of assigning intakes from the storage, disposal, and D&D of the facilities has not been addressed. This issue is especially important for the ORNL Isotopes Division because it processed and concentrated unusual radionuclides that would not be encountered during the normal D&D process.

8 SUMMARY AND CONCLUSIONS

SC&A evaluation of RPRT-0090 indicated the following finding and observations that need further clarification and evaluation.

8.1 FINDINGS

Finding 1: Scope of RPRT-0090 needs to be clearly defined.
SC&A finds that the scope of RPRT-0090 needs to be clarified in terms of whether (and how) it is meant to encompass the "reserved" portion of the ER for "cyclotrons, accelerators, and reactors" and whether NIOSH intends to address the full scope of radionuclides involved in waste management (including D&D), site-wide construction, and maintenance.

Finding 2: Incomplete radionuclide and radioisotope facility inventory.
A sampling of the radionuclides listed in Table 7-2 found a few missing when compared with operational and customer records. Likewise, a few ORNL facilities that historically handled radioisotopes are also not included in those cited and addressed in RPRT-0090. Given the operational diversity of ORNL accelerator and reactor operations, consideration should be given to an inventory scope that encompasses isotopic source terms broader than that of the Isotope Division.

Finding 3: Attachment A *in vitro* bioassay methods lack information about actual implementation.
In vitro *bioassay methods are outlined in Attachment A, but it does not include any discussion or references regarding their actual field implementation. The exclusion of comparable* in vivo *monitoring methods makes a review of ORNL monitoring capability incomplete.*

Finding 4: Feasibility of monitoring 28 radionuclides not adequately addressed.
While the 28 radionuclides were discussed in Section 7.2 and some of their characteristics were listed in Tables 7-4, 7-5, and 7-6 of RPRT-0090, the feasibility of monitoring for intakes for DR purposes was not completely addressed, particularly given the lack of routine bioassays in the earlier years. Methods for accounting for the lack of monitoring of these radionuclides need to be addressed in more detail, and an acceptable resolution derived. SC&A finds that it is not possible at this time to validate implementation without further onsite review, including document review and interviews with health physicists of the time period involved.

Finding 5: 1955 and 1956 intakes may not be bound by earlier coworker data.
Assessment of RaLa radioiodine releases at X-10 indicates the highest annual releases occurred during the campaign to process Hanford slugs during 1956. Therefore, the radioiodine production and releases during the years used for coworker development (1947–1949) do not appear to bound the production throughput, at least during 1956 and possibly 1955.

Finding 6: Adequacy and implementation of *in vivo* bioassay program not addressed.
Information is lacking for the actual implementation of the ORNL in vivo *program, including what and how radionuclides were monitored in practice, what and how workers were identified and included for counting, and how capability to monitor for MAPs, MFPs, and exotic radionuclides paced both technology developments and onsite monitoring practice (e.g., routine*

vs. nonroutine monitoring). SC&A recommends that the Work Group request a review of available records, particularly internal dosimetry program records and WBC nuclide libraries, and scheduling of interviews with appropriate ORNL dosimetry staff.

Finding 7: Unclear treatment of post-1988 monitoring capability during abandonment, deactivation, and decontamination and decommissioning phases.
After radionuclide production ended, the adequacy of monitoring and feasibility of assigning intakes from the storage, disposal, and D&D of the facilities has not been addressed. This issue is especially important for the ORNL Isotopes Division because it processed and concentrated unusual radionuclides that would not be encountered during the normal D&D process.

8.2 OBSERVATIONS

Observation 1: Inventory discrepancy.
A sampling of some of the inventory of the radionuclides for the early years indicated some discrepancies in inventory between Table 7-2 in RPRT-0090 and NIOSH's X-10 Inventory spreadsheet.

Observation 2: Specific alpha-emitting radionuclide needs to be identified for DR.
The specific radioisotope monitored is not always presented in NIOSH's X-10 Database as it generally is in the NOCTS files. Gross alpha results could be applied to many radionuclides. Is the information on the original bioassay cards available in the X-10 Database, and will the X-10 Database be used in DR or coworker model development?

Observation 3: Trans-plutonium radionuclides may need further analyses.
SC&A is concerned that assigning trans-plutonium gross alpha counting results as Am-241 intakes without consideration of other potential trivalent alpha-emitting actinides (such as Bk-249, Cf-252, Cm-242, Cm-244, etc.) and their individual radiotoxicity could result in underestimating the internal dose. It could be beneficial to determine if assigning the intake as Am-241 is claimant favorable, considering the exotic trans-plutonium radionuclides at ORNL.

Observation 4: Use of gross beta or gamma count data could result in underestimate of assigned dose.
Using gross beta or gamma count data without knowledge of the radionuclide the counter was calibrated with and the radionuclides in the bioassay sample could result in assigning the incorrect radionuclide and radioactivity content because of different counting efficiencies for the different energy of beta particles and gamma photons. Has this issue been addressed for DR for ORNL claimants? Additionally, bioassay data for at least one beta-emitting radionuclide (Ru-106) could not be located for several years that Table 7-2 indicated it was available.

Observation 5: The results in Table 7-6 depend on inventory used.
As outlined in Observation 1, there appear to be some discrepancies in the inventory used by NIOSH compared to those provided to SC&A for evaluation of RPRT-0090. These discrepancies change a few of the results of Table 7-6, as illustrated in Table 3 of this report.

Observation 6: Additional RaLa production information should be provided.
NIOSH should provide an evaluation and discussion of any potential differences in exposure

potential between commercial radioiodine production and the radioiodine produced via the RaLa operation to justify the extrapolation of exposures occurring during the years 1947–1949 to the unmonitored period (1955–1962).

Several of the above issues identified by SC&A may have resulted from NIOSH updating the inventory list that was used in preparing RPRT-0090 of 2018, whereas NIOSH's X-10 Inventory spreadsheet was dated February 2015.

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APPENDIX A: EXAMPLE OF EVALUATION OF CM-244 AND RU-106 BIOASSAY DATA

Table A-1. Cm-244 Data

Year	X-10 Inventory	RPRT-0090 Table 7-2 Cm-242	Number of Bioassays with Code 000, TP0, or CM0 in X-10 Database
1955	None	No	1
1956	None	No	0
1957	None	No	0
1958	None	No	2
1959	None	No	0
1960	None	No	0
1961	None	No	1
1962	None	Green [?]	0
1963	None	Green [?]	1
1964	0.002 gm	Green	Many
1965	None	No	Many
1966	3.5 gm	Green	Many
1967	19 gm	Green	Many
1968	144 gm	Green	Many
1969	213gm	Green	Many
1970	29.4 gm	Green	Many
1971	30.9 gm	Green	Many
1972	96.7 gm	Green	Many
1973	416.1 gm	Green	Many
1974	117 gm	Green	Many
1975	1,435 mg	Green	Many
1976	174 mg	Green	Many
1977	1,125 mg	Green	Many
1978	51.5 gm	Green	Many
1979	132745 mg	Green	Many
1980	10 mCi	Green	Many
1981	725 mg	Green	Many
1982	1,622 mg	Green	Many
1983	151,002 mg	Green	Many
1984	1,061 mg	Green	Many
1985	1,516 mg	Green	Many
1986	61,000 gm	Green	Many
1987	141 mg	Green	Many
1988	22 gm	Green	Many

* The *Green [?]* notation means it is not clear why this cell is marked green in Table 7-2.

Table A-2. Ru-106 Data

Year	X-10 Inventory	RPRT-0090 Table 7-2 Ru-106 *	Number of Bioassays with Code 000, 013, GB0, or RU6 in X-10 Database
1955	717	Green	2
1956	330	Green [?]	0
1957	397	Green [?]	0
1958	None	No	2
1959	1,360	Green [?]	0
1960	936	Green	Some
1961	233	Green	Some
1962	338	Green	Some
1963	810	Green	Some
1964	937	Green	Some
1965	500	Green	Some
1966	326	Green	Some
1967	221	Green	Some
1968	201	Green	Some
1969	35	Green	Some
1970	119	Green	Some
1971	85	Green	Some
1972	123	Green	Some
1973	105	Green	Some
1974	610	Green	2
1975	7	Green [?]	0
1976	346	Green	6
1977	133	Green	1
1978	704	Green [?]	0
1979	286	Green	Some
1980	350	Green	Some
1981	72	Green	Some
1982	360	Green	Some
1983	438	Green	Some
1984	132	Green	Some
1985	59	Green	Some
1986	81	Green [?]	0
1987	48	Green [?]	0
1988	15	Green [?]	0

* The *Green [?]* notation means it is not clear why this cell is marked green in Table 7-2.

APPENDIX B: EXAMPLES OF NOCTS ORNL RECORDS

Figure B-1. 1955 Plutonium, Uranium, and Strontium

Sample No. [redacted]

INDIVIDUAL URINALYSIS REPORT

Total Volume 1530 Total Hours 25 Name [redacted]

Collection Period: From 1-7-55 10 05 AM Badge No. [redacted] Meter No. 1861
(Date, Time)

To 1-8-55 10 00 AM. H. P. Representative [redacted]
(Date, Time)

	A	B	C
Analyzed For	Pu	U	Sr.
Volume Used	1430 ml.	100 ml.	1430 ml.
Counter No.	8	9	1
Date	1-11-55	1-12-55	1-13-55
SPECIMEN COUNTING DATA			
1. Total Count	102 cts.	28 cts.	365 cts.
2. Counting Time	985 min.	120 min.	70 min.
3. Gross c/m	0.10 c/m	0.23 c/m	18.2 c/m
BKG. COUNT (COUNTER & DISC)			
1. Total Count	35 cts.	13 cts.	382 cts.
2. Counting Time	1000 min.	110 min.	70 min.
3. Bkg. c/m	0.04 c/m	0.12 c/m	16.6 c/m
4. Bkg. (Reagents)	0.01 c/m	c/m	c/m
5. Total Bkg.	0.05 cts.	0.12 cts.	16.6 cts.
NET COUNTS/M	0.05 c/m	0.11 c/m	1.6 c/m
NET c/m/24 hr. Specimen	0.05 c/m	0.11 c/m	1.6 c/m
D/M	0.14 d/m	0.22 d/m	5.4 d/m
Per Cent of MPC	2.0 %	4.4 %	0.8 %

Assumptions:
1. The total volume represents a 24-hour output unless otherwise noted.
2. 136,500 d/m/μg for Pu.
3. Uniform Pu excretion per ml.
4. Excretion rate of Pu 0.01%/day of that in the body (the excretion rate is higher for Pu recently taken into the body).
5. Counter geometry ~50% for alpha, ~25% for beta.

X-491 A, B, C 4-21-52

A, B, C [redacted]

Figure B-2. 1955 Thorium

Form X-463

Date 11-15-55

Subject: Request for ~~Urine~~ feces Specimen

Re: [redacted] P. R. No [redacted]

Index No. [redacted]

It is recommended that the above named employee submit a ~~urine~~ feces specimen for ~~urinalysis~~ analysis for the 24 hour period ending at _____ AM PM on 11-17-55

Specimen containers may be obtained at Bldg Brought to him The employee is requested to leave his specimen at Bldg. _____ as soon as possible after the sampling period. He should attach this notice to the specimen container when he submits it for analysis.

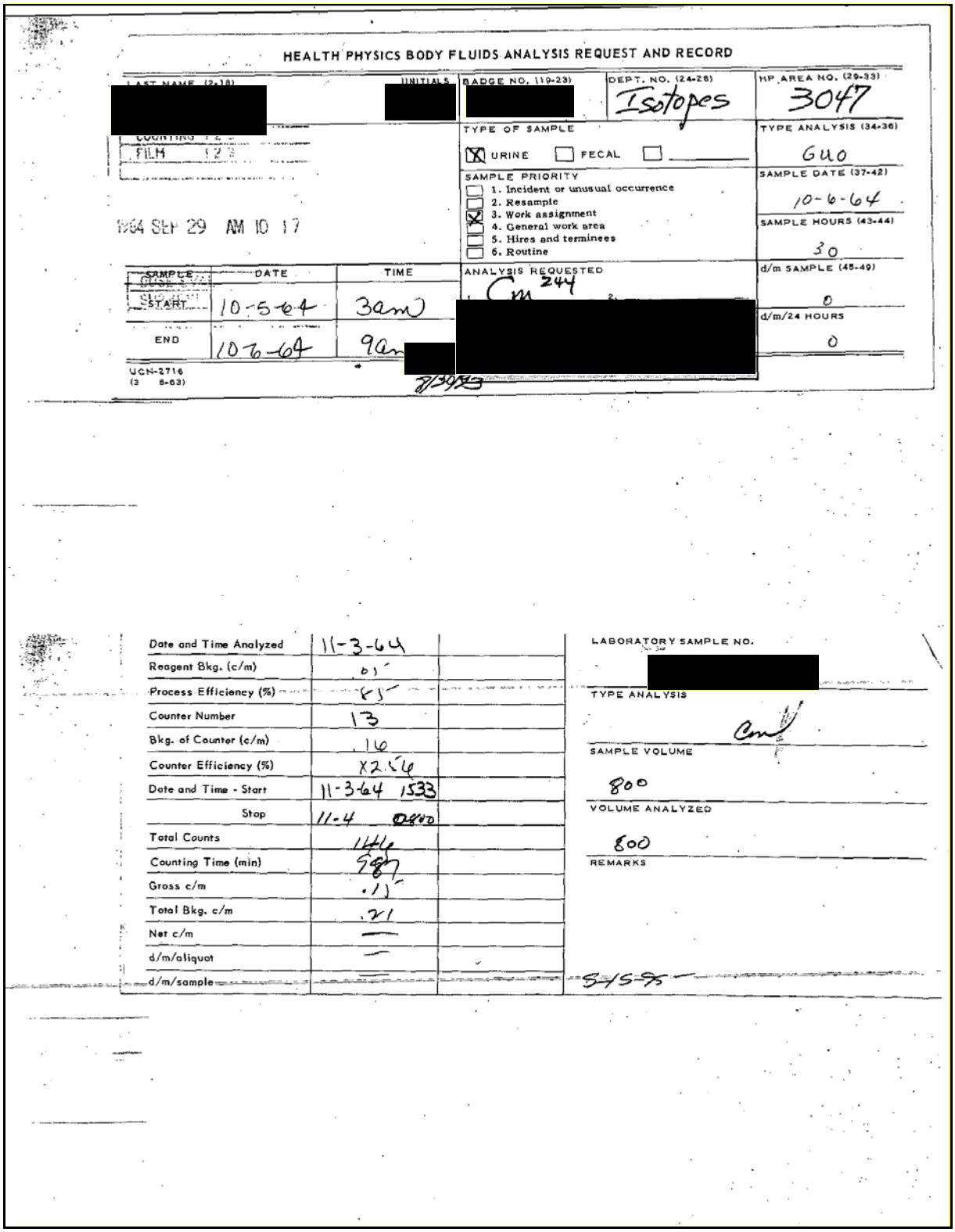
Requested by: [redacted]
Health Physics Division

in no. 6 watch door

The following is to be filled in on copies 2 and 3:

1. Radioactive material with which the employee was working: Thorium oxide
2. The approximate quantity of material involved:
3. The total length of time that the employee has worked with the material involved:
4. Conditions under which employee was working with the material (closed hood, open beaker, etc.): open rm

Figure B-3. 1964 Cm-244



HEALTH PHYSICS BODY FLUIDS ANALYSIS REQUEST AND RECORD

LAST NAME (2-18) [redacted] INITIALS [redacted] BADGE NO. (19-23) [redacted] DEPT. NO. (24-28) Isotopes HP AREA NO. (29-33) 3047

COUNTING FILM 123

1964 SEP 29 AM 10 17

TYPE OF SAMPLE: [X] URINE [ ] FECAL [ ]

TYPE ANALYSIS (34-36): GUO

SAMPLE PRIORITY
1. Incident or unusual occurrence
2. Resample
[X] 3. Work assignment
4. General work area
5. Hires and terminees
6. Routine

SAMPLE DATE (37-42): 10-6-64

SAMPLE HOURS (43-44): 30

d/m SAMPLE (45-49): 0

d/m/24 HOURS: 0

SAMPLE	DATE	TIME
START	10-5-64	3am
END	10-6-64	9am

ANALYSIS REQUESTED: Cm 244 [redacted]

UCN-2716 (3 8-63)

8/30/93

Date and Time Analyzed	11-3-64	
Reagent Bkg. (c/m)	.01	
Process Efficiency (%)	65	
Counter Number	13	
Bkg. of Counter (c/m)	.16	
Counter Efficiency (%)	X2.54	
Date and Time - Start	11-3-64 1533	
Stop	11-4 0840	
Total Counts	144	
Counting Time (min)	587	
Gross c/m	.11	
Total Bkg. c/m	.21	
Net c/m	—	
d/m/aliquot	—	
d/m/sample	—	

LABORATORY SAMPLE NO. [redacted]

TYPE ANALYSIS: Cm

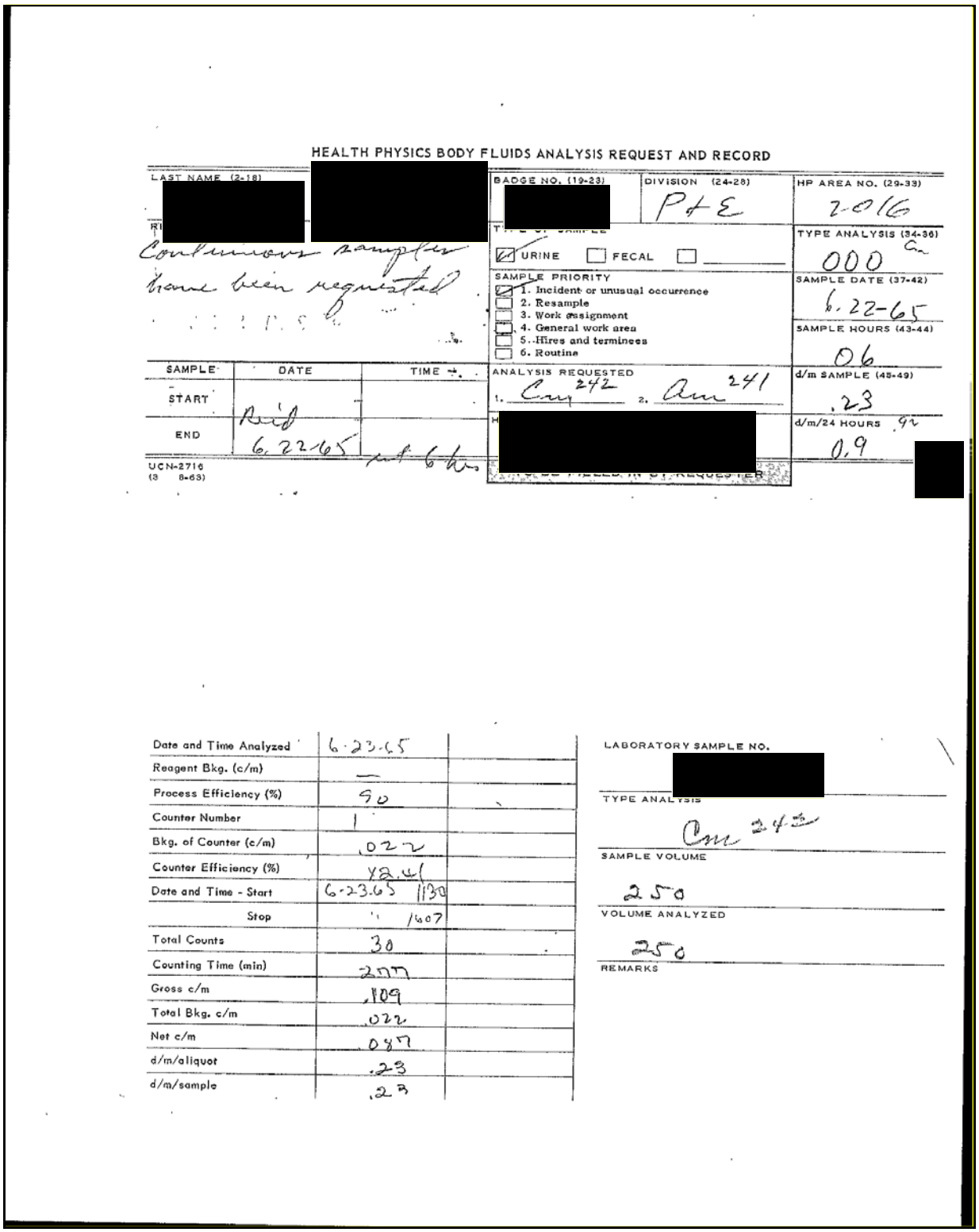
SAMPLE VOLUME: 800

VOLUME ANALYZED: 800

REMARKS

5-15-95

Figure B-4. 1965 Cm-242 and Am-241



HEALTH PHYSICS BODY FLUIDS ANALYSIS REQUEST AND RECORD

LAST NAME (2-18) [redacted]

BADGE NO. (19-23) [redacted]

DIVISION (24-28) P+E

HP AREA NO. (29-33) 2-016

Continuous samples have been requested

TYPE OF SAMPLE: ☑ URINE ☐ FECAL ☐

TYPE ANALYSIS (34-36) Cm 000

SAMPLE PRIORITY
- ☑ 1. Incident or unusual occurrence
- ☐ 2. Resample
- ☐ 3. Work assignment
- ☐ 4. General work area
- ☐ 5. Hires and terminees
- ☐ 6. Routine

SAMPLE DATE (37-42) 6.22-65

SAMPLE HOURS (43-44) 06

SAMPLE	DATE	TIME
START	Rec'd	
END	6.22.65	~ 6 hrs.

ANALYSIS REQUESTED 1. Cm 242 2. Am 241

d/m SAMPLE (45-49) .23

d/m/24 HOURS .92 0.9

UCN-2716 (3 8-63)

Date and Time Analyzed	6-23-65	
Reagent Bkg. (c/m)	—	
Process Efficiency (%)	90	
Counter Number	1	
Bkg. of Counter (c/m)	.022	
Counter Efficiency (%)	28.4	
Date and Time - Start	6-23-65 1130	
Stop	1407	
Total Counts	30	
Counting Time (min)	277	
Gross c/m	.109	
Total Bkg. c/m	.022	
Net c/m	.087	
d/m/aliquot	.23	
d/m/sample	.23	

LABORATORY SAMPLE NO. [redacted]

TYPE ANALYSIS Cm 242

SAMPLE VOLUME 250

VOLUME ANALYZED 250

REMARKS

APPENDIX C: EXAMPLE OF X-10 DATABASE FOR AM-241

Table C-1. Excerpt for Part of 1955 and 1956 Applicable to Am-241 from ORNL Database Containing 25,162 Entries

Bioassay Code [a]	dpm per Sample [b]	dpm per 24 Hours	Date Received
GU0	0	0000000.00	12-Dec-55
GU0	0	0000000.00	13-Dec-55
GU0	0.1	0000000.01	14-Dec-55
GF0	1.5	0000000.15	16-Dec-55
GF0	8.8	0000000.88	17-Dec-55
GF0	2.5	0000000.25	17-Dec-55
GF0	26	0000002.60	17-Dec-55
GU0	0	0000000.00	17-Dec-55
GF0	47	0000004.70	18-Dec-55
GF0	12	0000001.20	18-Dec-55
GF0	4.6	0000000.46	18-Dec-55
GU0	0	0000000.00	19-Dec-55
GU0	0.1	0000000.01	19-Dec-55
GU0	0	0000000.00	19-Dec-55
GF0	3.6	0000000.36	21-Dec-55
GU0	0.2	0000000.02	21-Dec-55
GU0	0	0000000.00	27-Dec-55
GU0	0	0000000.00	27-Dec-55
GU0	0	0000000.00	03-Jan-56
GF0	22	0000002.20	04-Jan-56
GF0	1.2	0000000.12	05-Jan-56
GF0	0.4	0000000.04	05-Jan-56
GF0	15	0000001.50	05-Jan-56
GF0	0.9	0000000.09	06-Jan-56
GU0	0.1	0000000.01	06-Jan-56
GF0	8.7	0000000.87	06-Jan-56
GF0	33	0000003.30	06-Jan-56
GF0	13	0000001.30	06-Jan-56
GF0	0.8	0000000.08	07-Jan-56
GU0	0	0000000.00	08-Jan-56
GF0	12	0000001.20	08-Jan-56
GF0	19	0000001.90	08-Jan-56
GU0	0	0000000.00	09-Jan-56
GU0	0	0000000.00	09-Jan-56
GU0	0	0000000.00	09-Jan-56

[a] GU0 = gross alpha in urine sample. GF0 = gross alpha in fecal sample.
[b] dpm = disintegrations per minute.

In addition to these data, the spreadsheet contains other information, such as the worker's social security number and name associated with each bioassay sample.

APPENDIX D: EVALUATION OF INTERNAL DOSE USING A FRACTIONAL INTAKE BASED ON AMOUNT OF MATERIAL IN PROCESS

Section 7.2 of RPRT-0090 describes a method of deriving internal doses for the 28 radionuclides requiring further evaluation that is based on assuming the intake potential can be bounded using a fraction of the total radionuclide inventory in process during a given year. Specifically, RPRT-0090 (p. 40) states:

> *An evaluation of the dosimetric consequences of each of the 28 radionuclides requiring further analysis was conducted. The committed dose to the maximally exposed organ from inhalation of 1×10^{-5} of the total annual inventory was computed for the year with the maximum recorded inventory…. The factor of 1×10^{-5} was selected based on the guidance in NUREG-1400, which postulates that 1×10^{-6} times the material handled could serve as a reasonable estimate of the quantity that could be inhaled (Hickey et al. 1993). A factor of 10 was added to ensure a conservative evaluation.*

Although NIOSH cites NUREG-1400 (Hickey et al, 1993), the underlying data forming the basis of this factor[7] is found in Brodsky 1977 and 1980. Brodsky 1977 focused the evaluation on the potential for tritium intake at various facilities and processes. Although tritium is not one of the 28 radionuclides evaluated in Section 7.2 of RPRT-0090, the volatile nature of tritium compounds (in particular tritiated water [HTO] and tritium gases) could be considered a conservative comparison to most other contaminants. Brodsky 1977 evaluated the following tritium intake situations summarized in Table D-1.

Table D-1. SC&A's Summary of Fractional Intake Data in Brodsky 1977

Industry or Activity Type	Form of Tritium	Protective Enclosure	Estimated Fractional Intake*
Luminous timepiece	Luminous paint	Hood, bench	6×10^{-6} per employee*; 1.2×10^{-4} for 20 employees
Luminous timepiece	Luminous paint, organic polymer	Open bench, modified with local exhaust	6×10^{-7} per employee*; (2×10^{-6} for 3 employees)
Luminous paint, mixing polymer, phosphor and binder	Luminous paint, organic polymer	Glovebox, single thickness rubber gloves	10^{-7} per employee
Luminous paint, research development	Gas, HTO, various organics	None	4×10^{-6} to 3×10^{-5} per employee
Luminous watch storage area	Solid paint on timepieces	Poorly ventilated room	7×10^{-7} per employee
Academic laboratories	Various	Fume hoods	$<5\times10^{-6}$ per employee
Academic laboratories – spill	HTO	Hood	10^{-2} highest observed employee dose

[7] While the term "factor" is used in RPRT-0090, the underlying literature uses the term "fractional intake" and thus that will be used in this evaluation.

Industry or Activity Type	Form of Tritium	Protective Enclosure	Estimated Fractional Intake*
Particle accelerators	Adsorbed gas and HTO	Unspecified	$<2\times10^{-6}$ per employee
Tritium gas processing	Tritium gas	Special glass vessels and glove boxes	$<10^{-10}$ per employee
Reactor operations	HTO in reactor coolant	Primary coolant system enclosure	10^{-5} (based on concentration in coolant) 10^{-10} of activity in reactor

*Total number of employees involved in these scenarios is not currently known.

As seen in Table D-1, the fractional intake based on the amount of tritium material in process for different situations varied from a high of 10^{-2} to a low of 10^{-10}. In the latter (maximizing) case, the intake fraction was for the highest exposed individual who was involved in a spill of approximately 73 millicuries (mCi) of tritiated water on the benchtop of a hood that was cleaned up. The worker involved in the cleanup was only wearing a set of gloves for personal protective equipment, and so it is likely a significant portion of the intake was caused by absorption through the skin. Several of the remaining examples were bounded by, or within range of, the NIOSH-assumed fractional intake value of 10^{-5}.

Brodsky 1980 also presents four situations that involved elements other than tritium. Table D-2 summarizes the estimated fractional intakes from these scenarios.

Table D-2. Summary of Fractional Intake Data from Brodsky 1980

Case Description	Fraction Deposited
I-131 escaping from open vial in hospital environment (145 mCi process)	1.2×10^{-6}
Technician mixed AmO_2-Au powder, compacted, sintered, rolled into foils over 3–5 years (9 Ci in process)	2×10^{-7}
Two technicians cut into Ir-192 pellets with lathe inside hot cell, hot cell ventilation off (2,000 Ci in process)	2×10^{-7}
Two technicians cut into Ir-192 pellets with lathe inside hot cell, hot cell ventilation off (75 Ci release)	1.4×10^{-7}
Hospital worker opening vial of I-131 for therapeutic administration (60 mCi in process)	8×10^{-7}

As seen in Table D-2, each of the five situations involved other radionuclides that were sufficiently bounded by the NIOSH assumed fractional intake value of 10^{-5}.

It should be noted that NUREG-1400 refers to the fractional intake factor of 10^{-6} as a “rule of thumb” and that to arrive at a more realistic estimate of the intake, several modifying factors should be applied, including the release fraction (R), confinement factor (C), and the dispersibility (D) of the material. The release fraction accounts for the physical and chemical properties of the material being evaluated. The confinement factor accounts for the configuration of the facility’s engineering controls where the material is being handled (such as a hood or glovebox). Finally, the dispersibility accounts for the physical and chemical processing of the material being evaluated (such as heating or grinding).

Using these modifying factors, a more realistic estimate of the intake can be calculated using the following formula:

$$\text{Estimated Intake} = (\text{Quantity of Material in Process}) \times (R) \times (C) \times (D) \times (10^{-6})$$

Table D-3 provides typical values for each of these modification factors. As seen in the table, application of the release fraction and confinement factor in the above equation will either not change or decrease the effective fractional intake below 10^{-6}. Application of the dispersibility factor will either not change the intake estimate or potentially increase the intake by a factor of 10 if cutting, grinding, heating or chemical reactions are occurring. However, it is likely that any such operations would occur (at a minimum) inside a vented hood, which would effectively cancel out the dispersibility factor. Therefore, based on the above equation, the fractional intake would only increase above 10^{-6} if a gas/volatile material were being handled in the open air while performing activities that increase the dispersibility.

Table D-3. Typical Values for Modification Factors Used in Arriving at a More Realistic Intake Based on the Quantity of the Material in Process and the Fractional Intake Factor (10^{-6})

Modification Factor	Description	Value
Release Fraction	Solid Material	0.001
	Non-Volatile Powders and Liquids	0.01
	Gases or Volatile Materials	1
Confinement Factor	Glovebox	0.01
	Vented Hood	0.1
	Open Air	1
Dispersibility	All Others	1
	Cutting, Grinding, Heating, or Chemical Reactions	10

The fractional intake method of estimating potential internal doses described above assumes normal routine conditions over a long period of time (i.e., 1 year) and does not necessarily address acute incident scenarios. However, Brodsky 1980 (p. 993) does discuss a fractional intake method under accident conditions:

> *Frank et al. …have found from data collected in their survey that usually no more than 10^{-6} of material in process will enter the body of a worker in the event of a release caused by an explosion or other dispersing incident. Even for volatile materials at elevated temperatures, no more than 10^{-5} of the material in process entered the body after release. In several accident cases involving Pu, Am and Ir, which the author evaluated at the University of Pittsburgh whole-body counter, estimated fractional intakes of material in process were 10^{-6} or less, even for workers handling material at arms' length at the time of the accident….*
>
> *However, in the case of specific tritium or iodine compounds that may penetrate protective gloves and enter the body through the skin, there may be special circumstances where higher fractional intakes are possible.*

Clearly, the amount of "material in process" considered for an accident or incident scenario would be significantly less than the amount of material considered during non-upset conditions over a long period of time (in the RPRT-0090 scenario, 1 year). Although the above quote notes that incidents involving iodine could produce higher acute fractional intakes, NIOSH used a different method to evaluate iodine, described in Appendix C of RPRT-0090.

CONCLUSION

Review of NUREG-1400 and the underlying documentation indicates that under routine conditions, the potential for intake of radioactive material is generally less than 10^{-6} multiplied by the amount of material in process. When the appropriate modification factors are included (release fraction, confinement factor), the value is likely much less than 10^{-6} under normal conditions. Estimates of intakes based on incidents or accidents are not specifically accounted for in this method and would have to be added to any routine estimate. Without specific knowledge of any incident conditions, frequency, and material amounts, it is difficult to assess how this might affect the total fractional intake estimate. However, SC&A agrees that use of a fractional intake factor of 10^{-5} represents a reasonably conservative methodology to estimate potential intakes to unmonitored workers under routine conditions in which off-normal occurrences are controlled and documented.