Preliminary Draft
For the Mound Work Group Review

MOUND INTERNAL DOSIMETRY DATA ADEQUACY AND COMPLETENESS

Prepared by

S. Cohen & Associates
1608 Spring Hill Road, Suite 400
Vienna, Virginia 22182

Saliant, Inc.
5579 Catholic Church Road
Jefferson, Maryland 21755

June 2010

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TABLE OF CONTENTS

ABBREVIATIONS AND ACRONYMS

ABWRH	Advisory Board on Radiation and Worker Health
ANL	Argonne National Laboratory
CEDE	Committed Effective Dose Equivalent
D&D	Decontamination and Decommissioning
DOE	U.S. Department of Energy
Dpm	disintegrations per minute
EEOICPA	Energy Employee Occupational Illness Compensation Program Act
ER	Evaluation Report
MCC	Monsanto Chemical Company
Mrem	millirem
MWP	Maintenance Work Permit
NIOSH	National Institute for Occupational Safety and Health
ORAUT	Oak Ridge Associated Universities Team
ORNL	Oak Ridge National Laboratory
PAAA	Price Anderson Amendment Act
pCi	picocurie
PMC	Plutonium Molybdenum (pg. 47)
POC	Probability of Causation
PPE	Personal protective equipment
R&D	Research and Development
RAM	Radiation Assessment and Measurement
RCG	Radioactivity Concentration Guideline
rem	roentgen equivalent man
RWP	Radiation Work Permit
SEC	Special Exposure Cohort
SC&A	S. Cohen and Associates (SC&A, Inc.)

1.0 INTRODUCTION

In its *Draft Mound SEC Issues Matrix* (SC&A 2008a, March 17, 2008), SC&A noted that the National Institute for Occupational Safety and Health (NIOSH) Evaluation Report (ER) has not adequately addressed how dose reconstruction would be accomplished for exotic radionuclides (i.e., other than plutonium, polonium, elemental tritium, and tritiated gas). Concerns have been raised regarding data adequacy for internal dosimetry; specific bioassay data are non-existent for many of the years when exotic radionuclides were handled at Mound.

The matrix items related to "exotic radionuclides" include:

- Matrix Item 1 – Exposure to radium, actinium, and thorium starting March 1, 1959
- Matrix Item 3 – Exposure to transuranium radionuclides (Am-241, Cm-244, Am-243, Np-237) other than plutonium
- Matrix Item 4 – Exposure to U-232, U-233, U-234, U-235, U-236, and U-238
- Matrix Item 7 – Fission and activation products
- Matrix Item 8 – Other radionuclides (Pa-231, La-140, Ba-140, Ca-45, Fe-55, Fe-59, Co-60, Zn-65, Sc-46, Hg-203, Ag-110m, Bi-210, Cs-137, Xe-131, Kr-85, I-131, etc.)

At the July 14, 2008, work group meeting, in response to Mound Matrix Items 1, 3, 4, 5, 7, and 8, NIOSH/Oak Ridge Associated Universities Team (ORAUT) compiled the *Major Isotopes, Process, Material, and Bioassay Road Map for the Mound Laboratory* (hereafter, referred to as the "roadmap;" ORAUT 2009) to characterize radionuclide usage and associated bioassay coverage. Information for the roadmap was derived from the *Mound Site Radionuclides by Location* (King 1995), supplemental incident reports, and decontamination and decommissioning (D&D) era Radiation Work Permits (RWPs). Although considerable effort was put into the roadmap, NIOSH indicated during the January 5–6, 2010, Mound Work Group meeting that the King document (King 1995), and thus the roadmap, conservatively lists all radioisotopes that **might have been** used in an area, for the purpose of determining bioassay requirements in the D&D era. The roadmap was not intended as a tool for dose reconstructors, but was prepared to assist the Work Group in evaluating the SEC petition (ABWRH 2010). According to NIOSH's statements at the work group meeting, the roadmap should not be construed as representative of the episodic nature of radionuclide handling, and they were aware of no situations where a potential exposure existed without the availability of bioassay.

At the January 5–6, 2010, work group meeting, SC&A was asked to provide specific examples of intervals in which a potential for exposure to radionuclides existed in the absence of internal monitoring. Several examples of such scenarios have been provided herein and are summarized in Table 1 of this report. Clarification on the purpose of the King document (King 1995; Mound 2001) is also provided, since this is a critical part of the characterization effort at the Mound Plant. Finally, additional information supporting radionuclide presence and potential exposure are provided for exotic radionuclides.

POINT OF CLARIFICATION: As a significant source of information for SC&A's Mound Matrix items and for NIOSH's roadmap (ORAUT 2009), Technical Manual MD-22153, *Mound Site Radionuclides by Location*, and its appendices have been referenced throughout the Special Exposure Cohort (SEC) petition evaluation and technical review, but with some inconsistency. The current version of the manual in SC&A's possession is Issue 3, approved by D. Morris on March 22, 2001. Changes incorporated with this revision are relatively minor; major sections of the document retain their identity, authorship, and effective dates from prior issues. For example, the Introduction retains its effective date of February 11, 1998, and Section 1, "Primary Research and Production Buildings," remains intact as Section Issue 1, Wayne C. King, effective date June 22, 1995. Within this paper, SC&A references Mound 2001 as representative of the entire manual (including Appendices). Since NIOSH/ORAUT references the document as King 1995 in the roadmap, SC&A retains this reference only when referring to its role as a roadmap source. Because the original section authorship, issue number, and effective date are retained in the current issue, SC&A assumes that the contents of "Eckman and King 1995" and Section 1 of "Mound 2001" are equivalent.

2.0 SUMMARY OF NIOSH POSITION ON DOSE RECONSTRUCTION

The *NIOSH Evaluation of Data Adequacy and Completeness Issues at Mound Laboratory* (NIOSH 2009) provides a detailed response to SC&A's concerns regarding the presence of exotic radionuclides at Mound and gaps in personnel monitoring data. NIOSH's position is summarized in NIOSH Response 1-1 of the document:

> *The presence of various radionuclides at Mound is not sufficient to establish an exposure potential for each radionuclide. Several conditions must be met to constitute and [sic] exposure potential, including but not limited to:*
>
> *(1) the radionuclide must be present in sufficient quantity to present a dosimetrically significant hazard;*
> *(2) the radionuclide must have a sufficiently long effective half-life to present a dosimetrically significant hazard;*
> *(3) the radionuclide must be in physical and chemical forms that presents an exposure potential;*
> *(4) the radionuclide must be handled in a way that presents the potential for intake (e.g., access to the breathable atmosphere).*
>
> *The predominant internal exposure at Mound was to alpha emitters and tritium. Beta and gamma emitters played a very minor role in Mound activities, and for the most part exist in trace quantities in research and production-scale operations.*

Furthermore, NIOSH indicates that SC&A's concerns are at odds with the *History of Mound Bioassay Programs* (Meyer 1992). NIOSH/ORAUT (NIOSH 2009) provided the following quote, attributed to Sheehan (2009b), as a description of the dose monitoring practices of the health physics group:

> ***For each of the radionuclides involved, Mound's RAM section had an internal radiation dose assessment support program.*** *All of these programs employed urinalysis as the source of data to estimate internal radiation exposure and evaluate the integrity of nuclear work station containment systems, process, procedures, techniques, ventilation management as well as employee job training and work attitudes. Radiological safety was made a pervasive, controlled atmosphere in which radioactive material was handled." (Sheehan 2009, emphasis added).*

SC&A has noted that the document referenced by NIOSH/ORAUT does not include this statement. Further conversations with Sheehan indicated that **the only** document provided to NIOSH from him in 2009 was *Mound's 24-Hour Urinalysis Program* (Sheehan 2009b). Either NIOSH/ORAUT has misquoted Sheehan, or their reference is incorrect.

NIOSH also maintains that gross alpha results can be used to bound the dose for all alpha emitters at Mound where radionuclide-specific data are not available. Using the gross alpha result, NIOSH would attribute dose to the radionuclide that gives the most claimant-favorable organ dose for the organ in question (the one with cancer for which a claim is being submitted), with case-specific information being considered when available (e.g., where a worker worked). This approach would be used to bound doses for all isotopes of actinium (via daughters), americium, curium, protactinium, neptunium, radium, thorium, and uranium.

Where bioassay procedures are unavailable for radionuclides handled at Mound, NIOSH repeatedly argues (NIOSH 2009):

> *… processes with these materials were episodic in nature, not ongoing, therefore monitoring would only be required when activity was ongoing, or a significant residual exposure potential existed.*

At the work group meeting in January 2010, NIOSH stated that they are aware of no specific examples in which Mound operations posed a potential for exposure to radionuclides outside the scope of the Mound bioassay capabilities, and that such an assertion on SC&A's part is contradictory to Meyer's first-hand description of Mound's radiation protection program. They referred meeting participants to their written response, specifically Attachment A, "Major Mound Source Terms and Radionuclides of Concern" (NIOSH 2009), and the table on page 24 of NIOSH 2009, "Response to Items in 'Application of Bioassay Methods by Radionuclide' Table in 'Data Completeness' Not Covered in the Discussion of Gross Alpha/Radionuclide-Specific Alpha Bioassay." Attachment A lists major Mound source terms, constituent radionuclides, and the major radionuclides of concern for each source term; it does not identify the locations or time periods in which these source terms produced exposure potential at Mound. The second document responds to specific issues or source terms raised in *Mound Internal Dosimetry Data Completeness* (SC&A 2009b).

The current position held by NIOSH is that they can reconstruct dose received by all workers at Mound who were potentially exposed to exotic radionuclides, with the exception of radium, actinium, and thorium exposures from October 1, 1949, to February 28, 1959. Additional consideration is being given to radon exposures in R-Building and SW-Building; however, a formal determination was not issued as of the date of this report. NIOSH maintains that gross alpha analysis can be used in lieu of radionuclide-specific bioassay for all alpha emitters present at Mound. For other radionuclides, they maintain that they were only handled in small quantities and/or that there was no potential for exposure.

3.0 RADIOLOGICAL HAZARDS AND EXPOSURE POTENTIAL

NIOSH/ORAUT's roadmap (ORAUT 2009) was compiled primarily to characterize radionuclide usage and associated bioassay coverage in response to SC&A's Mound Matrix items. Information for the roadmap was derived from the *Mound Site Radionuclides by Location* (King 1995), supplemental incident reports, and D&D-era RWPs. An additional source, *History of Mound Bioassay Programs* (Meyer 1992), was used to link potential exposures with bioassay techniques available at Mound, taking into consideration the quantities of radioactive material and established engineering and administrative controls. The roadmap columns include Location (building and/or room number), Program/Process, Time Frame, Radionuclides and Related Compounds, Quantity, Material Characteristics/Information, Bioassay Method, Exposed Individuals (number of individuals exposed), and Ref [Reference]. Incidents and other process information are listed under Material Characteristics/Information. RWPs from the D&D era (1995–2001) are listed as separate line items.

A revision to the roadmap, provided to the work group in July 2009, includes or references information contained in *Mound Site Radionuclides by Location,* Appendix B (hereafter referred to as Appendix B, King 1998). Information regarding classified operations and activities for R, SW, SM, T, WD, and WDA buildings is discussed only in Appendix B of the King document (King 1998). Appendix B describes the extent of radionuclide processing activities, the chemical forms handled in processes and programs, and the impact of weapons program-related activities on worker exposure. This information is a significant factor in fully appreciating the processes occurring in R, SW, SM, T, WD and WDA buildings, but it is only accessible to individuals possessing appropriate clearance and a need-to-know. Information in Appendix B (King 1998), as summarized in the roadmap, has direct relevance to the handling, research and development (R&D), and processing of exotic radionuclides at the Mound Plant.

During the January 5–6, 2010, Mound work group meeting, NIOSH indicated that the King document (King 1995) listed all isotopes that might possibly have been present in specific areas in order to support appropriate monitoring of D&D operations. NIOSH repeatedly emphasized that an isotope's presence is not sufficient to constitute an exposure potential. They offered several examples in which activity could be present without posing a potential for internal exposure (e.g., sealed sources, fixed and painted contamination, drums in a burial ground). This approach essentially dismisses *Mound Site Radionuclides by Location* and the roadmap as evidence of exposure potential, and serves to mitigate, on their part, SC&A's concerns about the adequacy of internal monitoring data. As evidenced in the following discussion, NIOSH maintains that gaps in bioassay data simply reflect intervals when there was no exposure potential, and requests that SC&A identify specific situations in which potential exposures were not appropriately monitored (ABRWH 2010, pp. 82–83):

> *MS. ROBERTSON-DEMERS: Bioassay capabilities do not equate to actually collecting samples.*
>
> *DR. ULSH: So you're saying that there were situations where they should have collected samples and they didn't?*

MS. ROBERTSON-DEMERS: Yes.

DR. ULSH: Okay. Let's talk about those. Give me some examples.

MS. ROBERTSON-DEMERS: I've given you an entire table where these things were noted as being handled in the Road Map, and there's no coverage of bioassay.

DR. ULSH: Okay. So we're talking about the Road Map now. Again, as we discussed earlier, the Road Map lists any element, any radionuclide that could have possibly been in a particular room, not that there was a confirmed presence of it, but just that it was possible.

And again, the piece that you are not considering is the exposure potential. If I walk through -- I'll use the same example again -- if I walk through a room, if I even stored in a room sealed sources, that does not equate to an exposure potential and it does not equate to a need to do bioassay.

If the Road Map is your basis, you are misinterpreting the Road Map.

In contrast to the statement above, the introduction to *Mound Site Radionuclides by Location* (the primary data source supporting the roadmap) outlines the following objectives (Mound 2001, emphasis added).

> *There are three major objectives of this document. The first is to **identify the radionuclides used** in each of the rooms for each of the buildings at Mound. This will include a listing of all compound forms of each radionuclide **for specific time frames because of specific programs**, which are also identified.*
>
> *The second objective is to determine information about a given radionuclide which would **facilitate internal dose assessment**. This includes the compound forms with which the radionuclides are associated, information to indicate the probable lung solubility class, the relative abundance of radioisotopes in each room for a given time frame, and the particle size.*
>
> *The **third objective** is to compile all information about ground surveys and core sample results to indicate possible intakes at specific locations **as a result of D&D** and other work performed.*
>
> *When doing either a **historic or current internal dose assessment for an individual**, the records do not always indicate the details of an incident or a chronic condition. **By knowing the approximate date and location of an exposure, this document can be used to determine which radioisotopes existed in the area**. Since chemical, temperature and processing, and particle size all can affect the lung solubility class, this information is of great help in determining*

> *the parameters used in the biokinetic model which predicts the internal dose from lung deposition. This information is also valuable for uptakes resulting from ingestion or wounds.*

Furthermore, the overview for each building within Mound (2001) indicates that dates provided in the document are representative of usage of radionuclides and not necessarily residual radioactivity (Mound 2001, emphasis added). While not every usage of radionuclides poses a potential for internal exposure, SC&A maintains that dates listed in the King document (Mound 2001) represent actual usage of radionuclides in active programs.

> ***All dates represent the duration of actual usage*** *of radioisotopes in their respective projects. It is clearly understood that* ***residual amounts*** *of all radioisotopes referred to in each room may still be found in floors, walls, and ceilings and* ***should be considered, up to the present, in every case for decontamination work****.*

During a recent interview with former workers, SC&A had the opportunity to discuss *Mound Site Radionuclides by Location* with one of the document contributors. This individual reaffirmed that the dates listed in the document represented the time period in which radionuclides were used, as opposed to being present as residual radioactivity. This individual clarified that the designation of major and minor radionuclides in the *Mound Site Radionuclides by Location* was determined based on the relative amount (i.e., quantity) of each radionuclide handled or processed. Although efforts have been made to contact the primary author, SC&A has been unable to locate him. However, the interviewee's interpretation may be supported by considering the following assessments of hazards present in Room SW-22, the "New Cave:"

- Describing the Cotter Concentrate program, King states, "Thorium-230 was the major radionuclide of concern with over 95% of the activity" (Mound 2001, Section 1, p 146 of 320).

- Another author reached a different conclusion, based on considerations other than activity: "Of all Cave Area radionuclides Pa-231 is of greatest concern. Although only a few milligrams are produced and stored each year a potential for exposure exists because of an RCG of 1×10^{-12} µCi/ml in air, only half that of Pu-238, and operations that do not provide for continuous containment" (Stought 1979).

NIOSH's characterization of the information in the King document (King 1995) seems to contradict statements drawn directly from the document. Interpreting King (King 1995) as listing all isotopes that "could have possibly been in a particular room" (ABRWH 2010) disregards the compilers' efforts to associate active projects with specific dates, locations, and chemical forms in support of historic and current internal dose assessment. Furthermore, SC&A has found additional references that substantiate process descriptions and time periods provided in King (Mound 2001). NIOSH maintains that exotic radionuclides (other than plutonium, polonium, and tritium) were handled episodically. In contrast, King (Mound 2001) identifies ongoing operations and research involving exotic radionuclides over extended periods of time;

their usage in processes, research, and analytical programs extended from the 1940s to closure of the Mound facility. Since *Mound Site Radionuclides by Location* claims to represent actual usage in specific rooms associated with specific programs, and since several of these programs can be substantiated through other sources, it seems unreasonable to dismiss the information on the basis of an unsubstantiated interpretation.

Table 1 lists several examples of programs and processes in which unencapsulated exotic radionuclides were handled or released in multiple locations throughout the Mound site. The table specifically identifies radionuclides and intervals for which no personnel monitoring data have been located to date. In some cases, there was a total absence of bioassay; in other cases, there were personnel who were not monitored handling material. Regardless of the bioassay **capabilities** available at Mound to detect internal uptakes, the availability of monitoring **data** more accurately represents the adequacy of personnel monitoring at Mound and the feasibility of dose reconstruction.

Furthermore, "dosimetric significance" must be considered in the context of worker compensation, rather than an operational internal dosimetry program. SC&A acknowledges that factors such as the activity of material handled (pCi, mCi, Ci, etc.), the established engineering and administrative controls, and the physical and chemical forms of material impact the dosimetric significance of an exposure potential. These factors must be taken into account through all stages of operations and handling, from receipt of radioactive material at the site through final disposal of radioactive and mixed waste. Processes which result in the separation of radionuclides and subsequent concentration of particular radionuclides, either intentionally or unintentionally, must also be considered. At times, separated material in the waste stream may contain higher concentrations of radionuclides than those identified for active process and operations activities, as outlined in *Mound Site Radionuclides by Location* (Mound 2001; King 1995), and thus in the roadmap (ORAUT 2009) prepared by NIOSH.

Although NIOSH has identified several factors that influence the potential for intake, they have not provided adequate information that would support an objective determination of the "dosimetric significance" of exotic radionuclides. Without this information or clear guidance from NIOSH, SC&A found it necessary to establish a working definition of "dosimetric significance" for evaluating exposure scenarios at Mound. The following factors were taken into consideration.

- Neither the Act nor the associated rules for the EEOICPA program define a *de minimus* dose for the compensation program.

- The DOE presently requires bioassay submission based on a 100-mrem dose criteria for an operational dosimetry program.

- During the Pre-1989 Dose Assessment Project, MJW Corporation identified H-3, Po-210, Ra-226, Ac-227, Th-228, Th-230, Pa-231, Th-232, U-233, U-234, U-235, U-238, Pu-238, Pu-239, Am-241, and Cm-244 as nuclides of dosimetric concern. This determination was based on a review of the bioassay data and procedures available at Mound, with a focus

on those workers potentially receiving an internal dose of 20 rem Committed Effective Dose Equivalent (CEDE) or greater (MJW 2002a).

- Doses of less than 100 mrem can impact the compensability of some claims, taking them from non-compensable to compensable.

Since Po-210, Ra-226, Ac-227, Th-228, Th-230, Pa-231, Th-232, U-233, U-234, U-235, U-238, Am-241, and Cm-244 were considered of importance in evaluating collective internal doses of 20 rem or more, these isotopes are clearly of "dosimetric concern" at dose levels effecting compensation. A number of radionuclides that were present at Mound and lack monitoring data were not considered in the Pre-1989 Dose Assessment Project. Many of the beta- and gamma-emitting radionuclides fall in this category. NIOSH has provided no objective evidence supporting their statement that these and other secondary and impurity radionuclides are of dosimetric insignificance, as NIOSH has claimed throughout *NIOSH Evaluation of Data Adequacy and Completeness Issues at Mound Laboratory* (NIOSH 2009) and repeatedly stated in work group meetings. Essential information for these exposure scenarios would include the relative activity of radioactive material handled, the ratios of "secondary radionuclides" to "primary radionuclides" in process and waste streams, and the engineering and administrative controls utilized to prevent exposure to "secondary radionuclides." NIOSH responses to SC&A's concerns regarding these issues have not been adequate to date.

From SC&A's perspective, a technical review involves a critical investigation of a program's effectiveness based on available documentation. The task is to **evaluate** the program, not to presume that it was comprehensive and effective. NIOSH appears to take a different approach. They have dismissed *Mound Site Radionuclides by Location* as insufficient evidence of exposure potential, not by offering new documentation that more clearly defines specific times and processes, but by re-interpreting the document's intent in direct contradiction of its internal claims. Data adequacy cannot be assessed without working definitions of exposure potential and dosimetric significance, along with mutually acceptable evidence defining when these circumstances were present. SC&A contends that *Mound Site Radionuclides by Location* (Mound 2001) is the best available comprehensive characterization of radionuclide usage at Mound over its operational period. SC&A sees no reason to waste further time and resources searching for documentation to substantiate or replace it. "Episodic use" could certainly explain fluctuations in the number of bioassay samples for a particular radionuclide from month to month or year to year, but this argument cannot explain away years (even decades) without specific bioassay data when available evidence indicates active usage of the isotopes.

3.1 POTENTIAL FOR EXPOSURE

During the January 5–6, 2010, meeting, NIOSH indicated that they see no evidence of potential exposures from exotic radionuclides during years when personnel monitoring was absent. Further information is provided in *NIOSH Evaluation of Data Adequacy and Completeness Issues at the Mound Laboratory* (NIOSH 2009). SC&A was asked to provide specific examples where there were potential exposures in the absence of internal monitoring. As with "dosimetric significance" in the preceding section, SC&A was obligated to adopt their own working

definition of "exposure potential" in the absence of specific direction from NIOSH or the work group. The operational exposure potential, as defined in this review, is exposure to unencapsulated radioactive material. Other considerations taken into account include:

- Positive contamination surveys
- Positive air sampling results
- Radionuclide quantities (where available)
- Bioassay monitoring requirements in the 1990s/2000s for the same area
- Radionuclides with half-lives >30 days

The effectiveness of administrative and engineering controls is another significant factor in defining potential exposures. As with other sites, Mound had numerous incidents indicating insufficient engineering and administrative controls. Repetitive incidents and poor radiological control design are indicative of a potential for exposure. Mound has a demonstrated history of radiological control failures site wide. Concerns regarding glovebox design and materials are discussed in detail under the polonium impurities section. Several additional examples are provided below. SC&A's purpose in citing these examples is to demonstrate weaknesses in the **general radiological controls** implemented at the Mound plant. The examples demonstrate inadequacies affecting facilities, procedures, containment, contamination control, implementation of corrective actions, and respect for posted hazards. While many of these examples took place within "primary" programs, such as plutonium production, where routine internal monitoring was in place, SC&A has no reason to assume that controls for "secondary" separations, research, and waste programs were so superior as to eliminate exposure potential for the workers involved.

3.1.1 Engineering Controls

Problems with hoods at Mound being improperly exhausted were discovered as late as 1977 (Kosuszek et al. 1977).

> *On October 17, 1977 Safety was notified by Engineering of their discovery that an exhaust duct from two fumehoods located in E-l07 was tied into the building's general recirculating room air system. This could be a risk of potential exposure to building occupants. From the facts found in this investigation, the committee also recognizes that other improper tie-ins may exist, especially wherever building ventilation systems have been modified, and therefore, recommend that an inspection program be implemented.*

At times, dryboxes became pressurized, which could lead to loss of containment and spread of contamination. Such an incident occurred in R-120 after workers changed an argon tank used to provide an inert atmosphere in the drybox. Pressurization of this drybox caused a glove to be blown off, resulting in the contamination of workers, as well as five visitors in the room. "In addition to Room 120, Room 120B, the west corridor, and Rooms 155 through 159 became hot" (Freeman et al. 1960).

Airflow at Mound was not always towards the area of maximum contamination potential. This was shown by a 1963 incident in R-149. The investigation into this incident found that an argon supply change pressurized a drybox and blew off a glove. In this event, "a slight positive pressure in room 149" caused some contamination spread to Room l50-5l and corridor 5 (Butz et al. 1963).

The flow control of Mound's dryboxes does not appear to be up to modern standards, as indicated by the following incident (again in R-120). In response to an internal uptake of approximately 15 rem that occurred during routine operations, the investigating committee concluded that the incident likely occurred when personnel were using a fumehood to introduce samples into a drybox line. The committee's findings included the following concerns (Wainwright 1973):

- *This fumehood is serviced by a forced air ventilation supply line, and the airflow rate into the hood from the room was observed to be substandard and variable. The supply line valve was found to be partially opened. These factors probably combined to provide a short reversal of airflow in the fumehood which resulted in a release of fine radioactive particulates to the laboratory.*

- *A second possible source was a small hole found in one of the glovebox gloves.*

- *A contributing factor to the exposure of the personnel was the turbulent airflow patterns caused by the room air supply systems which tended to spread the airborne contamination throughout the room.*

At times, facilities did not undergo adequate re-design to accommodate new materials being handled. According to an investigation report of a spill in R-149 (Bigler et al. 1960):

> *It was obvious from this investigation that the facilities for performing the work done in R-149 are inadequate. Contamination levels have been high in this lab at various times since this program began. It has been standard practice to require the wearing of respirators. The closed hoods used in this lab were not designed for the material being handled in them and do not afford proper protection or permit good handling techniques.*

Lessons learned from incident investigations did not necessarily result in effective, comprehensive corrections. A pressurized drybox incident in SW-13 in 1970 prompted the following observation (Madding and Carfagno 1970):

> *There is a striking similarity between this incident and one that occurred on the R-l27, 149 inert system on February 27, 1968. Applying the intent of the recommendations of the incident report to the SW-13 inert gas system, especially #4, would have prevented the SW-13 incident.*

The SW-13 incident report also mentioned that Mound had not characterized a material that had been released to the room from the incident (Madding and Carfagno 1970).

> *Room decontamination and monitoring were complicated by the metallic compound released from the vacuum boxes. While the use of respirators and protective clothing which occurred is considered reasonably safe, the hazards of this material have not been defined as indicated by discussions with the involved personnel.*

Although the recommendations from the R-127 and R-149 incident report were considered appropriate to have prevented the event in SW-13, it is interesting to note that the inert system in the R-Building labs experienced an extreme pressurization incident only 1 month after the SW-13 report. Over a period of 4 days in November 1970, pressurization of an inert atmosphere hood line cracked two hood fronts in R-127 and caused a hood front blowout in R-149. The 3/8" Lucite hood front ruptured into several sections, with pieces blown up to 10 feet from the hood. A primary contributing factor was that, "Budgetary considerations forced using a common purifying and pumping system for two separated installations with 'too many marginally safe procedures on this system.'" A researcher troubleshooting the system pressure concern selectively closed off each hood in R-127 while noting pressure changes on a Photohelic gauge. "Unfortunately, closing off this last hood deprived both the hood line in R-127, as well as in R-149, of any relief valving since the bubbler in R-127 is in the Photohelic line and the bubbler in R-149 [recently removed and sealed off due to a silicone oil leak] had not been replaced." Multiple corrections to the system, including independent bubblers for each box line and an independent purification system for the R-149 hood line, were recommended by the investigation team (Fauble et al. 1970).

The gloves used in dryboxes often had failures, which could lead to the spread of airborne contamination. In the following example, the incident investigation committee noted that the dryboxes were not kept in an orderly state (a great deal of trash was noted), and this incident was compounded by the lack of a functional contamination survey instrument in the area (Abrahamson et al. 1964).

> *At approximately 12:20 a.m. on October 1, 1964, the air monitor registered a high level of contamination in the R&R Building. The high level was first believed to be caused by the release of activity through a hole in a glove which was accidently produced by [name redacted] while working in an alpha box on recoverable trash. The contamination of R&R Building was caused by an accidental puncture of a glove by [name redacted].*

Another glove failure occurred on June, 29, 1970, resulting in a release of plutonium-238 activity in the B-2 laboratory of the PP-Building. This particular incident was classified as a "near miss," because nose wipes indicated potential internal uptakes. "The primary cause of this contamination release was thought to be a hole, too small to be detected by the unaided eye, in one of the glovebox gloves in either glovebox 041 or 042" (Freidline et al. 1970).

An incident at SM-35 occurred while the integrity of a glovebox glove was being checked (Amos et al. 1963).

> *In alpha box 7 a small pinhole was noted in the left hand glove and [name redacted] slowly brought his hands from the gloves to avoid spreading contamination. However excessive talc (used to aid in "sliding" in and out of the gloves) in the glove caused a puff of talc to come out into the room. Previously cold areas of the room were now contaminated including the floor, the hood fronts, and other alpha box gloves. The alpha air particulate monitor, 10–12 feet away showed an increase of 30,000 counts per minute.*

In some cases, glove failure was a result of poor maintenance, rather than a specific accident or event. A glove failure in SM-59A was discovered when an air monitor alarmed when a worker removed his hands from Box 8. Surveys indicated that Box 8 was not the source; previous work in Box 14 had resulted in the spread of contamination. Some personnel who had already left the work area were requested to return to Room 59A for survey by health physics and were found to be contaminated. Among the conclusions of the investigating committee (Mershad et al. 1966):

> *The direct cause of the incident was the opening up of a crack in the upper left hand glove of box #14. After the glove was changed, a visual inspection by [name redacted] revealed a crack about l/l6th inch in length along the inner side of the glove thumb. It was described as typical of a hole which develops from the deterioration or strain of the glove material over a period of time, and there was no evidence that the glove was cut or punctured.*

Drybox seals were also known to fail. For example, an incident in PP-Building A-1 laboratory was categorized as a "near miss," because "one Electronics Technician received a high nose wipe" (Kell and Combs 1971):

> *The contamination levels in Gloveboxes R-007 and R-006 and the subsequent contamination to [name redacted] was the direct result of an unsealed service penetration into Glovebox R-007. This penetration had been previously sealed but the sealant obviously had deteriorated to the point of opening up again.*

3.1.2 Work Practice Controls and PPE

Work practices and PPE in low-activity environments could be less stringent than those used in production areas. A contaminated acid burn in an analytical lab in PP-Building resulted from a technique and PPE that were not adequate for handling concentrated acid solutions. A health physics surveyor monitoring an [**redacted**] technician used his hand (with a surgical glove) to dip a wipe into a spill of 8N nitric acid. The acid penetrated the glove, resulting in a contaminated burn. An inter-office communication from Health Physics recommended that "orange" latex gloves, rather than surgical gloves, should be used for fumehood work, noting that, "Analytical is the only area where surgical gloves are used." A secondary correction was to

write a procedure specifying the use of tweezers or tongs in addition to heavier gloves when monitoring work with liquids (Davis 1974).

An unfinished and forgotten maintenance effort appeared to cause a 1963 contamination incident in SM-Building. The accidental disturbance of a plastic bag that had been used to seal what was believed to be an old vacuum line resulted in widespread contamination of the low-risk side of SM-Building. High levels of contamination spread to Rooms 21, 10, 38, 39, 17, 18, 9, and corridor 40, as well as other adjoining rooms. It must be noted that although this incident involved gross contamination and possible internal uptakes, it was made reportable only on the basis of cleanup costs (Chong et al. 1963).

> *Based on the findings it appeared this line had been cut, then the open end sealed with a plastic bag. Examination revealed a brittle bag, brownish yellow green in appearance. It appeared that solution was present in the bag some time ago and had since dried to form oxide dust. A slight touching of the bag caused it (the bag) to flake and disintegrate. This line was rebagged with three polyvinyl glove port covers. Counts in the immediate vicinity were well over 2 million.*

A metal tritide contamination incident in R-Building went undetected for 2 days. On September 6, 1978, a worker broke a capillary containing "a maximum of 0.3 Ci of tritium as a metal tritide" while attempting to straighten it in the rotating anode fixture. This sample, in addition to five other samples received on three different dates, was incorrectly presumed to be a deuteride sample, rather than a tritide sample. The worker notified two individuals of the break, one of whom returned the call the following day. "Since there was data on the duplicate sample, no request for additional analysis was made. No one expressed any concern about a possible spill." Two days later, two researchers discussing analytical results became aware of the misunderstanding about sample identity; one of these researchers recalled the sample break and requested a survey to investigate the possibility of contamination. Health Physics confirmed a spill and determined that contamination had been tracked throughout the R-Building corridors. Findings emphasized poor communication and labeling, resulting in the sample analysis being conducted in an uncontrolled area without special handling (Pardieck and Sheehan 1978):

- *Verbal communications between requestor and x-ray laboratory personnel did not establish identity of samples and requirement for special handling.*
- *Form 5188, Radioactive Material Transfer Tag, was not attached to the samples and did not accompany the samples to R-Building.*
- *The word tritide written on the request for analysis form was overlooked or misinterpreted by x-ray laboratory personnel.*
- *The analyst did not review request for analysis form prior to performing analysis.*

Ineffective passbox procedures led to a widespread contamination of T-Building. This incident was not detected when it occurred; the contamination was detected during routine surveys at a later time. The investigating committee also noted that no functional alpha survey meter was present in the area at the time of the incident (Heidelberg et al. 1964).

> *Radioactive contamination which was discovered spread widely throughout the "T" Building on June 11 was released from the passbox area of the south cell. . . The most probable method is the accidental drag-out of some of the highly contaminated damp debris left in the box. This damp material was then tracked throughout the building on the shoes of personnel. The spread throughout the building was speeded by the fact that Stairway 13 has been used by large numbers of persons not involved in cell operation; as a traffic route between the first and second floors of the "T" Building.*

A contamination event in corridor 16 of the PP-Building was attributed to lack of a procedure for the work being performed and a lack of communication as to how the task would be accomplished. The event occurred while pipefitters were removing contaminated drain pipes. The investigating committee noted that it was normal practice for workers to select from several different tools to cut or break a 4-inch cast iron soil pipe. The pipefitters on this occasion chose to use a sledge hammer rather than a soil pipe cutter. Lack of operating survey equipment was also an issue in this incident (Bond et al. 1986):

> *If the pipefitters normally make the decision in the field, an adequate number of optional tools should be close at hand and immediately available and guidance on restrictions should be provided. Methods or options, if to be selected on the job, should receive advance approval from the project engineer. All parties involved in the operation need to be made aware of approved methods.*
>
> *There was not an operating survey meter in the area due to portable alpha counter window contamination problems; steps should have been taken to solve the window contamination problem.*

Even routine maintenance activities could, and sometimes did, cause widespread contamination events. For example, the changing of roughing filters on several dryboxes in SM-35 resulted in elevated nose wipes (potential internal uptakes) from 19 people in the low-risk area of the building. The filters were changed out using what is now considered a standard bag out method. It must be noted that at the time, no written procedure existed for performing filter changes; the methods were the result of passed-on knowledge. Airborne contamination was determined to have spread from room 35 into rooms 34, 26, 19, 20, 15, 9, 10, 40, 11, 28, 2, 3, 28, and 38. Because of the rapid spread of airborne contamination, the investigating committee recommended a thorough investigation of the building ventilation system, with the findings or recommendations of such an investigation to be carried out as soon as possible (Meyer 1964).

Waste containers were known to rupture outside of areas considered contaminated. Room 26 in the following example was not normally surveyed, because it was not an operating area. The

trash had produced enough pressure to split open the metal can containing it, and the committee concluded that tracking caused the spread of high levels of contamination (Rogers et al. 1965):

> *At approximately 9:15 A.M. February 26, 1965 a package of hood trash was discovered to be ruptured in an open drum located in room 26 of the SM building. The incident resulted in the release of activity into the room and caused minor external contamination of an AEC visitor and four Mound Laboratory personnel.*

Furthermore (Rogers et al. 1965):

> *Rooms 26, 34 and 19 were severely contaminated (>1,000,000 c/m on the floor); adjacent laboratories and corridors were contaminated to a lessor* [sic]*extent. Most contamination outside of room 26 was probably caused by tracking of evacuating personnel.*

And (Rogers et al. 1965):

> *The ruptured #12 can (marked "burnable 2-24-65") was split down the seam and around the top and bottom. The lid of the inner container (press lid can 5-7/16" diameter × 5-7/8" high) was off and trash was scattered in the drum. The containers were deformed indicating pressure had built-up in the package. The other packages marked "burnable 2-22-65" also contained wet paper and were beginning to bulge slightly.*

Activities other than major production campaigns have been known to produce exposure potential. Even work with sealed sources is not immune from accidents. An actinium spill occurred in R-114 when a source being used for gamma experiments leaked in the safe in which it was being stored. "The source itself consisted of actinium perchlorate in a water solution total volume being 6 to 8 ml., contained in a ground-glass stoppered flask about 10 mL in volume. The flask was contained in a brass cylinder with a threaded stopper, which had been greased." After health physics arrived, an air sample was taken. "The air sample was too hot to count so complete protective equipment was used" (Madding and Carfagno 1970).

3.1.3 Safety Culture

Disregard for radiological posting exacerbated some incidents, as was cited for a spill in R-149 (Bigler et al. 1960):

> *A spill of radioactive material in Room-149 of the "R" Building during the morning of May 19, 1960 resulted in the contamination of the floor of the lab and the adjoining corridors. "Danger - Radioactivity" signs were used to mark off the area of contamination and decontamination of the corridor was started immediately. Despite the placement of signs, some of the personnel in the building continued to track through the contaminated area while it was being decontaminated, thus hampering the clean-up operation. In one case, an*

employee failed to comply with a health surveyor's request not to walk in the contaminated portion of the corridor.

In 1973, the area manager for the Atomic Energy Commission (AEC) expressed concern about workers' disregard for standard procedures in regard to an uptake incident in R-120. He wrote to the Mound director (Wainwright 1973):

I feel it necessary to bring to your attention an attitude of disregard for standard procedures which was clearly displayed at the time of this occurrence. Although this attitude may have had little or no effect on the degree of seriousness of this incident, such laxity of discipline can be expected to increase the potential for subsequent problems, if allowed to continue.

A 1964 contamination incident in SM-35, like several other incidents described above, was exacerbated by lack of a functional alpha survey meter. The incident was detected after the fact by an air monitor alarm just outside the low-risk change room. The change room and shower area were determined to have been lost to airborne contamination when workers were found to be even more contaminated after showering. The investigating committee noted, "The rule that 'No Operating Monitoring Devices - NO WORK' should be strictly enforced." It seems that this rule was routinely ignored at Mound. Another statement in the investigating committee's report raises doubts about the adequacy of air monitoring practices and contamination control with respect to workers' breathing zones: "Four constant air monitors were operating, but the background level of the building was of such magnitude that the warning bells had been turned off or were malfunctioning. The instrument controls were set so that no warning light would flash unless the contamination level reached the highest level recorded by the instruments" (Meyer 1964).

At times, areas were posted to require respiratory protection when work resumed in an area before decontamination from previous accidents was completed. For example, "The development laboratory has been a respirator area most of the time for the past several months due to the material being handled and because of the several radioactive spills which have occurred" (Witzerman et al. 1963).

3.1.4 Explosions and Fires

Contamination was likely to have spread to normally inaccessible areas, such as above ceiling panels, electrical raceways, and crawl spaces, due to a number of incidents involving explosions and/or fires (Fiely et al. 1966).

The Committee investigated an incident which occurred on October 21, 1966, yellow control side, Room 21 of the Special Metallurgical Building. The incident resulted in a release of Plutonium-238 of unknown quantity when a double contained vessel nearly full of drybox sieved material intended for recovery of the isotope exploded and dispersed a quantity of the waste material in the south

portion of the laboratory. . . A large quantity of the material was adhering to the ceiling above the point of explosion.

Room 21 was highly contaminated especially in the south end where the explosion took place. [Name redacted], the Health Physics Supervisor for the "SM" Building, initiated an immediate survey of the entire building. The results indicated that Corridor 40, side Corridor 41, and the south end of Corridor 66 were contaminated.

Another example of an explosion took place in R-116A. It involved a source M-218, a standard 5-curie plutonium-239 unit. The source contained 80.0 g plutonium metal and 39.37 g beryllium metal in a welded tantalum cylindrical capsule. Workers were attempting to re-can this sealed source when it exploded. "Contamination of the room was severe, but was almost confined to R1l6A and R1l6" (Blanke et al. 1960).

An explosion and fire in SM-38 lifted the false ceiling panels across the room. As a result of the explosion and fire, contamination was spread to room 38, corridors 66 and 41, and to a lesser extent, the yellow production areas of the SM Facility. It is likely that contamination spread to the areas above the ceiling panels and to other areas affected by air exchange as a result of the concussion wave of the explosion. The investigating committee requested cessation of "all spent anion exchange resin drying operations" and "a demonstrated safe disposition of the spent nitrated resin" (Ofte et al. 1965).

During the opening of a calorimeter can, a mild explosion caused contamination spread in SM-10. Only one can was open of four, each of which contained about 8 g of plutonium oxide. Significant levels of contamination were detected in adjacent areas, "the corridor outside room 10 and extending the full length of the low-risk corridor, rooms 2, 9, 18 and 21. All of which showed a rise in airborne and surface contamination, low to intermediate levels." The investigating committee could not determine a conclusive cause for the spread of contamination to room 2, but they concluded that it must somehow share an air supply with the corridor (Adams et al. 1962).

4.0 RADIUM, ACTINIUM, AND THORIUM USAGE BEYOND FEBRUARY 28, 1959

NIOSH has acknowledged a single class of employees for which they cannot estimate radiation doses with sufficient accuracy. "NIOSH cannot estimate internal Ra-Ac-Th exposures from the arrival of K-65 sludge in October 1949 through February 28, 1959, when the related area of work was decontaminated and decommissioned and sufficient monitoring was in place" (NIOSH 2007).

Although the major production campaigns associated with Ra-226, Th-228, and Ac-227 occurred prior to March 1959, exposure to these radionuclides continued well beyond the NIOSH-proposed SEC period, whereas the availability of isotope-specific bioassay data dropped dramatically. Actinium and thorium separation/production continued throughout the 1970s. Research and development, waste management, and decontamination activities took place throughout much of Mound's operational history. Collectively, these programs involved direct handling, contamination potential, and known environmental releases. Specific examples include:

- Th-229 extraction from U-233 occurred from 1966 through 1981 (Mound 2001).
- Th-232 was used as a substitute for Pu-238 in R&D and analytical programs in the plutonium processing area from the early 1960s through 1980 (Mound 2001).
- Ac-227 was processed and shipped between 1964 and 1970. It was also used in analytical programs from the late 1960s through plant closure (Mound 2001).
- Cotter Concentrate, processed for rare isotope extraction from 1970–1979, contained isotopes of thorium, actinium, and radium (along with other exotics discussed below). Mound had more than 1,000 drums of this material on site (Stought 1979).
- Thorium ore and sludge from the 1955 Thorium Refinery program remained on site until 1975. This corrosive material deteriorated drums. Re-drumming campaigns, as well as material transfers into and out of the bulk storage facility, occurred from 1955 through 1975. Leaking drums and material transfers produced extensive soil contamination. Hazardous levels of radon and thoron have been found in some facilities (Mound 2001, Abbott 1990, Draper 1994).

In addition to Th-229, Th-230, and Ac-227 extractions identified above, several other radioisotopes were processed at Mound. The Cotter Concentrate program extracted Pa-231, as well as Th-230. Despite relatively low quantities, both Th-230 and Pa-231 were identified as radionuclides of concern. According to Stought (1979):

> *Several hundred grams of this mixture* [Cotter Concentrate] *are extracted and stored each year. The bulk of it, 97%, is thorium-232 which has a very long half-life and so is of little concern. But the remaining 3% is thorium-230 whose RCG for airborne contamination is the same as for plutonium-238, 2 × 10-12 μCi/ml air, and so the mixture must be treated with respect.*

> *Of all Cave Area radionuclides, Pa-231 is of greatest concern. Although only a few milligrams are produced and stored each year a potential for exposure exists because of an RCG of 1 × 10–12 uCi/ml air, only half that of Pu-238, and operations that do not provide for continuous containment.*

Contaminated soils and buildings could potentially expose Mound workers long after operations were discontinued. For example, the corrosive sludge from the thorium refinery project was removed from Building 21, repackaged, and shipped off site in 1974–1975 (Mound, no date). The interior was washed and painted, and the facility was used to store drums of Cotter Concentrate from 1976–1987. According to a 1987 memo, health physics personnel made regular entries into Building 21 to conduct radiological surveys and air monitoring. Shoe covers, smocks, and smoke-checked respirators were required for brief entries (Draper 1987). In 1990, a request to use Building 21 for storage of low-level boxes was contra-indicated, because high levels of Rn-220 (thoron) appeared to necessitate supplied air, which was not available at Building 21. Additional concerns included potential thoron penetration of respiratory protection equipment and inadequate means to detect and quantify potential intakes (Abbott 1990). A D&D characterization report from 1996 states, "Radon levels inside Building 21 have been reported as high as 2.0 Working Levels. The source of this contamination is due to the delivery, storage, and removal of the Bulk Thorium Sludges" (EG&G Mound 1996). Since the conditions in the 1990s were attributed to residual contamination from the bulk storage of thorium sludge, and detection of thoron uptake was not considered feasible in 1990, SC&A assumes that these conditions were present during the intervening years, potentially exposing workers who participated in decontamination efforts, radiological assessments, and handling of the Cotter Concentrate drums.

The ER indicates that in-vitro urine data are the primary source of information to support dose reconstruction, and outlines bioassay program requirements associated with specific projects. For example, a bioassay program for Pa-231 required monthly bioassay for research personnel involved in Pa-231 extractions between 1956 and 1960 (NIOSH 2007); SC&A has located bioassay data for Pa-231 from 1955–1959. However, Pa-231 was utilized in analytical work in R-120 and other production and development areas from the late 1950s through the late 1960s; it was chemically extracted from Cotter Concentrate in SW Building from 1970–1979; and it was separated, characterized, and analyzed in R-Building from 1956–1987. A site expert interview conducted by NIOSH indicates that workers performing Pa-231 separations were concerned about uptakes and desired a bioassay program (NIOSH 2008a). However, no Pa-231 bioassay data have been found for the years between 1959 and 1993.

A bioassay program for Th-232 sludge re-drumming calls for Thorium Refinery personnel to provide 24-hour urine specimens on a monthly basis. According to NIOSH (2007), "several hundred drums were periodically re-drummed in the summer months between 1954 and 1966." Some drums remained in use after construction of Building 21 for bulk storage; at least 117 leaky drums remained outside Building 21 as late as 1973 (Mound, no date). Even acknowledging the seasonal nature of this work, the limited quantity of available bioassay data does not appear adequate to represent monthly bioassay samples from all participating personnel throughout the duration of this mission.

It is interesting to note that bioassay data for "other" radionuclides were far more prevalent during the proposed SEC period than during later years. There were 238 Th-232 bioassay results for the SEC period, based on the MJW other radionuclide files alone. In contrast, a total of 84 sample results were located by SC&A for Th-232 for the period from 1960 to 1967 in the *Data Base of Excretion Data of Other Radionuclides* (ORAU 2003a) and the *Database of Ra-Ac-Th Excretion Data* (ORAU 2003b), as well as logbooks and bioassay reports. No results were located for 1962. Similarly, there were 180 bioassay results for Th-230 during the SEC period; there were no sample results for the 1960s and 1970s, when Th-230 was utilized in analytical programs and was extracted from Cotter Concentrate.

An uptake incident from October 1978 demonstrates that radium, actinium, and thorium continued to pose an exposure potential well after decontamination and entombment of the former processing facility. The incident summary reviewed by SC&A does not clearly indicate the location or work activities of the exposed individual, although it refers to a prior health physics audit of the "Cave Area." This seems to imply that the exposure may have been related to legacy contamination from the entombed Old Cave, rather than current processing or research (Jenkins 1978).

> *A gamma scan of the nosewipe indicated the presence of Ra-226. An analysis of the nosewipe by alpha spectroscopy indicated the presence of Po-2l0, Th-230, Ac-227, Th-227, Ra-223, Ra-226 and U-238.*
>
> *A gamma scan of plaster from the area indicated the presence of Ac-227.*
>
> *An analysis of the air filter after 11/2* [at least 4 days post exposure] *by alpha spectroscopy indicated the presence of Po-210, Th-230, U-233, U-238 and the following decay products of Ac-227: Th-227, Ra-223, Rn-219, Po-215 and Bi-211.*
>
> *In an effort to minimize the chances of missing one or more of the nuclides that may have been present in the urine, the chemical procedures used to prepare the urine samples were not the same as is used routinely; therefore, a comparison of these data with data from the subject's routine urine samples would not be meaningful" (Jenkins 1978).*

The incident summary also indicates a potential for loss of control of internal monitoring samples: "The nosewipe was set aside in the counting room without its identification." The author's confidence in the correct identification of the nosewipe is "quite high," because "it was the only positive nosewipe observed in the last month," and because alpha spectroscopy for the nosewipe and the air filter resulted in a similar mix of radionuclides (Jenkins 1978).

The health physics report for this incident does reflect a diligent effort to evaluate the individual's internal exposure, and the analysis conservatively estimated an uptake of less than 1% of a maximum permissible body burden. However, the incident demonstrates that D&D of the Old Cave in 1959 did not eliminate exposure potential to airborne actinium, radium, thorium,

and their decay products. The event also indicates that standard bioassay procedures in the late 1970s were not capable of detecting all of these radionuclides. Communication among workers, supervisors, field health physics, and the bioassay lab would be essential for ensuring that correct (non-standard) procedures were used to analyze for exotic radionuclides. Since these isotopes were actively handled in research, analytical procedures, separations, and waste management well beyond 1959, SC&A maintains that these isotopes represent an inadequately monitored exposure potential.

The ER and subsequent white papers have not demonstrated that quantities handled were insufficient to affect worker dose. The responses NIOSH has provided to date do not satisfy SC&A's concerns regarding the feasibility of assigning dose in the absence of personnel monitoring data.

5.0 URANIUM AND TRANSURANICS (AM-241, CM-244, NP-237, U-233, U-234, U-235, AND U-238)

Uranium-233 projects occurred in 1958 and 1959 and from 1966 through the late 1970s. Uranium-234 was produced from chemical separation of aged Pu-238 from 1962 to 1979. Other isotopes of uranium, such as U-232, U-235, U-236, and U-238, were also handled at Mound.

Several U-234 separation campaigns occurred at Mound in R-Building and SM-Building. The petition evaluation report identifies U-234 separation from Pu-238 occurring from the mid-1950s to 1972 (NIOSH 2007, pg. 24). A pilot program for separating U-234 from Pu-238 by tri-butyl solvent extraction and ion exchange was conducted in SM-Building from 1965–1970; the ensuing production occurred in R-Building from 1970–1980 (Mound 2001). Uranium-234 was listed as being present during compatibility studies involving assembly and disassembly of heat source units and "hot" metallographic analysis (Mound 2001; Wagner and Stought, no date). A successful U-234 separation process and investigation of a second extraction process were reported in 1963 (Eichelberger 1963). According to *Mound Facility Activities in Chemical and Physical Research: July-December 1979* (Mound 1980), "Mound Facility has been separating and recovering high-isotopic purity uranium-234 from aged plutonium-238 since 1964." Gram quantities of high-isotopic purity U-234 were shipped to Oak Ridge National Laboratory at least through 1979.

Isotopic separation of uranium isotopes, especially U-235 and U-238, using a chloride separation technique occurred in R-Building starting in 1980. This project also involved work with Pm-147 and calcium separation analysis (Mound 2001).

One of Mound's primary missions was to produce tritium for the weapons program. Research and development activities for this program utilized metal tritides, including uranium tritide. Facilities for tritium recovery also involved the use of uranium in the R-Building, SW-Building, and T-Building. King (Mound 2001) lists uranium as a secondary radionuclide of concern during these operations, behind tritium. Uranium beds containing pulverized powder were used as storage beds in the tritium program. Enriched uranium, in the form of metal and scrap, was listed on an inventory in 1963 (Blanke 1963).

The report of a small uranium fire in SW-Building provides some details of interest regarding uranium handling and monitoring in the tritium program. The incident investigation report indicated that the uranium that ignited had been treated no differently than previous uranium samples. The tritium was removed from the uranium tritide complex by heating, and the uranium cooled to room temperature in a reaction vessel. The material in the reaction vessel was approximately 0.3 grams of fine powder. Air was introduced to oxidize the uranium; a red glow was observed. The uranium was then left open to air in a fume hood from Friday until Tuesday morning. An individual shook the reaction vessel and poured the uranium into a plastic waste bottle, which contained oxidized waste from approximately 10–15 previous thermal decompositions. Turning away after pouring the uranium into the bottle, the worker saw a red glow reflected in the glass front of an instrument (Nunn et al. 1980):

He turned around and saw a flame rising about two inches above the mouth of the plastic waste bottle and the entire bottle glowing red. He warned [name redacted] to evacuate the area. As [name redacted] walked past the burning plastic container, he tried to pull it out of the hood with his hand into a fire retarding trash container located in front of the hood. Some of the burning material fell outside the trash container to the floor and on a chair near by. A paper schematic on the chair was ignited, and the material on the floor continued to burn.

The [redacted] individuals extinguished the fire while another worker called for emergency assistance. There were no firm conclusions as to why the uranium had not completely oxidized in the reaction vessel (Nunn et al. 1980).

Urine samples were collected from [redacted] individuals involved in the incident. The samples were analyzed for tritium uptake (increases from background were judged to be insignificant). No uranium bioassay data are available, despite the fact that all material in the waste container had undergone thermal decomposition, which was known to remove "greater than ninety-nine percent of the tritium." It appears that radiological monitoring for this situation, from a beta monitor in the room to the urine bioassay, focused exclusively on tritium, while ignoring the uranium.

There are uranium urinalysis data from 1958–1959, 1965–1966, 1972, 1984, 1989–1991, and 1993–2006. There is limited radionuclide-specific data for U-233, and no specific data for U-234 prior to 1966. Ten workers were monitored for uranium during 1958 and 1959, and five samples labeled as U-233 were processed during 1965 and 1966. After 1966, there is a gap broken by one uranium bioassay in 1972 and samples collected for several individuals in 1984. No U-234-specific bioassay program was associated with the U-234 processing from 1954–1958 (within the proposed SEC period) or from 1970–1980 (well after the proposed SEC period). Beginning in 1984, isotope specific (i.e., U-234, U-235, and U-238) results are seen in the bioassay data.

While processing and handling of uranium compounds continued until the 1990s, uranium bioassay data are scarce from 1966 until the D&D era. The ER does not indicate how process data can be used to assign dose to unmonitored exposed workers for all isotopes of uranium. SC&A questions whether NIOSH can bound the exposures to uranium based on the minimal bioassay data available, particularly given the inherent limitations of fluoroscopic analysis techniques used during the 1950s–1985. The responses provided to date do not satisfy SC&A's concerns regarding the feasibility of assigning dose in the absence of personnel monitoring data.

5.1 AMERICIUM, CURIUM, AND NEPTUNIUM

Transuranics such as americium, curium, and neptunium were handled during R&D activities, including chemical separations. They were also used in analytical procedures involving vaporization. Available documentation indicates that activities involving these isotopes occurred from 1956 to 1987. Bioassay records are limited to 1 cluster of Am-241 results for an individual

in 1987, 38 Cm-244 results for 6 workers in 1983, and 2 Cm-244 results for an individual in 1986. The paucity of bioassay data for Am-241 and Cm-244 could limit validity of dose estimation; it is also unclear in which programs the monitored people were involved. In this context, the extension of the available data to unmonitored individuals would become questionable. No Np-237 bioassay data have been located for the period of concern. The numbers and dates of available bioassay data do not represent the entire exposure period for any of these radionuclides. The ER and subsequent white papers do not demonstrate that quantities handled are insufficient to affect worker dose. The responses provided to date do not satisfy SC&A's concerns regarding the feasibility of assigning dose in the absence of personnel monitoring data.

6.0 FISSION AND ACTIVATION PRODUCTS AND OTHER RADIONUCLIDES

The most predominant source of activation products at Mound, which has not been adequately addressed in NIOSH's evaluation, was the production of Po-210. There are two periods of time for which NIOSH has excluded Mound workers from the SEC class. The first is the period from February 1, 1949–September 30, 1949, and the second is the period after February 28, 1959. SC&A has concerns regarding the feasibility of dose reconstruction for fission and activation products associated with the polonium process at Mound during both of these excluded periods.

The polonium production process, which transferred to the Mound Plant in February 1949, involved the separation of the bismuth slug from the aluminum can through chemical dissolution. Both the aluminum used in fabricating the can and the bismuth contained impurities of iron, silicon, cobalt, lead, tin, zinc, silver, chromium, vanadium, and gallium. Upon being irradiated, these impurities produced gamma- and beta-emitting radionuclides, which at the time of bismuth processing created a radiological hazard (BWXT 2002; Moyer, 1956).

The chemical separation produced a waste stream with a higher concentration of these gamma and beta-emitting isotopes than was present in the original slug. Waste processing and handling would also result in a potential for exposure to these impurity radionuclides. Aqueous waste from Po-210 was originally transferred to HH-Building via waste lines. In 1959, the waste treatment was moved from HH-Building back to T-Building. From 1949 to 1971, WD-Building supported the polonium operations. Aqueous wastes containing polonium from HH-Building, and subsequently from T-Building, were transferred to WD-Building via underground sewer lines for polonium wastewater treatment. WD-Building also received polonium waste or waste byproducts from R, T, and HH buildings, where research and processing took place, as well as wastewater from the H-Building laundry. Potential for exposures existed from contaminated areas, breaches of pipes and sumps, leakage from filter banks through 1969, hot maintenance activities, sludge packaging, and denitrification and scrubbing of gases (Mound 1993, Mound 2001).

This issue is closely linked with previous SEC determinations. The polonium process was transferred to Mound Plant in Miamisburg, Ohio, from the Monsanto Chemical Company (MCC) Dayton Laboratory in Dayton, Ohio, where unmonitored exposures to activation products and neutrons have contributed to the granting of SEC status. NIOSH stated the following in the MCC ER (NIOSH 2006):

> *Initially part of the Manhattan Project, the Dayton facility separated polonium-210 from naturally occurring materials to produce polonium-beryllium source neutron generators for atomic bomb initiators. By 1944, the decision was made to transmute bismuth-209 into polonium-210 via neutron bombardment. That reaction can be stated as: 83 209Bi + η→ 83 210Bi → 84 210Po + β-. The beta radiation from the irradiated slugs was so intense that they could not be handled without lead gloves and tongs (ORAUT-TKBS-0016-6)* [ORAUT 2004]. *Polonium impurities produced a number of activation products that were beta*

emitters. Silver-112 was a particular problem with beta particles of 3.94 MeV and caused the irradiated ingots to generate high dose rates. Other beta-emitting radionuclides of concern were antimony-124 (2.31 MeV), iron-59 (1.57 MeV), cobalt-60 (1.48 MeV), cesium-137 (1.176 MeV), bismuth-210 (1.160 MeV), tin-121 (0.42 MeV), zinc-65 (0.327 MeV), and mercury-203 (0.214 MeV).

NIOSH has determined at this time there is a lack of sufficient monitoring and source term data for nuclides other than polonium between 1943 and 1949 at MCC. Although polonium bioassay data used in conjunction with co-worker data from Mound Laboratory and ambient environmental polonium internal intakes could be used to support internal dose reconstruction, due to lack of information and internal exposure data for the use and production of radionuclides other than polonium, NIOSH has concluded that there are insufficient data available to support internal dose reconstructions with sufficient accuracy at MCC for the time period 1943 through 1949. This inability to complete internal dose reconstruction at MCC for the 1943 through 1949 time period is because of a lack of information and internal exposure data for radioisotopes other than polonium, such as antimony-124; bismuth-210; cesium-137; cobalt-60; iron-55 and -59; lead-210; mercury-203; polonium-208 and -209; selenium-75; silver-112; strontium-90; tellurium-121 and -132; and tin-121; as well as radium and thorium.

The presence of these impurities at Mound is well established. Activation products were readily identified in the waste streams from the polonium process and were considered a radiological hazard. Longer-lived activation products from this process were identified during characterization activities in the 1990s. NIOSH reaffirmed the presence of impurities in the aluminum cladding and their presence at T-Building in the roadmap.

> *Both waste streams had high degrees of beta and gamma radiation from the irradiated impurities in the aluminum can and in the bismuth metal.* [ORAUT 2009, pg. 81 of 110]

NIOSH has not provided an adequate explanation for excluding the period from February 1, 1949, through September 30, 1949. Work on polonium processing began in R-Building and T-Building as early as February 1949. Polonium processing at the Mound Plant, as at the Dayton facility, began with the dissolution of the aluminum can from the bismuth slug; this step and associated waste streams constitute a major source of activation products. The process used to separate Po-210 was equivalent for both facilities during this time period. In addition, neutron monitoring was not implemented at Mound until August 1949. Absence of neutron monitoring was also cited as a reason for granting the Dayton Laboratory SEC class.

During the January 5–6, 2010, Mound Work Group meeting, NIOSH indicated that the radiological controls in the T-Building at the Mound Plant were substantially better than those at Dayton Laboratory. While controls were somewhat improved over those at Dayton, the existing situation did not preclude the potential for exposure to impurity radionuclides. The potential for

exposure existed throughout the polonium processing years, as demonstrated by analysis of radiological controls and incidental releases. Additional documentation of concerns regarding radiological controls is presented in Attachment 1 of this report.

Gloveboxes used for polonium processing at Mound were primitive by modern standards. In the early days of the project, all work was performed on an open bench top or in standard laboratory fume hoods. All rooms where radioactivity was handled additionally had air locks and negative ventilation. "Urine analysis, used as a monitor of activity in the body, indicated that the aforementioned installations and precautions were inadequate" (McEwen 1949). The solution was to use a drybox (glovebox in modern terms), which was originally developed "to maintain an inert atmosphere around the enclosed operation rather than to prevent the spread of activity." As the need for more effective confinement of radioactivity became apparent, work was initiated to design "Special Hoods" for this purpose. Because gloveboxes were not available on the market at the time, Mound's dryboxes were designed and built in-house. The main objective was a design "that would adequately confine activity, but would not materially handicap personnel or require drastic changes in the design and operations of the existing process equipment" (McEwen 1949).

"Plywood of the proper thickness was to be used for the basic hood structure because of its relatively low cost, ease of fabrication, adaptability for alterations, and suitability to the installation of services" (McEwen 1949). The surfaces of the boxes were to be varnished or painted to resist the chemicals used in processing. Attempts were made to standardize hood design. This attempt achieved some success, but true standardization did not occur. "Some further standardization might be accomplished in later designs, but as long as the material of construction is limited to plywood, improvements in design and standardization will probably be minor. The greatest improvement would be the discovery of a better construction material" (McEwen 1949).

The following incidents from the 1960s were associated with failures of the original design gloveboxes that had been used from the start of polonium operations, providing evidence of exposure potential throughout the period of use. "The items below, list the incidents which have occurred in either T-267 or T-270 over the years 1963-64-65, which can be **attributed to faulty glove boxes that have now been in use sixteen years**" (Guillet 1965, emphasis added).

1. *None in 1963.*
2. *01/17/64 - Faulty plastic pass box.*
3. *04/28/64 - Contaminated water leaking thru low risk barrier.*
4. *10/5/64 - Pressurized acid supply line.*
5. *10/30/64- Fire in wooden hood.*
6. *05/20/65 - Repeat of 4/28/64 - dried out caulking*
7. *03/29/65 - Pass box repair needed.*

In addition to the polonium impurities, beta/gamma-emitting radionuclides were handled in other operations at Mound. In some cases, the beta- or gamma-emitting radionuclides were identified as the major radionuclide(s). For example, isotopic separation of Sr-90 and Y-90 occurred in

R-Building from 1960–1962. In the NIOSH response on data adequacy and completeness, Sr-90 is listed as a major radionuclide of concern for this program. NIOSH indicates in their response that this project was limited to two radiochemists; however, they have not considered the potential exposures to support staff or the fact that no bioassay data exist for Sr-90 from this time period for either the radiochemists or support workers. There are no isotope-specific or gross beta/gamma bioassay results for Sr-90 until 1993, and NIOSH has not specified how dose will be reconstructed for prior years.

A campaign to separate Ba-137m and Cs-137 was also conducted onsite during 1968 and 1969 (Essig and Neubert 1969, Mound 2001). No bioassay data for either isotope have been located from this period. Cesium-137 bioassay was not performed until 1993, and no method has been proposed to reconstruct internal doses prior to this time.

Several other operations, including incineration, use of a glass melting-furnace, neutron accelerator source irradiations, and handling of orphan sources, created exposure potential to fission products. SC&A acknowledges beta/gamma emitters were often handled in the presence of alpha emitters. However, NIOSH has not presented a method of reconstructing doses from beta/gamma emitters from alpha bioassay data, nor have they addressed those situations in which beta/gamma emitters were handled without associated alpha emitters.

Workers at Mound experienced exposure potential to beta- and gamma-emitting radionuclides throughout the site's history, extending before and after the NIOSH-proposed SEC period. During the July 2008 work group meeting, NIOSH/ORAUT indicated that no gross beta results had been located to date. Limited bioassay sampling was done for Cs-137 (1993–1995), Co-60 (1993–1995), Mn-54 (1994–1995), and Sr-90 (1993–1997). In the *Health Physics – 5 Year Plan* (Monsanto 1967), there is mention of the development of a promethium urinalysis procedure for Mound. Whether the procedure was implemented and monitoring data collected is unknown. No promethium bioassay data have been located to date. There is an absence of beta/gamma internal monitoring for a majority of the years when beta/gamma emitters were present at Mound, particularly during the production era. With several of these isotopes persisting into the D&D characterization period, it is apparent that they were present in higher activities, and accompanied by additional shorter-lived radionuclides, during the period when polonium was actively processed.

Again, NIOSH has indicated that the predominant internal exposure at Mound was to alpha emitters and tritium. They further state that beta/gamma emitters played a minor role in Mound activities, and for the most part only existed in trace-quantity research and production-scale operations (NIOSH 2009). NIOSH has not produced objective data regarding the quantities of material handled or processed or the concentrations of these radionuclides in various source terms, including waste streams. While they maintain that these radionuclides are not of dosimetric significance, they have not quantitatively defined what constitutes dosimetric significance.

In summary, the responses provided to date do not satisfy SC&A's concerns regarding the feasibility of assigning dose in the absence of personnel monitoring data for beta/gamma-

emitting radionuclides. The ER and subsequent white papers do not demonstrate that quantities of beta/gamma emitting radionuclides are dosimetrically insignificant to all organs, nor have they provided objective evidence that radionuclide concentration data are available such that the relative dose of beta/gamma emitters can be determined from available personnel monitoring data for the predominant radionuclides in the process and waste streams.

NIOSH has not provided an adequate justification for excluding Mound workers from February 1949–September 1949 or for periods after February 1959 when no bioassay data are available. They have granted an SEC at MCC Dayton Laboratory for an equivalent polonium process in the period immediately prior to this, and for Mound starting in October 1949. NIOSH has not provided an adequate explanation for why it is not feasible to reconstruct dose for Sb-124, Bi-210, Cs-137, Co-60, Fe-55, Fe-59, Pb-210, Hg-203, Po-208, Po-209, Se-75, Ag-112, Sr-90, Te-121 through Te-132, and Sn-121 at the Dayton Laboratory, yet dose reconstruction is feasible for the same radionuclides at the Mound Plant. The current position that fission and activation products at Mound are not of importance due to their trace concentration is in conflict with the position taken in the MCC petition evaluation. The lack of information and internal exposure data for impurity radionuclides has not been resolved for the period of active polonium production at the Miamisburg location.

7.0 USE OF GROSS ALPHA AS A SURROGATE FOR RADIONUCLIDE-SPECIFIC BIOASSAY

NIOSH has indicated that gross alpha results can be used to bound the dose for all alpha-emitting radionuclides at Mound where isotope-specific data are not available. The gross alpha results would be credited to the radionuclide resulting in the highest dose to the organ of concern, with case-specific information being considered when available (e.g., where a worker worked). This methodology encompasses all alpha-emitting radionuclides (i.e., Am-241, Cm-244, Pu-238, Pu-239, Ac-227 via daughters Th-227 or Ra-223, Ra-224, Ra-226, Th-228, Th-229, Th-230, Th-232, Pa-231, Np-237, U-233, U-234, U-235, and U-238) at Mound for which there are no isotope-specific bioassay data.

To appreciate the intricacies of the bioassay data collected for alpha emitters, the following items must be understood.

(1) The different radiochemical and subsequent counting techniques utilized
(2) The method for designating the radionuclide to which an individual was exposed

SC&A has attempted to obtain very specific information from radiochemists and chemists throughout the evaluation process; however, those consulted were not familiar with the particular radiochemical techniques. SC&A has conducted interviews with personnel responsible for radiochemical analysis of bioassay samples in an effort to clearly understand the radiochemical analysis, the method for identifying radionuclides present in the urine, and the protocol for documenting bioassay results.

7.1 GROSS ALPHA DETERMINATION (AKA PLUTONIUM BIOASSAY)

The history of plutonium bioassay is relevant to the detection of other alpha-emitting radionuclides, since this technique was not specific to plutonium, but also carried other alpha-emitting radionuclides through the radiochemical process. The initial plutonium technique (gross alpha determination) developed in 1956 was developed to monitor for plutonium; however, the technique was actually more of a gross alpha procedure. It was later referred to as the "gross alpha determination." The first step of the procedure was to cause an alkaline-phosphate precipitate to form by adding ammonium hydroxide. Most of the solids and the polonium remained in supernate (i.e., urine). Purified cerium carrier in the +3 and +4 valence state was added to the sample. The cerium phosphate carrier brought down plutonium, thorium, uranium, protactinium, and americium (possibly curium). The cerium precipitate was initially mounted on a disk and counted for gross alpha activity. Although the radiochemical process actually brought down multiple actinides, the sample results were considered as plutonium, unless it was known that an individual was working with another radionuclide, such as uranium or thorium (Sheehan 2009a).

Whether all alpha emitters were captured along with the plutonium or were captured at the same efficiency as plutonium is not clear. A similar procedure was implemented for analyzing actinides in urine at Argonne National Laboratory (ANL) using alkaline phosphate precipitation

followed by co-precipitation with bismuth phosphate and then cerium fluoride, isolated heavy metals, including thorium, neptunium, actinium, plutonium, and trans-plutonium elements (ANL 1989). Although uranium is present after the alkaline phosphate precipitation step, ANL indicates that uranium does not come through in the co-precipitation process with bismuth phosphate and then cerium fluoride (ANL 1984).

The gross alpha procedure did not isolate radium isotopes. A radium bioassay procedure, utilized during the Ac-Ra-Th production campaign, used barium sulfate to isolate the radium isotopes from the other actinides (e.g., Ac, Th, U, Pu, and Pa). The barium sulfate step was eliminated once the Ac-Ra-Th program ended; it required much more labor and was no longer necessary, because Mound was no longer doing radium bioassay (Sheehan 2009a). The gross alpha technique, at a minimum, could not be used as a surrogate for radium after the barium sulfate step in the gross alpha radiochemical procedure was discontinued.

Actinium-227 is a beta emitter. There was no attempt after the Ra-Ac-Th separation project to characterize Ac-227 from its alpha-emitting daughter by the Mound bioassay group.

While the gross alpha radiochemical procedure may have precipitated Ac-227, the sample counting procedure with a low background proportional counter yielded gross alpha results. The gross alpha count would not be inclusive of Ac-227, a beta emitter. Because Ac-227 was not directly analyzed, radionuclide progeny (i.e., daughters) must be used as a surrogate for the parent. Where daughters are used to derive parent radionuclide activities, the knowledge of the ratios of the daughters to parents in the respective source terms must be known, and the differential effects of biokinetics between parent and daughter radionuclides must be considered. Data can be obtained from process information; however, the age of the material at the time of uptake is generally unknown, and equilibrium between parent and daughter depends on the age of the material being processed. Equilibrium may or may not exist. For dose reconstruction, NIOSH might assume that Ra-223 is in equilibrium with Ac-227 in urine. However, this assumption would not support estimates of Ac-227 from gross alpha results, because the gross alpha procedure is selective against radium. Thorium-227 is detectable by the gross alpha procedure according to Mound radiochemists; however, the state of equilibrium must be known. Given the Ac-227 separation activities conducted at Mound, it is unclear how the alpha-emitting daughters can be used to extrapolate Ac-227 uptakes. The claimant-favorable assumption would be to assume 100% pure Ac-227, which cannot be detected by an alpha count.

7.2 ANION EXCHANGE

From mid-1966 to mid-1967, Mound implemented an anion exchange technique for most urine samples, which was selective for plutonium or uranium. In 1967, laboratory technicians returned to using the gross alpha procedure, but they began to do some specific analyses for “special samples” (e.g., Th-228, Th-230) by running samples through a column to select the nuclide(s) of interest. In 1967, Mound also implemented pulse-height analysis for “special samples.” Interviews with a former worker indicate that pulse-height analysis was conducted when they wanted more specific information on the radionuclide in the urine (SC&A 2008b). Documented

details on when Mound used pulse-height analysis versus gross alpha measurement with a proportional counter are not available.

Plutonium-specific data using an anion exchange column, starting in mid-1966, allowed sequential elution off the column to obtain isotope-specific samples. The adjustment of the pH allowed for isolation of plutonium, americium, neptunium, and uranium in sequential steps. In general, an 8 M HNO_3 solution was used to strip off the plutonium, and this material was electroplated along with an internal tracer (Sheehan 2009a). "Mound Laboratory 24-Hour Urinalysis Procedures for Actinides," *History of the Mound Bioassay Programs* (Meyer 1992) indicates that thorium, curium, trans-plutonium, strontium, and polonium are not absorbed onto the column and are carried in the rinsate. With the implementation of anion exchange, the focus of routine bioassay was on plutonium; analyses of other radionuclides depended on notification from the field or technical staff regarding which radionuclides were handled by the personnel submitting specimens. The default routine analysis did not include all alpha emitters. Unless the field indicated potential exposure to an isotope other than plutonium, the bioassay lab only pulled off the plutonium fraction from the column. Once electroplating and use of an internal tracer began, pulse-height analysis was used (Sheehan 2009a). The routine use of anion exchange, rather than the rapid gross alpha technique, would preclude the analysis of all alpha emitters, once again putting a reliance on isotope-specific bioassay. Appropriate analysis for radionuclides other than plutonium during this period required effective communication between the field and the bioassay laboratory.

It is important to note that the Field Health Physics group was responsible for communicating the need to analyze a bioassay sample for radionuclides that were not routinely isolated and counted. The information was provided by several Health Physics Supervisors who had responsibility over specific areas of the plant. For example, there was a supervisor for G-Area, T-Area, SM/PP-Area, and SW Area (Sheehan 2009a). For the technical staff who worked directly with special radionuclides, the field health physics staff was generally aware of the isotopes being utilized and the need for special analysis. In the case of the maintenance staff, Maintenance Work Permits (MWPs) generated for each maintenance job provided some information on monitoring needs. These MWPs have not been located to date. For maintenance staff, it was more difficult to identify those working with radionuclides other than polonium or plutonium. There was a period of time when maintenance workers were restricted to either the polonium areas or the plutonium areas (SC&A 2008b). After the bioassay laboratory added copper powder to their radiochemical procedure, they were able to remove the polonium from plutonium samples, and workers were allowed to work in both areas (SC&A 2008b). The communication between the field and the bioassay staff was relatively successful in the case of an incident, but it was not always effective for routine operations.

As a point of clarification, SC&A has not implied that the Mound bioassay program lacked the capability to detect alpha emitters other than plutonium and uranium during the 1966–1967 time period, as argued by NIOSH. The rapid gross alpha technique was still available; however, a memo from Sheehan (1966) referenced above actually states:

> *Starting with this report all 24-hour urinalysis results are being reported as plutonium and uranium as we are now using anion exchange separations* ***which are selective****.* [Emphasis added.]

The procedure using the anion exchange column, while capable of isolating multiple actinides in sequential steps, only pulled off the plutonium fraction from the column, unless the field indicated there was a potential exposure of an isotope other than plutonium.

Procedures are available for both the gross alpha determination and the anion exchange procedure at least as early as 1968, and results from this era generally do not indicate which procedure was used.

Based on a review of Meyer (1992) and the PURECON database, it appears that, from 1956 to mid-1966, results from analysis of 24-hour urine samples were reported as gross alpha and total alpha, without specification of an isotope. Results continued to be reported as gross alpha or total alpha until 1970, with exceptions in some records from mid-1966 to mid-1967. The bioassay results themselves provide little assistance in determination of whether gross alpha or anion exchange were used after 1970. There is a notable change in the recording practices in 1971. In general, the bioassay result was assigned to the radionuclide that the individual was working with at the time. As such, during the 1970s, results identified in PURECON as Pu-238, Pu-239, ***or*** other specific radionuclide may be misleading, because the gross alpha determination was not selective for particular alpha emitters. In the case of anion exchange, the radiochemical method allowed for the discrimination between plutonium, americium, neptunium, and uranium. Pulse-height analysis, starting in 1967, was conducted on special samples when more specific information on the radionuclide in the urine was desired; however, the use of this technique is not clearly stipulated in the bioassay record. A preponderance of the bioassay results in PURECON are for Pu-238, Pu-239, and/or total or gross alpha. The plutonium data in PURECON indicate that a majority of the results were assigned to Pu-238 from about 1971 through 1980. In 1981, isotope-specific data started to appear in the record (e.g., Pu-238, Pu-239, etc.)

7.3 ALPHA SPECTROSCOPY AND ANION EXCHANGE CHEMISTRY

In 1980, Mound implemented alpha spectroscopy and anion exchange chemistry, which allowed for radioisotopic analysis (Meyer 1992).

> *During 1980,* [name redacted] *had the technicians use specific anion exchange chemistry, plutonium-242 as a tracer with electrodeposition of sample mount, alpha spectroscopy counting and the processing of sample blanks. Running blanks allowed the calculation of the detection limit.*

In 1980 and thereafter, radionuclide-specific analysis replaced the gross alpha technique. At this point, NIOSH's proposal to bound dose with gross alpha results is no longer feasible. Personnel monitoring is now specific to the radionuclide and results listed in the dosimetry file. For example, if an individual worked with Pa-231, Ac-227, Pu-238, and Pu-239, and bioassay results

were available for Pu-238 and Pu-239, the individual was effectively unmonitored for Pa-231 and Ac-227. SC&A has provided several examples of situations post-1980 where there was a potential for exposure to alpha and beta emitters for which no bioassay data are available. No technique has been proposed by NIOSH to assign dose from unmonitored alpha emitters during this time period.

NIOSH has not provided relative ratio data for unmonitored alpha emitters to monitored alpha emitters for all processes.

In summary, there are significant gaps in bioassay collection for alpha-emitting radionuclides other than plutonium and polonium (through 1973), compared to the years when the radionuclides were present at Mound. As a result, NIOSH is relying on gross alpha results to represent "other alpha emitting radionuclides." The gross alpha determination procedure is not a catch-all procedure for all radionuclides of concern; the procedure is particularly inadequate for radium and actinium. With the use of the anion exchange procedure, there are questions regarding the ability to capture curium, thorium, trans-plutonium, strontium, and polonium (post-1973) in the analyzed sample. Anion exchange was performed for plutonium only unless otherwise specified by the field radiological control staff. This communication was not always effective, particularly in the case of support workers.

7.4 AIR SAMPLING

A brief evaluation was done of the availability of air sampling data available on the SRDB, versus the buildings and time periods when thorium was handled. The available air sampling data are limited to specific locations and time periods; air monitoring information exists for PP-Building (1981, 1982, and 1984), R-Building (1980–1990), SM-Building (1981, 1982, and 1984), SW-Building (1972 and 1995), WD-Building (1978 and 1994), and Building 21 (1987). It is apparent that the available air sampling data do not represent all areas and time periods at Mound. Furthermore, NIOSH has stated in the ER (NIOSH 2007):

> *Mound health physics program records include a number of air samples for alpha- and beta-emitters. Typically, such data are of limited use in dose reconstruction due to the great uncertainties associated with the doses estimated based on measured air concentrations. When possible, dose reconstructions are usually performed based on data related to a specific claim (including bioassay data), which provides a much more direct assessment of the uptake of a given radionuclide and the resulting organ dose.*

Although NIOSH indicates in the ER that measured air concentrations can be useful in estimating maximum doses when bioassay data are unavailable, this cannot be done where air concentration data are not available.

7.5 ITEM OF CLARIFICATION FROM THE NIOSH EVALUATION OF DATA ADEQUACY AND COMPLETENESS ISSUES AT THE MOUND LABORATORY

7.5.1 Item #1

NIOSH has indicated the following (NIOSH 2009, pg. 7):

> *As has been explained repeatedly [(1) "NIOSH Responses to Mound Matrix Items," July 5, 2008, page 19; (2) Mound Working Group meeting on May 27, 2009 transcript page 163; (3) Mound Working Group meeting on July 14, 2008 transcript pages 213–215] NIOSH does not propose to use gross beta results as this bioassay was not used at Mound. The mention of it in the ER was an error.*

Furthermore,

> *NIOSH does not propose to use gross beta results (see previous response).*

NIOSH/Oak Ridge Associated Universities Team (ORAUT) indicated that in lieu of isotope-specific bioassay data, they could rely on gross alpha and gross beta results (NIOSH 2008b):

> *Gross alpha and gross beta results are usually credited to the radionuclides resulting in the highest dose among those to which exposure is possible and plausible when circumstances of the potential exposure are not known.*

The SC&A review must be conducted on the current version of the ER which does state that gross beta results will be used. If the ER is incorrect in stating the above, a revision to the ER is warranted.

7.5.2 Item #3

NIOSH correctly identified on page 26 of their response to data completeness that Po-210 has only a stable daughter (Pb-206); therefore, it is inappropriate to refer to Po-210 daughters as radioactive. SC&A will eliminate Po-210 progeny from the list of radionuclides at Mound; however, NIOSH should do the same, as they have identified Po-210 daughters in the roadmap on pages 65, 68, 70, and 96 (ORAUT 2009). NIOSH also referred to Po-210 progeny as a major radionuclide of concern in Attachment A of their response. These corrections should clear up any further confusion.

7.6 SUMMARY

NIOSH maintains that the exotic radionuclides were handled episodically. Furthermore, they infer that when this episodic handling occurred, bioassay was conducted. NIOSH has presented *Major Mound Source Terms and Radionuclides of Concern* in Attachment A of *NIOSH Evaluation of Data Adequacy and Completeness Issues at the Mound Laboratory* (NIOSH 2009). Because the table does not indicate relevant periods for these major Mound source terms, reviewers are unable to link the information provided by NIOSH with available bioassay methods and data in order to critically assess the data adequacy. NIOSH also did not provide a definition of "major radionuclide of concern," nor have they substantiated their position that other radionuclides produce insignificant doses. As a result, SC&A was forced to rely on its own research to identify exposure scenarios with no supporting bioassay.

SC&A has provided several examples representing potential exposure to radionuclides other than plutonium, polonium, and tritiated water or tritiated gas. In these situations, radionuclide-specific bioassay data are absent. In many cases, gross alpha bioassay is of questionable value in isolating these radionuclides, due to the radiochemical methods used. In the case of beta/gamma emitters, there is a complete absence of bioassay until the mid-1990s when the D&D effort began.

NIOSH has indicated that exotic radionuclides were handled in trace quantities, and that their handling was episodic in nature. NIOSH has minimized the relevance of the King document (King 1995; Mound 2001) by characterizing it as listing "any radionuclide that could have possibly been in a particular room" and restricting its intent to determination of bioassay requirements during the D&D era. It is clear from the document itself that the dates provided for radionuclides of concern represent the "usage of radionuclides" and not necessarily "residual radioactivity" or radionuclides with a "possible presence." While the King document (Mound 2001) was utilized for identifying radionuclides that might potentially be encountered during D&D, the determination of bioassay requirements during that period was based on the potential for a worker to receive 100 mrem CEDE. While NIOSH has denied its usefulness in dose reconstruction, one of the stated purposes of the King document (Mound 2001) was to "facilitate internal dose assessment." Episodic presence of exotic radionuclides is not indicated from the King document (Mound 2001), and potential fluctuations in operations cannot account for years and decades in which internal monitoring data are minimal or non-existent. The bottom line is that the King document (Mound 2001) and corroborating evidence indicate active handling of unencapsulated exotic radionuclides for extended periods of time beyond the NIOSH-proposed SEC period.

Where information concerning quantities of materials handled has been readily available, SC&A has included this information; however, months of further research would be required to compile this information for the range of projects described herein. NIOSH has provided no objective evidence supporting their statement that exotic radionuclides, some of which are impurities from major production processes, were present in trace or research quantities. Essential information for these exposure scenarios would include the relative activity of radioactive material handled, the ratios of "secondary radionuclides" to "primary radionuclides" in process and waste streams,

and the engineering and administrative controls utilized to prevent exposure to "secondary radionuclides." Given that the impurity radionuclides were of no interest to the operations, it is unlikely that analytical information is available in many cases.

Dosimetric significance in terms of the compensation program is not defined by the Energy Employee Occupational Illness Compensation Program Act (EEOICPA) or the associated rules. There is no de minimus dose specified. The dosimetric significance was therefore determined based on the requirement for bioassay sampling at 100 mrem CEDE, the radionuclides defined as nuclides of dosimetric significance during the Pre-1989 Dose Assessment Project at Mound, and the sensitivity of Probability of Causation (POC) codes. Clearly, alpha-emitting radionuclides such as Ra-226, Ac-227, Th-228, Th-230, Pa-231, Th-232, U-233, U-234, U-235, U-238, Am-241, and Cm-244 are of dosimetric significance in compensation, as exposures can potentially lead to doses on the order of rems. The requirement for submitting bioassay in the mid-1990s indicates a potential for exposure to result in a CEDE of 100 mrem. If such doses could be encountered during the D&D era, this implies that workers should have been monitored for these radionuclides during the production era, when concentrations were much higher. During the production era, workers were potentially exposed to beta/gamma emitters that would not persist to the D&D era, due to their relatively short half-lives (i.e., <1 year). Furthermore, lack of monitoring or limited monitoring for the same exotic radionuclides have resulted in designation of SEC classes at other EEOICPA sites, indicating their dosimetric significance in the compensation program. Most notably, an SEC class was defined at Dayton Laboratory based on the lack of monitoring for "Sb-124, Bi-210, Cs-137, Co-60, Fe-55, Fe-59, Pb-210, Hg-203, Po-208, Po-209, Se-75, Ag-112, Sr-90, Te-121 through Te-132, and Sn-121," which also existed at Mound Laboratory. Although the engineering controls were better established at Mound Laboratory, these controls did not preclude potential exposures to these radionuclides for the periods of February 1949–September 1949 and after February 28, 1959.

NIOSH has indicated that in order for a potential to exist, material must be handled in such a way as to make it accessible to the breathable atmosphere. The examples provided include handling of unencapsulated sources in various physical forms. Operations included handling of contaminated soil, repackaging of leaking or grossly contaminated drums, chemical separations including vaporization processes, emanation of gaseous radionuclides, handling and processing of radioactive waste, and D&D of radiologically contaminated facilities. From a routine processing standpoint, there was no difference between these exotic radionuclides and the primary radionuclides, yet NIOSH acknowledges potential exposure for plutonium, polonium, and tritium, and denies the same potential for exotic radionuclides handled in the same manner. Exotic radionuclides were clearly present in physical and chemical forms, which presented a potential for exposure.

NIOSH indicates that radiological safety at Mound presented a controlled atmosphere in which radioactive materials were handled. The radiological controls and practices were proceduralized; however, the numerous incidents and failures in radiological and administrative controls paint a different picture. The "pervasive, controlled atmosphere in which radioactive material was handled" appears to be contradicted by:

(1) Incidents numbering in the hundreds for the production areas

(2) Multiple intakes identified for many Mound workers, some of which are not associated with incidental exposures

(3) Administration of chelation therapy to over 60 Mound workers

(4) Recurring failures of engineering controls

(5) Inadequacies in facility procedures

(6) Inadequacies in implementation of corrective actions

(7) Incomplete radiological characterization prior to D&D efforts

(8) Price Anderson Amendment Act (PAAA) fines received for ineffective implementation of radiological controls and personnel monitoring

(9) Radiological control audit reports

After being sued by a union in 1995, DOE agreed to enhance the radiological control program at Mound by installing personal contamination monitors, installing an automated record-keeping system, performing radiological characterization, assessing the need for personal air sampling and enhanced continuous air monitoring, accrediting the bioassay laboratory, and establishing a validation program for internal dosimetry methodologies. This raises questions regarding the appropriateness of the personnel and field monitoring in prior years, given the lack of characterization and the questionable effectiveness of egress monitoring. These factors clearly demonstrate the potential for exposure, not only to primary production radionuclides, but to exotic radionuclides as well.

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Ag-110, Co-60, Cs-137, Fe-59, Po-208, Po-209, Se-75, Sn-121, Sr-90, Te-125, Te-127, Te-129, Sb-124, Zn-65	In the polonium production process, the bismuth slug was separated from the aluminum can through chemical dissolution. The aluminum used in fabricating the can and the bismuth contained impurities of iron, silicon, cobalt, lead, tin, zinc silver, chromium, vanadium, and gallium. Upon being irradiated, these impurities produced gamma-emitting isotopes that, at the time of bismuth processing, created a radiological hazard. The radionuclides went into the waste stream.	1949–1971	T-Building R-Building	Unencapsulated material	1949–1971	Radionuclide impurities from the dissolving operations (air, contamination, waste stream); process, leakage from filter banks through 1969, hot maintenance in T-53, waste treatment, sludge packaging, denitration & scrubbing of gases. There were six process line incidents in 1964–1965 in Rooms T-267 or T-270. Gross contamination spread through all the corridors and many of the working areas on the low-risk side of the T-Building.	Mound (2001); Guillet (1965); BWXT (2002); Moyer (1956); Mound (no date)
Ag-110, Co-60, Cs-137, Fe-59, Po-208, Po-209, Se-75, Sn-121, Sr-90, Te-125, Te-127, Te-129, Sb-124, Zn-65	Processing of aqueous wastes from the Po-210 separations process (including radionuclide impurities)	1949–1971	WD/HH/T	Unencapsulated material	1949–1971	Aqueous waste from Po-210 was originally transferred to HH-building via waste lines. In 1959, the waste treatment was moved from HH-building back to T-building. From 1949 to 1971, WD-Building supported the polonium operations. Aqueous wastes containing polonium from HH-Building and subsequently, T-Building were transferred to WD-Building via underground sewer lines for polonium wastewater treatment. In 1971, it was renovated to process Pu-238 waste water. WD-Building also received polonium waste or waste byproducts from R, T and HH Buildings, where research and processing took place, and wastewater from the H-Building laundry.	Mound (1993); BWXT (2002); Mound (2001); Mound (no date)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Co-60, Sr-90, Cs-137	Room-floors have a high "fixed" radiation potential because of pipes and sumps from the old polonium program.	No Dates	T-Building (T-43, T-44, T-48, T-50, T-57, T-58, T-59, and corridor-51)	Fixed contamination, internally contaminated pipes	Co-60: <1993 and >1995; Cs-137: <1993 and >1995; Sr-90: <1993 and >1997	Potential of exposure with disturbance of floors or breach of pipe and sump systems. Potential exposures exist for processing years, as well as post-processing years.	Mound (2001)
U-234	A pilot program was developed to separate U-234 from Pu-238 by tri-butyl solvent extraction and ion exchange.	1965–1970	SM-Building (SM-38)	Unencapsulated material	1965–1970	Process moved to R-149 for production.	Mound (2001)
Th-232	Th-232 was substituted for Pu-238 compounds for modeling purposes in R&D and for analytical studies. The compounds of thorium are identical to those of Pu-238 used in these areas.	1961–1967 1965–1978 1967–1980	SM-Building (Rooms 61, 62), R-Building, PP-Building	Unencapsulated material	1962, 1968–1971, 1973–1978, 1980		Mound (2001)
Th-228, 232 Ra-226 Radon, Thoron	Thorium sludge operations: re-drumming, transfer to bulk storage, removal / repackaging, decontamination, and entries into contaminated facility.	1955–1976 (Active Storage); 1976-demolition and remediation	Warehouse 15. In and around Building 21. Outside Areas 1, 3, 9, and 12.	Unencapsulated material		Drums of corrosive sludge were stored at Mound from the cancelled thorium refinery project. Drums leaked, and frequent repacking was necessary. 20%–40% of the drums were repackaged annually. This was initially done in Warehouse 15, but elevated radiation levels forced the work outside. Building 21 was built in 1966 for bulk storage of thorium materials. Material was loaded from above through a removable-panel roof. The sludge was repackaged and shipped off site in 1975. Initial cleanup in 1976 - interior wash and paint. Later entries: Drums of Cotter Concentrate were stored in Bldg 21 from 1976–1987. Regular entries were made by health	Mound (2001); Mound (no date); Abbott (1990); Draper (1987); Draper (1994); EG&G Mound (1996)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
						physics personnel for radiological surveys and air monitoring.	
Th-228, Th-230, Th-232, Ac-227, Cs-137, Ra-224, Ra-226, Radon, Thoron	Soil contamination: Area 2 was a disposal trench for empty thorium drums and for Po-210 contaminated sand. Area 7 was a historic disposal area used for disposal of empty thorium drums and radiologically contaminated equipment. It also contains a historic septic tank contaminated with Ac-227 from SW-Building. Thorium contaminated soil was moved from Area 1 to Areas 8 and 12. Area 21 was used for historic storage area formerly used for storage of high-risk wastes from the SW Building (Cs-137 and Ra-226).	Sources of contamination date back at least to 1955. Contamination reported as late as 2000.	Outside Areas: Area 1, Area 2, Area 3, Area 7, Area 8, Area 9, and Area 12; Warehouses 9 and 15; Building 21.	Unencapsulated material; potential exposure from resuspension of soil	Ac-227: 1960–1963, 1965–1988, 1990–1993; Cs-137: 1955–1992, 1996-2000, Th-228, 230: 1959–1987, 1990–1992; Th-232: 1962, 1968–1971, 1973–1978, 1980–1987, 1990–1992; Ra-224: 1959–2000; Ra-226: 1960–1963, 1968–1993; Sensitivity of urine bioassay for Ac-227 ~100X lower than for Pu. (Cheng and Halcomb, no date)	During characterization efforts in the late 1980s, 1990s, and early 2000, several outdoor contamination areas were identified. Redrumming of thorium sludge resulted in apparent widespread contamination due to fugitive dust emissions. Th and Ra were reported for surface soil readings. Bldg 21 - Contamination of surrounding grounds resulted from loading and/or removal of sludge. At least 117 drums were still stored outside Bldg 21 and deteriorating as late as 1973. Surface soil samples - 34,000 pCi/g (80 ft from bldg); 54.3 pCi/g thorium (150 ft from bldg). In general, all surface samples were much higher around Bldg 21 than farther down the hill. Exposure Potential examples: After sludge was removed from Bldg 12, contaminated soil was scraped, moved to Area 12 behind PP, & covered. A corner of the trench was disturbed during removal of WTS overburden, causing Th contamination at "Rader's Hill" across from Bldg 100. HP used the area around Bldg 21 for emergency spill response training 1970–1977.	Mound (2001); Stought et al. 1988; Cheng and Halcomb (no date); Draper (1994); Davis (1994); Draper (1987); Mound (no date); Mound (1993); EG&G Mound (1996).

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Co-60	Outside soil contamination areas from polonium processing and spills to the environment.	1949-remediation	Outside areas (Area 5, Area 20, Area 22)	Unencapsulated material; potential exposure from resuspension of soil	Co-60: 1949–1993, 1996–closure	December 1970 - Waste line break near Building 48, located north of the WD Building.	Mound (1993)
Sr-90 / Y-90	Isotopic separation of Sr-90 and Y-90 through various techniques.	1960–1962	R-Building (R-167)	Unencapsulated material	1960–1962	Sr-90 was the primary radionuclide.	Mound (2001)
Pa-231, Pa-233 Am-241 Cm-244 U-233 Ra-226 Ce-141, 144 Th-230, 232	Work included isotopic separation of Pa-231, Pa-233. Characterization and analysis of cerium-141, cerium-144, americium-241, and Pa-231.	1956–1987	R-Building	Unencapsulated material	Pa-231, 233: 1960–1987; Th-230: 1959–1987; Th-232: 1962, 1968–1971, 1973–1978; 1980–1987; Am-241: 1956–1982, 1984–1985; Cm-244: 1956–1982, 1984–1985; Ra-226: 1960–1963, 1968–1987; U-233: 1960–1964, 1967–1971, 1973–1983, 1985–1988; Ce-141, 144: 1956–1987	Incident: Cm-244 spill, 7/8/83.	Mound (2001)
U-235, U-238, Ca-45, Pm-147	Isotopic separation of uranium radioisotopes, especially U-235 and U-238, using a chloride complex separation technique. The work was done on Pm-147. Ion exchange resins were used with Ca-45 for calcium	1980–Present	R-Building (R-169)	Unencapsulated material	Pm-147: 1980-closure; Ca-45: 1980-Closure; U-235: 1980–1988,	Dates defined per Mound (2001).	Mound (2001)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
	separation analysis.				1992; U-238: 1980–1983, 1985–1988, 1992		
U-234	U-234 separation from aged Pu-238 was done. A homogeneous oxalate precipation technique was used along with two cycles of ion exchange on resins.	1962–1972 (ER) (1970–1980 WK)	R Building SM Room 1	Unencapsulated material	1970–1971, 1973–1980	U-234 was separated from Pu-238from the mid-1950s–1972. U-234 separation activities occurred from 1964 at least through 1979, with gram quantities shipped to ORNL. An incident occurred in R-149 in 1970; no uranium bioassays were collected from those involved.	Mound (2001); Mound (1980); NIOSH (2007)
U-234	Heat Source Program where plutonium molybdenum shards (PMC) were hot pressed into disks and machined to required dimension.	1968–1975	R-Building (R-145, 147)	Unencapsulated material	1968–1971; 1973–1975	Heat Source program involved machining and pressing. Compatibility studies included assembly and disassembly of heat source units and "hot" metallographic analysis. U-234 is listed as a third radionuclide of concern. An incident occurred 8/26/65 involving Pu-238, Pu-239, U-234.	Mound (2001); Wagner and Stought (no date)
Ac-227	Decontamination of contaminated equipment from R-building	1956–1982	R-Building (R-198)	Unencapsulated material	1960–1982	Ac-227 is listed as a secondary radionuclide.	Mound (2001)
Cm-244	Analytical laboratory supporting the heat source program.	1965–1978	R-Building (R-133)	Unencapsulated material	1965–1978	Analytical laboratory supporting the heat source program.	Mound (2001)
Th-228,230,232 Cs-137, Sr-90 U-234, 235, 238 Po-208, 209 Pa-231	Analytical support for work in R-120 and other production and development areas in R-Building.	1950s–late 1960s	R-Building (R-140)	Unencapsulated material, removable contamination	Sr-90: 1950s–late 1960s; Cs-137: 1950s–late-1960s	Early programs consisted of analytical support for work in R-120 and other production and development areas in R Building.	Mound (2001)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Am-241 Ac-227	Analysis of plutonium compounds, alloys, and mixtures along with related radionuclides were studied by chemical analysis and emission spectroscopy. A variety of solutions of oxide samples were commonly prepared for these analyses. This involved vaporization of compounds from arcing.	late–1960s-closure	R-Building (R-140)	Unencapsulated material, removable contamination in glove boxes and general area identified in post-operation era	Ac-227: late-1960s–1988, 1990–1993; Am-241: late-1960s–1982, 1984–1985, 1988, 1990–1993; Sensitivity of urine bioassay for Ac-227 ~100X lower than for Pu.	Analytical program for plutonium compounds, along with related radionuclide compounds, included chemical analysis and emission spectroscopy. Vaporization of compounds from arcing can result in small diameter particles. This room contained removable contamination in excess of 1,000,000 dpm in some areas post-operations. Isotope identification confirmed the presence of Am-241. Am-241 and Ac-227 are listed as secondary radionuclides.	Mound (2001) Cheng and Halcomb (no date)
U-238, Np-137, Am-241, Cm-244	Plutonium research reactor fuels program explored a variety of uranium and later Pu-239 metal and Pu-239 dioxide compounds for possible application into the fuel program. Also used were Np-237, Am-241, Cm-244 in the metal and oxide form. The supporting resarch laboratory used an electron microprobe to characterize materials through a vaporization technique.	1956–1969	R-Building (R-155, 159)	Unencapsulated material	Np-237: 1956–1969; Am-241: 1956–1969;Cm-244: 1956–1969; U-238: 1960–1964, 1967–1969	The plutonium reactor fuels group explored a variety of uranium and later plutonium compounds for possible application in the fuel program. U-238 was the primary radionuclide of concern. Also used were Np-237, Am-241, and Cm-244.	Mound (2001)
U-235, U-238	Tritium processing and research and development activities	1965–Closure	R-Building, SW-Building	Unencapsulated material	U-238:1967–1971, 1973–1983, 1985–1988, 1992	Facilities for tritium recovery, including solid tritiated metal compounds, gas recovery, purification, analytical analyses, and other tritium handling processes involved the use of U-235 and U-238. Uranium was used in storage beds. U-238 is a secondary radio-nuclide	Mound (2001)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Ac-227	Ac-227 (oxide and nitrate) was processed in a separation procedure to acquire a pure radionuclide.	1964–1968	SW-Building (SW-140), R-Building	Unencapsulated material	1965–1968	Ac-227 oxide & nitrate were processed to acquire pure radionuclide. No radium was involved in this process. Separations & shipments were conducted from 1964 through 1967 using material irradiated in 1952–1953. Ac-227 separation from Th-227: shipments were made 1968 - 1970.	Mound (2001); Mound (2003); Monsanto (1968), Essig and Neubert (1969), Monsanto (1970) Nason, et al (1970) Neuberg (1971)
Ba-137m, Cs-137	Ba-137m was separated from its parent Cs-137 by residue adsorption in a phosphate medium as a part of the Radioactive Isotopes Separation program.	1968–1969	Unknown	Unencapsulated material	Cs-137/Ba-137m: 1968–1969		Essig and Neubert 1968
Am-241	Production of alpha sources by plating various metal substrates.	1956–1962 1962–1965	R-Building (R-116, R-120), SW-Building (SW-219)	Unencapsulated material	Am-241: 1956–1965	Major radionuclide of concern in R-116.	Mound (2001)
Th-230, Pa-231, Th-232, Ac-227, Ra-223, Ra-224, Ra-226, U-234, U-235, U-238	Chemical recovery of Th-230 and Pa - 231 from "Cotter Concentrate." Several rooms served as staging areas for the 55-gallon drums of "Cotter Concentrate."	1970–1979	SW-Building (SW-22, SW-140 and support rooms)	Unencapsulated material	Ac-227: 1970–1979; Pa-231: 1970–1979; Th-230: 1970–1979; Th-232: 1970–1971, 1973–1979; Ra-223, 224, 226: 1970–1979; U-234, 235, 238: 1970–1971, 1973–1979	Concentrate was processed for uranium, cobalt, copper, and nickel isotopes as early as 1971. Th/Pa analysis and separations occurred in 1971 and 1972. Production facility was completed in 1974. Th-230 production 339 g (6.85 Ci) from 1974–1979 (NIOSH 2007, p. 24). Th-230 represented over 95% of the activity. Other concerns: Th-232; Ac-227; Ra-223, 224, 226; Pa-231; U-234, 235, 238. An incident involving Ac-227 occurred on 10/30/78 in SW-132.	Nason (1972); Nason (1973); Monsanto (1975); Mound (2001); Hertz et al. 1983; NIOSH 2007; Stought (1979)

Table 1: Examples in which a Potential for Exposure to Radionuclides Existed in the Absence of Internal Monitoring Data

Radionuclides	Description	Time Period	Location/Area	Form	Gaps in Bioassay	Comments	References
Th-229, U-232, U-233, Tl-208	Thorium-229 was separated from aged U-233 and its daughters. This was done with a chloride complex separation technique.	1966–1975	SW-Building (SW-22, SW-140)	Unencapsulated material	Th-229: 1966–1975; U-232, 233: 1967–1971, 1973–1975; Tl-208: 1966–1975		Mound (2001)
Th-229, U-233	Modification of the Th-229 Separation Program where U-233 was the precursor of the process.	1975–1981	SW-Building (SW-22, SW-140)	Unencapsulated material	Th-229: 1966–1975; U-233: 1967–1971, 1973–1981		Mound (2001)
Po-209	Process to retrieve Po-209 from old Po-210 work.	1976–1979	SW-Building (SW-140, SW-132)	Unencapsulated material	Po-209: 1976–1979	Po-210 daughters are major radionuclides of concern.	Mound (2001)
Radon, Cs-137, Ra-226, Th-230, Th-232, Sr-90	Widespread contamination particularly near the old cave area.	See comment	Outside and under SW-building	Unencapsulated material; potential exposure from resuspension of soil		During characterization efforts in the late 1980s, 1990s, and early 2000, several outdoor contamination areas were identified. SW-19 was built above the entombed old cave; floor drains ran through fill, contaminating soil under SW-2 & SW-20 (adjacent rooms).	Mound 2001; Stought et al. 1988

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