

ORAU TEAM Dose Reconstruction Project for NIOSH

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Evaluation of Issues in the Use of General Area Air Sampling for Argonne National Laboratory-West Internal Dose Assessment		ORAUT-RPRT-0089 Effective Date: Supersedes:		Rev. 00 04/19/2022 None	
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ACRONYMS AND ABBREVIATIONS

AC	number of air changes
ADU	ammonium diuranate
AEC	U.S. Atomic Energy Commission
ANL	Argonne National Laboratory
ANL-W	Argonne National Laboratory-West
BZ	breathing zone
cfm	cubic feet per minute
CFR	Code of Federal Regulations
cm	centimeter
d	day
DCAS	Division of Compensation Analysis and Support
DOE	U.S. Department of Energy
dpm	disintegrations per minute
EBR-I	Experimental Breeder Reactor-I
EBR-II	Experimental Breeder Reactor-II
FASB	Fuel Assembly and Storage Building
FCF	Fuel Cycle Facility
ft	foot
g	gram
GA	general area
GM	geometric mean
GSD	geometric standard deviation
HEU	highly enriched uranium
hr	hour
in.	inch
INL	Idaho National Laboratory
ITF	Inspection and Testing Facility
kg	kilogram
LEU	low-enriched uranium
Ipm	liters per minute
m	meter
mCi	millicurie
MFP	mixed fission product
min	minute
MPC	maximum permissible concentration
MPL	maximum permissible limit
NBS	National Bureau of Standards
NIOSH	National Institute for Occupational Safety and Health
NRC	U.S. Nuclear Regulatory Commission

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NUMEC	Nuclear Materials an	d Equipment Corporat	tion	
ORAU	Oak Ridge Associate	ed Universities		
PAS pCi	personal air sampler picocurie			
RAS-TREAT RCG ROS	Reactor Analysis and radiation concentration regression on order s	d Safety Division-Trans on guide statistic	sient Reactor Experiment ar	nd Test
SAS SEC SNM SRDB Ref ID	stationary air sample Special Exposure Co special nuclear mate Site Research Datab	r bhort rial ase Reference		
μCi	microcurie Identificat	ion (number)		
UKAERE	United Kingdom Ator	nic Energy Research	Establishment	
wk	week			
yr	year			
ZPPR ZPR-III	Zero Power Plutoniu Zero Power Reactor	m (later Physics) Read III	tor	
μm	micrometer			

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1.0 PURPOSE

The National Institute for Occupational Safety and Health (NIOSH) evaluation report for Argonne National Laboratory-West (ANL-W) Special Exposure Cohort (SEC) Petition SEC-00224 (NIOSH 2016) proposed using air sampling data to bound potential unmonitored exposures to thorium, uranium, and plutonium when mixed fission products (MFPs) were not part of the radioactive source term. The areas where these potential unmonitored actinide exposures occurred are generically referred to as the "actinide-only areas" throughout this report.

The primary purpose of this report is to address issues raised by SC&A and Salient about the unmonitored actinide internal dose approaches NIOSH proposed in the SEC-00224 evaluation report. Those issues are documented in SCA-TR-2016-SEC009, *Review of Petition Evaluation Report for SEC-00224, Argonne National Laboratory-West Regarding the Use of General Area Air Sampling for Internal Dose Assessment* (SC&A and Salient 2016). Those specific issues are summarized in Section 3.0 and are primarily addressed in Sections 5.0 and 6.0.

A secondary purpose of this report is to provide additional details about the calculation of the unmonitored actinide intakes to be used for the ANL-W dose reconstructions. Those additional details are provided throughout Sections 2.2, 4.0, 5.0, 6.0, and 7.0.

As part of the efforts described above, further evaluations of the ANL-W radiological source terms and Experimental Breeder Reactor-II (EBR-II) Complex air sample data were performed, which included reviewing records that were captured after the SEC-00224 evaluation report was issued. Those evaluations indicated that some of the dates for the potential exposure periods needed to be adjusted and that the air concentrations in several areas were not bounded by 10% of the maximum permissible concentration (MPC) as indicated in the SEC-00224 evaluation report. As a result, another purpose of this report is to present the basis for revising some of the unmonitored actinide intake and internal dose approaches proposed in the SEC-00224 evaluation report. The bases for those deviations from the SEC-00224 evaluation report are provided in Sections 4.1, 4.2.2.1, and 4.2.3, and the deviations are summarized in Section 8.3.

This report does not address unmonitored actinide exposures in areas where MFPs were also present. ORAUT-TKBS-0007-5, *Idaho National Laboratory and Argonne National Laboratory-West – Occupational Internal Dose,* provides an approach for addressing those exposures (ORAUT 2010a).

2.0 BACKGROUND AND TERMINOLOGY

2.1 BACKGROUND

ANL-W facilities occupied two main areas on the Idaho National Laboratory (INL) site, the Experimental Breeder Reactor-I (EBR-I) Complex and the EBR-II Complex. Since its beginning in 1949 until February 2005, ANL-W was operated by the University of Chicago under the U.S. Department of Energy (DOE) (and its predecessors) Chicago Operations Office. In February 2005, DOE merged ANL-W with INL under the Idaho Operations Office; the operating facilities were collectively renamed as the Materials and Fuels Complex.

In addition, ANL-W was originally known as the Idaho Division (ID) of the Argonne National Laboratory (ANL) and was also referred to as the "Idaho Site" in ANL documents. In this report, the site is referred to as ANL-W.

The SEC-00224 evaluation determined that workers at the EBR-I Complex could have been exposed to dispersible forms of uranium without MFPs being present. It also determined that workers at the EBR-II Complex could have been exposed to dispersible forms of thorium, uranium, and plutonium

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without the MFPs being present. Because there are inadequate actinide bioassay data to estimate the potential intakes of thorium, uranium, and plutonium when the MFPs are not present, the SEC-00224 evaluation report recommended estimating the potential unmonitored actinide intakes for the possible periods of exposure based on the gross alpha radioactivity air sampling results for those areas. The following are brief excerpts of what was originally recommended in Sections 7.2.1.2 and 7.2.2.2 of the SEC-00224 evaluation report (NIOSH 2016).

EBR-I Complex Uranium

Because there are inadequate uranium bioassay data to estimate the potential intakes of uranium without mixed fission products at the EBR-I Complex after 1957, NIOSH will bound the potential intakes of uranium by assigning uranium intakes based on 10% of the maximum permissible concentration (MPC) that ANL-W was using. A review of the available air monitoring data indicated that the airborne alpha radioactivity at the EBR-I Complex was typically below 10% of the MPC. Because it cannot be determined when and where the workers within the EBR-I Complex worked, these uranium intakes will be assessed for all workers any time they were within the EBR-I Complex after 1957.

EBR-II Complex Uranium

The potential for uranium exposures within ZPPR [Zero Power Plutonium (later Physics) Reactor] was minimal because encapsulated and coated fuel sources were used (SRDB 138139). ZPPR Health Physics used air monitoring data and/or contamination smear data to control potential uranium exposure at this reactor during normal operations. Loading and unloading procedures were carefully monitored to detect any loose contamination on fuel plates. Exposures to uranium without mixed fission products can be bound using 10% of MPC (maximum permissible concentration) from available air monitoring data.

Because of FCF [Fuel Cycle Facility] Hot-Line start-up activities, FCF machine shop activities, and Cold-Line fuel production at the FCF, ITF [Inspection and Testing Facility], and FASB [Fuel Assembly and Storage Building], more-than-incidental intakes of depleted and enriched uranium without mixed fission products present could have occurred from August 1967 to as late as 1994. For the period of August 1967 through June 1983, no uranium bioassay data could be found for ANL-W workers. Beginning in July 1983, there is a significant increase in uranium bioassays for ANL-W workers, and the bioassay data can be used to estimate the intakes and doses from these uranium exposures.

and

Because it cannot be determined when and where the workers within the EBR-II Complex worked, these uranium intakes will be assessed for all workers any time they were within the EBR-II Complex during the period of August 1967–June 1983. For periods that workers were only assigned EBR-I Complex dosimeters, no additional uranium intakes will be assessed using this approach.

EBR-II Complex Thorium

Because there are inadequate thorium bioassay data to estimate the potential intakes of thorium without mixed fission products during the period of August 1963–November 1967, NIOSH will bound the potential intakes of thorium by assigning thorium intakes based on 10% of the maximum permissible concentration (MPC) that ANL-W was using. A review of the available air monitoring data indicated that the airborne alpha radioactivity in FCF Room 25 was typically below 10% of the MPC. Because it cannot be determined when and where the workers within the EBR-II Complex worked, these

thorium intakes will be assessed for all workers any time they were within the EBR-II Complex during the period of August 1963–November 1967.

EBR-II Complex Plutonium

The potential for plutonium exposures within ZPPR was minimal because encapsulated and coated fuel sources were used (SRDB 138139). ZPPR Health Physics used air monitoring data and/or contamination smear data to control potential plutonium exposure at this reactor during normal operations. Loading and unloading procedures were carefully monitored to detect any loose contamination on fuel plates. Exposures to plutonium without mixed fission products can be bound using 10% of MPC (maximum permissible concentration) from available air monitoring data.

Because there isn't adequate plutonium bioassay data to estimate the potential intakes of plutonium without mixed fission products during the period of April 1970–December 1972, NIOSH will estimate those intakes using the gross alpha radioactivity air sampling results that were collected during RAS-TREAT [Reactor Analysis and Safety Division-Transient Reactor Experiment and Test] sodium-loop experiment work in FCF Rooms 22 and 27. In the instances where the air samples were counted for alpha radioactivity more than once due to the presence of short-lived alpha-emitting radionuclides, the latest result for gross alpha radioactivity will be used for this approach, since isotopes of plutonium are long-lived alpha-emitting radionuclides. Because it cannot be determined when and where the workers within the EBR-II Complex worked, these plutonium intakes will be assessed for all workers any time they were within the EBR-II Complex during the period of April 1970–December 1972.

Additional details about the ANL-W site, its operations, and radiological control practices can be found in the SEC-00224 evaluation report (NIOSH 2016). Because that information is extensive, it was not included in this document for brevity. The information in the SEC-00224 evaluation report provides details about the locations, processes, and radioactive source terms for the actinide-only areas at ANL-W (NIOSH 2016). In the subsequent sections of this report, additional details are provided: 1) regarding how the SEC-00224 evaluation report recommendations above were interpreted (Sections 2.1 and 4.0), 2) how the unmonitored actinide intakes were calculated (Sections 2.2, 4.0, 5.0, 6.0, and 7.0), and 3) how they will be applied (Section 7.4).

2.2 TERMINOLOGY

The following paragraphs explain some of the terminology in this report.

The term "significant" means "practically significant" as opposed to "statistically significant." In other words, in this report "significant" means that the difference (effect size) is large enough to influence how a parameter is used or treated in practice.

The term "workroom" refers to a work location having a single discernable airspace.

For this report, breathing zone (BZ) is defined as the volume of air surrounding a worker where the characteristics of the aerosol are close to being identical to the air inhaled by the worker (NRC 1984). Thus, the results of a BZ air sample are considered to be close to being identical to the air inhaled by the worker.

The issues being addressed in this report primarily involve the relationship between the general area (GA) and BZ air concentrations. GA air concentrations are generally measured using stationary air samplers (SASs). Whereas, BZ air concentrations can be measured using SASs, portable air samplers, or personal air samplers (PASs). As indicated in NUREG/CR-4033, PAS can be

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considered an adequate BZ sampling method in almost all situations (NRC 1984, p. 17). However, SASs and portable air samplers are only considered to be BZ samplers when their sampling heads are within the BZ (NRC 1984, p. 25). A "lapel air sampler" is a specific type of PAS that has the sampling head attached to the lapel of the worker's clothing.

The relationships between BZ and GA air concentrations are typically discussed in terms of BZ-to-GA air concentration ratios, which are abbreviated as BZ:GA ratios throughout this report.

During the SEC-00224 evaluation years of 1951 to 1979, the applicable air concentration limit values were expressed in terms of maximum permissible limit (MPL), MPC, and radiation concentration guide (RCG). Based on the available Air Sample Data sheets for ANL-W, uses of the term MPL were found as early as April 1959 and as late as December 28, 1959. After that, the term MPC was used on those sheets for the period of December 29, 1959 through early 1973. By October 1973, the site was using the term RCG on the Air Sample Data sheets and continued using that term beyond the evaluation years (i.e., beyond 1979). Based on the available monthly and weekly ANL-W Health Physics reports, the term MPC was used in those reports before February 1966. From at least January 1967 through the end of the evaluation period, only the term RCG was used in the Health Physics reports, even during the periods when MPC was used in the Air Sample Data sheets. Because those terms were mostly interchangeable to ANL-W and because the values of whatever concentration limit was being applied were typically documented in the Air Sample Data sheets, only the terms MPC or air concentration limit are used in this report.

3.0 SUMMARY OF THE ISSUES

The following is a summary of the two main issues raised in Section 4.0 of SCA-TR-2016-SEC009 (SC&A and Salient 2016). Because the issues were not numbered in SCA-TR-2016-SEC009, they have been numbered below to assist with referring to and addressing a specific issue. Excerpts from Section 4.0 of SCA-TR-2016-SEC009 are in the italicized text below:

Issue 1

SC&A's review of FCF air data, typical daily operations, and assessment of NIOSH's proposed use of GA air sampling data identified two issues of concern. The <u>first</u> concern centers around the use of GA air samplers with a low airflow. Low airflow rates required sampling times of up to <u>4 days</u>, which necessarily correspond to periods when no work was performed (and no workers were present). Thus, it is reasonable to conclude that air concentrations during <u>non-working hours</u> differed significantly from air concentrations that would have exposed workers during normal facility operations that were likely limited to an 8-hour shift Monday through Friday during many periods.

Issue 2

A second and more serious concern regarding the use of GA air sampling data is the generic lack of parity between air concentrations measured by GA air samplers and lapel air samplers worn by the individual workers. Study data, including those of two nuclear fuel processing facilities cited in this review, have consistently demonstrated the poor correlation between GA and BZ air sample data with BZ/GA ratios that spanned several orders of magnitude.

In addition, Section 3.2 of SCA-TR-2016-SEC009 recommended multiplying the GA air samples results by a factor of 10 to resolve the lack of parity between GA and BZ air concentrations (SC&A and Salient 2016).

In this report, Section 4.0 provides an overview of the actinide air sampling at ANL-W. Sections 5.0 and 6.0 address the specific issues. Section 7.0 discusses intake calculation for unmonitored actinide

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intakes, and Section 8.0 provides the Oak Ridge Associated Universities (ORAU) Team's conclusions. Attachment A provides the statistical evaluation of the ANL-W EBR-II Complex air sample data, and Attachment B provides diagrams of the ANL-W actinide-only areas.

4.0 OVERVIEW OF ACTINIDE AIR SAMPLING, 1958 TO 1976

The available information indicates that ANL-W predominantly used air monitoring to demonstrate compliance with contamination control requirements, posting requirements, and for determining respiratory protection requirements. Based on the low surface contamination levels and low air monitoring results for alpha radioactivity in those areas, the probability for a significant unmonitored intake of thorium, uranium, or plutonium to have occurred would have been low.

There are five major types of air sampling: (1) fixed GA air sampling, (2) portable GA air sampling, (3) fixed BZ air sampling, (4) portable BZ air sampling, and (5) personal BZ air sampling. The term "fixed" refers to an air sampler that is not mobile and has a fixed location. The term "portable" refers to a mobile air sampler that can be relocated as needed. The term "personal" refers to a portable air sampler that is mounted somewhere on a worker. One of the most common types of PASs is the lapel air sampler where the sampling head is clipped to a worker's lapel. The U.S. Nuclear Regulatory Commission (NRC) states that air samples collected by lapel air samplers and air samplers within about 1 ft of the worker's head are considered to be representative of the BZ air (NRC 1992). As a result, the terms BZ air sample, personal air sample, and lapel air sample are interchangeable throughout this report. The order of the five types of air sampling listed above generally represents the reverse hierarchy for the preferred types of air sampling for estimating worker exposures, with personal BZ air sampling usually being the most preferred type. However, there are potentially significant sources of error unique to BZ air sampling that can generate questionable results. Those sources of error are addressed later in this document.

In the ANL-W actinide-only areas, actinide air concentrations appear to have been measured using a combination of the first four types of air sampling with fixed GA air sampling eventually becoming the most common type. For the period in question (i.e., 1958 to 1976), no indication of personal BZ air sampling was found in the available radiological safety program documents for ANL-W. However, at least some of the samples from portable air samplers were BZ air samples. A 1967 memorandum on the health physics aspects of Cold-Line operations provides more than one indication that ANL-W was sampling a worker's BZ for some of the air samples (Stoddart 1967).

A review of the gross alpha air sampling results for the ANL-W actinide-only areas indicates that the majority of the elevated sample results were due to shorter decay periods before the final count was performed on the air samples. As a result, the radioactivity on those elevated air samples was still dominated by short-lived radionuclides attributable to radon and thoron progeny. This is demonstrated in a handful of the air sample results with final recounts performed 1,000 or more minutes after the samples were first counted. Numerous samples with final recounts roughly 200 minutes after the samples were collected show the radioactivity on the air samples dropping by a factor of 2 or more. The air sample data indicates that the general practice at ANL-W was to recount the air samples only until the results dropped below the level of concern, which was usually 10% of the MPC but sometimes below 1% of the MPC. If the final counts on all of the air samples were performed several days after the air samples were collected, the mean air concentrations for ANL-W facilities would have been significantly lower. The air samples collected in the FCF were affected the most by radon and thoron progeny, which was most likely due to the much larger mass of concrete used for the construction of the FCF. When Cold-Line fuel production moved from the FCF to the new FASB, there was a significant drop in the mean air concentration for the Cold-Line fuel production. The majority of that decrease was most likely due to lower levels of radon and thoron progeny in the air at the new facility.

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Figure 4-1 provides one of the more extreme examples of how significant the short-lived radioactivity was in ANL-W air samples. It is one of the few air samples with multiple recounts going out to about 3 days. In Figure 4-1, the alpha radioactivity on the air sample decayed from 2,356 dpm to only 5 dpm over 4,245 minutes (70.8 hours), which is a reduction by a factor of 471. Most ANL-W air samples did not receive a decay count much past the 200-minute mark. For this air sample, the alpha radioactivity had reduced by a factor of 1.9 after the first recount for gross alpha radioactivity at 208 minutes.



Figure 4-1. Example of radioactive decay on an FCF air sample (ORAUT 2018a).

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4.1 SOURCE TERM AND AIR SAMPLING AT EBR-I COMPLEX, 1958 TO 1975

The period during which internal uranium exposures might not have been monitored, without MFPs being present, was from January 1, 1958, through June 13, 1975 (NIOSH 2016). Section 7.2.1.2 of the SEC-00224 evaluation report only indicated that potential intakes of uranium without mixed fission products could have occurred at the EBR-I Complex after 1957 (NIOSH 2016). That section did not provide an end date for those potential intakes. However, the end of Section 5.1.1.1 in the SEC-00224 evaluation report indicated that the decontamination and decommissioning of the EBR-I Complex was completed on June 13, 1975 (NIOSH 2016). Therefore, January 1, 1958 through June 13, 1975 was used as the period when unmonitored uranium intakes could have occurred in the EBR-I Complex. Early air sampling at the Zero Power Reactor-III (ZPR-III) was done by putting a large piece of filter paper on the (clamp-down) head of a high-volume air mover, not by permanent air samplers (ORAUT 2014a). Based on the available air sample datasheets, a variety of high- and low-volume air samplers were being used at the EBR-I Complex.

The EBR-I Complex consisted of several nuclear reactors and did not contain any radioactive material production facilities (NIOSH 2016). As indicated in the SEC-00224 evaluation report, the potential for actinide-only internal exposures at the EBR-I Complex was limited to unirradiated uranium at the ZPR-III (NIOSH 2016). The unirradiated uranium source terms at the EBR-I Complex were limited to uncoated depleted uranium metal plates, unirradiated reactor fuels, and radioactive check and calibration sources. The available information indicates that all of the EBR-I Complex reactor fuels were either clad in nonradioactive metal or coated with Teflon-based protective coating (called Kel-F) (NIOSH 2016), making them an unlikely source for internal exposures. Any uncoated or unencapsulated radioactive check and calibration sources were also an unlikely source for internal exposures due to how they would have been handled and stored to maintain their usefulness as a check and calibration source. Therefore, the primary source term for potential internal uranium exposures was the oxidized uranium on the uncoated depleted uranium metal plates. This contamination was typically found on the gloved hands of workers handling the bare plates (NIOSH 2016). However, in August 1961, all of the depleted uranium metal plates from ZPR-III were replaced with new plates that had a protective coating. The original depleted uranium metal plates were deteriorating and becoming an increasing pyrophoric hazard. The new plates had a protective coating to minimize future deterioration and pyrophoric hazards (ANL 1960–1961).

According to a former ZPR-III employee who worked as an operator, supervisor, and experimenter while there, they never saw more than trivial contamination levels of depleted uranium. The effort was put on contamination control. Air contamination never seemed to be a problem. Most of the dust was depleted uranium, but not much was suspended (ORAUT 2014a, 2015a). This information is supported by many of the gross alpha radioactivity air concentrations reported by the site for EBR-I Complex, which were often reported as being less than 1% of the MPC.

Based on the radioactive source term information and information from worker interviews, the amount of potentially dispersible uranium without MFPs being present was low, making internal uranium exposures at the EBR-I Complex unlikely.

4.2 ACTINIDE AIR SAMPLING AT EBR-II COMPLEX, 1963 TO 1976

The following sections provide information about the specific actinide-only areas at the EBR-II Complex.

Because of the number of elevated air sample results for the EBR-II Complex thorium and uranium areas and the FCF plutonium areas, a closer review of that data was performed for this report. Because the evaluated air sampling data were scattered throughout numerous documents in the Site Research Database (SRDB), all of those air sample results were put in sequential order and

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consolidated into three documents (ORAUT 2018a, 2018b, 2018c). One document contains all of the thorium-only area air data, one the uranium-only area data, and the third the plutonium-only area data for those areas. The original SRDB Reference Identification (SRDB Ref ID) numbers and page numbers were added to the top of each page in red font. The three new documents include bookmarks, highlights, and notations about how certain records were interpreted or corrected for this evaluation. The observations made during the closer review of the air sample results for the EBR-II Complex thorium and uranium areas and the FCF plutonium areas resulted in those datasets being put through a statistical evaluation.

4.2.1 <u>Thorium Areas, 1963 to 1967</u>

Unmonitored internal thorium exposures could have occurred without MFPs being present from August 1963 through November 1967 (NIOSH 2016). During this period, portable Filtronics air samplers with a flow rate of 1 cfm were used to collect air samples in the Mold Preparation Room (FCF Room 25), which was the only location where the thoria (ThO₂) was used (ORAUT 2018a).

The gross alpha radioactivity air concentrations reported by the site for FCF Room 25 were normally less than 10% of the MPC even during a thoria spill incident (ORAUT 2018a). Even then much of the radioactivity on the air samples was likely still attributable to short-lived radon and thoron progeny (ORAUT 2018a). Air concentrations in FCF Room 25 were generally low because only small quantities of thoria were being used inside of a ventilation hood. Further, the thoria was mixed with ethyl alcohol before coating the casting molds, which would have reduced its ability to become airborne (NIOSH 2016).

Figure 4-2 is the Radiation Survey Report for a September 18, 1963 thoria spill (ORAUT 2018a). Figures 4-3 and 4-4 are the air sample results for the air samples that were collected during the incident and during the cleanup, which was performed immediately after the incident. Even when thoria was spilled from the ventilation hood to the floor, the air concentrations never exceeded 10% of the MPC of 132 dpm/m³. It should be noted that ANL-W used MPCs of 66.6 dpm/m³ $(3 \times 10^{-11} \,\mu\text{Ci/cm}^3)$ and 132 dpm/m³ $(6 \times 10^{-11} \,\mu\text{Ci/cm}^3)$ for FCF Room 25 versus the MPC for ²³²Th. Based on the 1963 addendum to National Bureau of Standards (NBS) Handbook 69, the MPC for ²³²Th was 1 × 10⁻¹¹ μ Ci/cm³, which is equivalent to 22.2 dpm/m³ (NBS 1963). Document No. ORAUT-RPRT-0089

LOG NUMBER 9-7 **RADIATION SURVEY REPORT** LOCATION: FCF Room 25 DATE: 9-18-63 TIME: 1115 REQUESTOR: REDACTED SURVEYOR: REDACTED REQUEST: Monitor And Assist with thorin spill ifrom Hood onto floor, REDACTED personal Trousers + Shoes not covered by toe rubbers. RESULTS: REDACTED Trousers - 200 c/m/60 cm² ~, cleaned to Background shoes - 350 c/m/60 cm² ~, cleaned to Background Room was not surveyed before cleaning. personnel used Comfo respirators, rubber gloves distore covers. Room Floor WAS VACCUMED & wiped with Water & Rhgs. see Log book for more complete details. Room cleaned to 210 d/m/100 cm²/2 Except for two per molding on the contract of 29 d/m/200 cm²/2 Except for two per molding of the sample to the same to t INSTRUMENTS USED: Air Sample Taken: JUNO Results: see Attachments #1, 344 GM Was respiratory equipment used? Fast Neutron Slow Neutron Type: Comfo Other (Specify) PAC-3 IHS-1D-10*

Figure 4-2. Radiation Survey Report for a thoria spill (ORAUT 2018a).

				AIR	SAMPLE	DATA							
Date Time	on Time 17-63 9-12	off B-63	Run Time		Suspected A	ctivity		Pro	tection Worn		First Count	Factor	B8 =
9-18-63 14 Room F	20 113 low Rate	38	1289 min	2	1211			Supplie	ed Air		Final	Count ion of MPC	;
25 -	$M^3/hr \div 60$		A 43 / t_	MPC(40) 132	dpm//	M ³	Assault	Masks		2		Br
FCF L	cfm ÷ 35.4		_ M°/min.	MPC	40) <u>0000</u>	apm//	N	None		×	\geq	10% 🗋	
Operation Code	Sampler Type	Volum	3 G M ³					*			<	10% 🕱	X.
<u>Nowtine</u> Filter Media	al c	Counter(s	Used		/		Conve d/m/	ersion Factor M ³ x 4.55 x	10 ⁻¹³ = µ	:/cm³			
Gelmm (Sampled By:	SIASSE	KLI	Counted By:	or Ti	INA	Self-	Absorp	tion Factor:				ĸ	
F. W. La Remarks:	ee		F.W.L.	ee					8				
1×12 35,	289 4	=391	13										
		\vdash	GROSS COU	TIV Ounts	BKGD N		-	5		2 V	ity	/	
the and Tim Count Tim	tal Count	ount Time Minutes	ounts per linute	ackground er Minute	ounts per linute	ounter Yield	elf-Absorptic	isintregratio _l er Minute	/m/M ³	ercent of M	Pe of Activ	ecay Time	nitials
9-18-63 1141	15-38	5	308	41	267	18.6	-	1435	-37	~/	Br	3 min	FA
1 158	800	5	160	1	15-9	24.8	13	1034	27	20	à	15min	FA
1538	410	5	82	1	81	24.81	3	527	14	10	×	240mi	FA
9-19-63 1250	152	5	SO	/	29	2451	1,3	153	4	3_	-	1512min	Folz
						++	-						-

Figure 4-3. Air sample collected during the thoria spill (ORAUT 2018a).



Figure 4-4. Air sample collected during cleanup of the thoria spill (ORAUT 2018a).

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4.2.2 <u>Uranium Areas, 1967 to 1976</u>

The SEC-00224 evaluation report currently indicates that the period that unmonitored internal uranium exposures could have occurred without MFPs being present was from August 1967 through June 1983 (NIOSH 2016). However, the potential unmonitored uranium exposure period has been reduced to August 1967 through June 1976 for this report. This is a result of information that was captured after the SEC-00224 evaluation report was written. Previously available information combined with the new information helped to determine that the FASB only handled encapsulated uranium, which means it was not a significant internal dose concern.

Section 7.2.2.2 of the SEC-00224 evaluation report indicated that the potential for unmonitored uranium intakes at ZPPR was minimal and any unmonitored exposures could be bound using 10% of the MPC (NIOSH 2016). However, the SEC-00224 evaluation report did not provide any date range for when those minimal uranium exposures could have occurred. As part of researching what that date range should be, it was determined that the potential for the unmonitored actinide exposures at ZPPR would have only been possible when there was a cladding breach in a plutonium fuel element or uranium-plutonium fuel element. In the event of a cladding breach for a uranium-plutonium fuel element, any internal doses attributable to uranium would have been negligible compared to the internal doses attributable to plutonium. Therefore, only unmonitored plutonium intakes will be assessed for ZPPR workers.

4.2.2.1 Basis for Exposure Period Change

For the SEC-00224 evaluation report, the primary references for the FASB were the 1981 master site plan for ANL-W (ANL-W 1981), a FASB vault safety review from 1972 (Abrams 1972), and a mostly unlabeled floorplan for the FASB that was used for the radiological survey reports (ANL-W 1983). Page 17 of ANL-W (1981) indicates that the FASB was equipped for the assembly of EBR-II driverfuel pins into fuel elements, fabrication of EBR-II driver and experimental subassemblies, and inspection and storage of the elements and finished subassemblies. Page 46 of ANL-W (1981) indicates that FASB use was limited to preirradiation inspection, assembly, and testing of EBR-II fuel. A preliminary proposal from July 1969, which was not used as a reference for the SEC-00224 evaluation report because it was dated before the FASB was constructed in 1971, indicated that the FASB operations were supposed to include all of the Cold-Line fuel production processes (ANL 1969a). Because of the information in the preliminary proposal document, it was assumed for the SEC-00224 evaluation report that all of the Cold-Line processes took place at the FASB, including the ones that involved processing unencapsulated uranium.

Since the SEC-00224 evaluation report was written, two new pieces of information have been found. In Appendix E of a 1978 FASB Vault Safety Assessment, which was the firefighting plan for ANL-W, statements indicated that only encapsulated uranium was handled at the FASB (ANL-W 1978, pp. 151–153). During its Cold-Line fuel production era, the FASB consisted of three major areas: the West Room, East Room, and Storage Vault. Based on Appendix E of ANL-W (1978), the East Room was primarily for the nonradiological processes (e.g., sodium metal work and subassembly production) and the West Room was where the uranium fuel was manufactured. Appendix E of ANL-W (1978) also states, "The west room of FASB, including the vault, houses only jacketed or contained uranium alloy." It also indicates that both irradiated and unirradiated samples were examined in the West Room, which were potentially small samples of unjacketed uranium (ANL-W 1978). In addition to the 1978 FASB Vault Safety Assessment, detailed floorplans of the FASB from 1971 were captured after the SEC-00224 evaluation report was written (ANL-W 1971). Those floorplans depicted equipment layouts for the three main parts of the FASB (i.e., East Room, West Room, and Vault). There is no equipment for uranium metal alloy melting, pin casting, skull recovery

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(uranium-bearing melt residue recovery), and pin shearing operations in those diagrams (ANL-W 1971). Those were the major Cold-Line processes that involved work with unencapsulated uranium.

The continued use of FCF Room 20 and the use of vendor-produced fuel elements appear to have eliminated the need for the FASB to have any fuel casting capability. The pin casting work in FCF Room 20 was shut down in December 1969 and put on standby until 1982. The available information indicates that enough acceptable vendor-produced fuel elements (clad fuel pins) were being provided to the site by December 1969. Initially, there were some problems with the vendor producing acceptable fuel elements, which is likely why FCF Room 20 was kept on standby (ANL 1968, 1969b, 1970a, 1970b, 1970c).

A review of the available FASB air sample data indicates that the FASB gross alpha air sampling data support this determination. The 95th percentile of the unadjusted gross alpha air concentrations for the FASB is only 3.9% of the MPC. It is also worth noting that many of the higher results occurred after May 1977 when the typical sample decay period appears to have been reduced to only 23 minutes on average. Therefore, a significant fraction of the radioactivity for those samples' results is likely attributable to radon progeny. None of the FASB air samples with sample decay times of 1,440 minutes (24 hours) or greater had a result above 0.6% of the MPC (ORAUT 2021a).

A review of the available FASB survey data after 1975 indicates that the GA survey records for the FASB also support this determination. GA surveys of the FASB Storage Vault were performed daily before April 1976, twice weekly from April 1976 to February 1977, weekly from March 1977 to September 1977, and twice monthly after September 1977. For the FASB production areas, GA surveys appear to have always been performed monthly. The higher contamination survey frequency for the Storage Vault indicates that the Storage Vault was considered to be a higher risk area for contamination than the FASB production areas. With the exception of a couple FASB Vault surveys, all of the FASB GA alpha contamination survey results that were reviewed were reported as <50 dpm/100 cm², which was the minimum reporting level for alpha contamination in the surveys.

4.2.2.2 Review of Air Sampling Data

During the period from August 1967 through June 1976, air sampling data for 1970, 1973, and 1975 were more sparse than other years. This was likely due to the intermittent nature of the Cold-Line fuel production work at ANL-W and the transfer of those production facilities to the new FASB. In addition, beginning around June 1968, ANL-W started receiving vendor-produced Cold-Line fuel pins and fuel elements to initially supplement and eventually replace ANL-W's Cold-Line fuel pin and fuel production (ANL 1968). In December 1969, Cold-Line fuel pin and fuel element production at the FCF was put on standby until 1982 (NIOSH 2016). Radiological survey records indicate that Cold-Line fuel production did not start at the new FASB until at least February 1972 (NIOSH 2016). Even though some significant gross alpha radioactivity air concentrations for EBR-II Complex uranium-only areas were reported by the site, the majority of the gross alpha radioactivity air concentrations for those areas were reported as 10% of the MPC or less (ORAUT 2018b). For the uranium-only areas at the EBR-II Complex, ANL-W normally used a MPC of 132 dpm/m³ (6 × 10⁻¹¹ μ Ci/cm³) from 1967 through September 1972. After September 1972, a MPC of 220 dpm/m³ (1 × 10⁻¹⁰ μ Ci/cm³) was normally used.

Beginning in May 1975, ANL-W changed the format of the Air Sample Data sheets, and the Time On and Time Off information was no longer being routinely reported for the air samples. However, the Run Time for the air samples was still being reported. Based on the few records with sample on/off information in the Remarks column of the new form, the Date and Time column on the new forms only provide the sample count dates and times. Figure 4-5 is an example of the revised Air Sample Data sheets (ORAUT 2018b). Also by May 1975, routine air sample collection times at FCF periodically began going up to and over 10,080 minutes (~7 days).

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Figure 4-5. Example of a post-April 1975 air sample record (ORAUT 2018b).

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4.2.3 Plutonium Areas, 1970 to 1975

At the EBR-II Complex, there were two distinctly separate sets of plutonium work being performed: one at ZPPR and the other at the FCF.

4.2.3.1 Zero Power Plutonium Reactor

Section 7.2.2.2 of the SEC-00224 evaluation report currently states the following (NIOSH 2016):

The potential for plutonium exposures within ZPPR was minimal because encapsulated and coated fuel sources were used. ZPPR Health Physics used air monitoring data and/or contamination smear data to control potential plutonium exposure at this reactor during normal operations. Loading and unloading procedures were carefully monitored to detect any loose contamination on fuel plates. Exposures to plutonium without mixed fission products can be bound using 10% of MPC (maximum permissible concentration) from available air monitoring data.

Based on those statements, potential unmonitored intakes of plutonium without MFPs at ZPPR will be based on 10% of the MPC value being used for the air monitoring program at ZPPR. Based on the available air monitoring data, an MPC of 4.4 dpm/m³ ($2 \times 10^{-12} \mu$ Ci/m³) was used for the airborne alpha radioactivity at ZPPR, which makes the bounding air concentration 0.44 dpm/m³.

Because the SEC-00224 evaluation report did not provide a date range for when those potential unmonitored intakes were possible, a basis for that date range has been provided as part of this report. The earliest indication of plutonium contamination at ZPPR was during the October 9, 1968, inspection on the receipt of the original driver fuel. The receipt inspection of that fuel found low levels of plutonium contamination on two fuel elements (Koplin 1975). However, ANL-W procedures indicate plutonium intakes were unlikely during the receipt inspections because the workers wore respirators and because the fuel was handled inside of a hood until it was confirmed as being free of contamination (ANL-W 1968a, 1968b). Therefore, plutonium found during receipt inspections was not considered a potential intake hazard.

On September 1 and 2, 1970, a reactor safety survey was performed at ZPPR. Section C.4, "Health Physics Procedures," of the report for that safety survey indicated that no leaky fuel had been encountered during the entire operating period of the ZPPR. However, during the safety survey, a minor plutonium contamination incident occurred. Some plutonium came loose from a small plutonium source on a planchet that was used to check plutonium monitor calibrations. The report also indicated that only the source storage container was contaminated (ANL-W 1970). Based on the available information, this plutonium contamination was not likely airborne or an intake hazard. In addition, the plutonium contamination was likely in the form of nonrespirable flakes. Even though a plutonium intake was unlikely from this incident, September 1, 1970, will be used for the start date for the potential unmonitored plutonium intakes at ZPPR.

On July 31, 1975, a plutonium bioassay program was initiated for ANL-W workers (Madison 1975). Therefore, the period that unmonitored intakes of plutonium without MFPs were potentially received at ZPPR is assumed to be September 1, 1970, through July 31, 1975.

4.2.3.2 Fuel Cycle Facility Areas

The SEC-00224 evaluation report currently indicates that the period when unmonitored internal plutonium exposures could have occurred without MFPs being present was April 1970 through December 1972 for FCF. The date range for the potential plutonium exposure period at the FCF was based on the period when the Mark-II type RAS-TREAT sodium-loop experiment work was being

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performed at the FCF (NIOSH 2016). However, the potential unmonitored plutonium exposure period for the FCF work has been extended through April 1973 for this report.

At the FCF, the Mark-II type RAS-TREAT sodium-loop work began in April 1970 and was initially limited to Rooms 27, 28, and 29. In early 1971, a glovebox was installed in Room 22 (i.e., the former FCF Machine Shop) to handle the Mark-II type RAS-TREAT sodium-loop experiments. The glovebox became operational sometime after May 1971. After 1972, RAS-TREAT sodium-loop experiment work at the FCF appears to have been performed exclusively in the Argon Cell, and the experiments were irradiated to much higher levels (Cook et al. 1975). However, additional plutonium air monitoring data for FCF Room 22 was found after the SEC-00224 evaluation report was completed. The more recently discovered data indicates that plutonium exposures without the MFPs were likely possible at the FCF through April 1973 (ORAUT 2018c). Plutonium air monitoring was likely performed in FCF Room 22 until the room was decontaminated. Therefore, the potential period for unmonitored plutonium exposures at the FCF without MFPs has been extended through April 1973.

Based on the available air monitoring data, ANL-W used an MPC of 4.4 dpm/m³ (2 × 10⁻¹² μ Ci/m³) for the FCF plutonium areas. With the exception of the FCF sodium-loop work areas during 1970 to 1973, the gross alpha radioactivity air concentrations reported by the site for plutonium-only areas were normally less than 10% of the MPC (ORAUT 2018c). However, some of the elevated air sample results during those years were likely attributable to short-lived radon and thoron progeny. Based on the decay times for the final counts on the elevated samples, the final counts for nearly all of the elevated air samples were performed before all of the short-lived radon and thoron progeny could have decayed. The final counts for the air samples should have been performed after a 3.5-day (5,110-minute) decay time to provide sufficient time for all of the short-lived radon and thoron progeny to decay out of the air samples. Additionally, some of the elevated results could have been attributable to uranium, since uranium work was also periodically performed in those FCF rooms. All but one of those results were below 10% of the MPC for uranium.

Until June 1972, portable Filtronics air samplers with a flow rate of 1 cfm were used to collect air samples in the plutonium area (FCF Room 22) (ORAUT 2018c). After June 1972, a special alpha sampler with a flow rate of 1 cfm was used in the plutonium area (ORAUT 2018c).

5.0 RESPONSE TO ISSUE 1

There are two distinctly separate parts to Issue 1 in SCA-TR-2016-SEC009 (SC&A and Salient 2016). Because of that, each of those parts is addressed separately below.

5.1 PART 1 OF ISSUE 1

The <u>first</u> concern centers around the use of GA air samplers with a low airflow. Low airflow rates required sampling times of up to <u>4 days</u>, which necessarily correspond to periods when no work was performed (and no workers were present).

The ORAU Team is not certain how to address the first part of Issue 1 because lapel samplers typically have much lower flow rates and because it is contradictory to the argument in SCA-TR-2016-SEC009 that only lapel air samplers can provide representative BZ air samples (SC&A and Salient 2016).

At ANL-W, the air samplers in the thorium and uranium areas had flow rates typically ranging between 1.0 and 2.0 cfm, and the air samplers in the plutonium areas had flow rates of 1 cfm. As a comparison, most lapel samplers only operate at 2 lpm, which is equivalent to 0.07 cfm. Minimum detectable concentration calculations, based on the site's 5-minute count time, indicate that the ANL-W air samplers could detect <10% of the MPC for an 8-hour sample period and could detect

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nearly 10% of the MPC for a 4-hour sample period. Whereas, a lapel sampler would need to sample for approximately 60 hours to detect 10% of the MPC with a 5-minute count time, or sample for 24 hours to detect 10% of the MPC with a 60-minute count time. Therefore, the concern that low airflow rates required sampling times of up to 4 days for GA air samples is not substantiated.

5.2 PART 2 OF ISSUE 1

... Thus, it is reasonable to conclude that air concentrations during <u>non-working hours</u> differed significantly from air concentrations that would have exposed workers during normal facility operations that were likely limited to an 8-hour shift Monday through Friday during many periods.

The ORAU Team has evaluated this and agrees it is a legitimate concern, even though a number of factors would mitigate any biases that are not favorable to claimants by using the reported air concentrations as is. The following are a few of the key factors that would mitigate those potential biases:

- 1. Typical low levels of removable contamination in those areas are not indicative of areas with significant levels of airborne radioactivity during operational or nonoperational periods,
- 2. Not accounting for the actual occupancy in those areas,
- 3. Not accounting for the respirable fraction of the airborne radioactivity,
- 4. Not accounting for the times respirators were worn, and
- 5. ANL-W not allowing for adequate decay of the short-lived radionuclides before performing final counts on the air samples.

With two exceptions, the ANL-W air sampling data for the actinide-only areas were evaluated, and calculations were performed, to address the air sample dilution concern. The air sampling data for the EBR-I Complex were not evaluated or adjusted because of the small potential for internal uranium exposures at the EBR-I Complex. As indicated in Section 4.1, very little of the uranium in the EBR-I Complex was dispersible. The air sampling data for the EBR-II Complex ZPPR were not evaluated or adjusted because of the small potential for internal plutonium exposures at ZPPR. As indicated in Section 7.2.2.2 of the SEC-00224 evaluation report, the potential for plutonium exposures within ZPPR was minimal because encapsulated and coated fuel sources were used (NIOSH 2016). These determinations are also supported by EBR-I Complex and ZPPR air sampling data, which indicate that significant levels of airborne uranium and plutonium contamination without MFPs present were rare and that air sampling appears to have mostly been performed as a precautionary measure. Therefore, it is reasonable to bound the potential unmonitored uranium and plutonium intakes attributable to those facilities based on an average concentration of 10% of the MPC without further adjustment.

Only the gross alpha radioactivity air concentrations were evaluated because potential unmonitored actinide exposures when MFPs were not present was the only unmonitored internal dose concern at the EBR-II Complex. As indicated in Section 4.2, the evaluated air sampling data for each area were put in sequential order and consolidated into three documents (ORAUT 2018a, 2018b, 2018c). Additionally, a review of the available air sampling data indicates that ANL-W inconsistently applied its alpha self-absorption factor of 1.3 to the air sample results for alpha radioactivity. Therefore, whenever the air sample results indicate that no alpha self-absorption factor was applied or was not known to have been applied (as in the monthly Health Physics Reports), those results were multiplied by a factor of 1.3.

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For the four air sample datasets that were evaluated and adjusted for potential dilution, statistical evaluations were performed on each of those datasets to verify that their distributions could be represented by a lognormal distribution. The four air sample datasets included the air sample results for the EBR-II Complex thorium and uranium areas and the FCF plutonium areas, with the air sample results for the uranium areas being divided into two separate datasets. Because all four of those air sample datasets had a reasonable fit to a lognormal distribution and because the sample times varied significantly with the shortest duration air samples often having the highest results, time-weighted geometric mean (GM) air concentrations were calculated for each of those air sample datasets. In the instances of overlapping sample periods that were from different work areas for a given dataset, no samples were excluded from the calculation of the time-weighted GM air concentration, even though a worker could not have been simultaneously at both locations. The primary reasons for not excluding overlapping air samples include partial overlaps and differing sample run times. Attachment A provides the details on how those statistical evaluations were performed along with the resulting timeweighted GM air concentrations and geometric standard deviations (GSDs) for each of those datasets. Sections 5.2.1 and 5.2.2 provide more details about the two different approaches that were used to adjust the four air sample datasets for potential dilution.

5.2.1 EBR-II Complex Air Sampling Data, 1963 through 1974

With the exception of the ZPPR data, all of the available air sampling data from the actinide-only areas for August 1963 through December 1974 were evaluated. As discussed above, the ZPPR air sample data were not evaluated or adjusted because of the small potential for internal plutonium exposures at ZPPR. With the exception of short duration samples, all of the air concentrations for this period were adjusted to eliminate sample dilution from sampling during nonoperational periods. To do that, the concentrations of airborne alpha radioactivity were assumed to drop to zero during the nonoperational periods. The adjusted air concentrations were calculated based on the volumes of air sampled during the operational periods. Those adjusted air sample volumes were calculated using the reported sampler flow rates and calculated operational times, which are referred to as potential exposure times below and in the calculations (ORAUT 2021a, 2021b, 2021c).

Based on an FCF worker interview, a second work shift was started at the FCF in the late 1960s that continued until the Hot Fuel Examination Facility-North came online in March 1975 (ORAUT 2015b; NIOSH 2016). Based on sample start and end times and notes in many of the air sample records, two 8-hour shifts were being worked at the FCF by August 1967. Therefore, assuming 16 hours of exposure time for each full workday is considered reasonable for the period of August 1967 through March 1975. For all other periods, a single 8-hour shift was assumed for each workday.

Given that ANL-W normally exchanged air samples on Monday to Friday during the first or second shift, short duration samples were likely collected outside of that period because of some radiological work being performed during off-shift hours. Adjusting the short duration samples to eliminate sample dilution could cause the results for the samples collected during off-shift hours to be zeroed out or inappropriately biased high. Therefore, the short duration samples with sample times of 18 or less hours were not adjusted to eliminate potential sample dilution. The up to 18 hours assumes that two 8-hour shifts were worked and allows for up to 2 extra hours for additional work outside of the scheduled shifts (e.g., performing radiological surveys, collecting air samples).

For sample times greater than 18 hours, up to 16 hours of exposure time was assumed for each full workday (Monday through Friday, excluding federal holidays). Only the observed federal holidays for a given year were accounted for in these calculations because no information was found about which holidays were observed by ANL-W. The amount of time worked during partial workdays was calculated based on the sample exchange times and the assumed workday start and end times. The available records indicate that the workday started at 8:00 a.m.

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For FCF air sample results that are only summarized in the monthly health physics reports, all exposure periods for those air samples were assumed to be 16 hours, unless additional information indicated otherwise. This was assumed for the period before March 1975, because the available information indicated that two 8-hour shifts were being worked at the FCF from August 1967 to March 1975 (ORAUT 2015b; NIOSH 2016). After that, FCF operations returned to a single day shift (ORAUT 2015b; NIOSH 2016). However, this exposure period assumption was not reduced to 8 hours, because no summarized air sample results from the monthly health physics reports were used after 1969.

Equations 5-1 and 5-2 are provided to better illustrate how these air concentration adjustments were performed.

$$T_{\rm O} = T_{\rm S} - T_{\rm N} \tag{5-1}$$

where

To	=	total operating time during the sampling period (min)
Ts	=	total sampler run time during the sampling period (min)
T_N	=	total nonoperational time during the sampling period (min)

Note that the calculation of T_N is actually rather complicated. For more details on how T_N was calculated please refer to the spreadsheets for those calculations, which are in ORAUT (2021a, 2021b, and 2021c).

$$C_{\text{Adj}} = \frac{(A)(AF)(CF)}{(F)(T_{\text{O}})}$$
(5-2)

where

C_{Adj}	=	adjusted GA air concentration (dpm/m ³)
A	=	gross alpha radioactivity on the sample (dpm)
F	=	sampler flow rate (cfm)
To	=	total operating time during the sampling period (min)
AF	=	1.3 self-absorption factor for the measured alpha radioactivity, but only included it in these calculations when it was omitted in the results reported by ANL-W. The ANL-W Air Sample Data sheets clearly indicate when this factor was or
CF	=	volume conversion factor used by ANL-W (35.4 ft ³ /m ³)

5.2.2 EBR-II Complex Air Sampling Data, 1975 to 1976

As previously indicated, beginning in May 1975 ANL-W changed the format of the Air Sample Data sheets; air sample on/off dates and times were no longer routinely reported. Because of that, the air sample results from January 1975 through June 1976 could not be adjusted to eliminate the influence of the nonoperational periods from the samples. Therefore, instead of adjusting the air concentrations, the intake calculations were modified to address the air sample dilution concern. In the approach that involved adjusting the air sample results, a major assumption for that approach was that the airborne radioactivity dropped to zero during the nonoperational periods at the facility. Using that same assumption, the sample dilution concerns can be eliminated by assuming that the workers were exposed to unadjusted air concentrations day and night for 1 year (24 hr/d 365 d/yr). It should be noted that this approach only works when the daily operational period for the facility is the same as the daily exposure period assumed for the workers (i.e., no second or third shifts at the facility). For

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the period that this approach was being used, the FCF was only operating for one work shift per workday. For more details on this approach, including the equations that were used, refer to Section 7.1.2.2.

6.0 RESPONSE TO ISSUE 2

A second and more serious concern regarding the use of GA air sampling data is the generic lack of parity between air concentrations measured by GA air samplers and lapel air samplers worn by the individual workers. Study data, including those of two nuclear fuel processing facilities cited in this review, have consistently demonstrated the poor correlation between GA and BZ air sample data with BZ/GA ratios that spanned several orders of magnitude.

The end of Section 3.2 in SCA-TR-2016-SEC009 suggests applying a factor of 10 to convert all GA air concentrations to BZ air concentrations as a means for addressing the lack of parity between GA and BZ air concentrations (SC&A and Salient 2016). The suggested factor of 10 in SCA-TR-2016-SEC009 is based on the data from only two studies (Brunskill and Holt 1967; Caldwell, Potter, and Schnell 1967), both of which were completed in 1967. Those studies represent two of the earliest known studies involving lapel air samplers, which is a specific type of BZ PAS device. Because many other air sampling studies exist, most of which are more recent than 1967, this report evaluated the results from some of those studies. The results from those studies are discussed in the subsections below.

6.1 APPLICABILITY OF STUDIES FROM OTHER SITES

There are several highly variable parameters that can affect the outcome of air sampling studies, often making most of their results and conclusions only applicable to the specific conditions and location of the study. Therefore, it is important to determine the applicability of an air sampling study from one site or workroom before applying its results to another site or workroom.

6.1.1 Key Parameters in ORAUT-RPRT-0097

ORAUT-RPRT-0097, *Breathing Zone to General Area Air Concentration Ratios in Small Workrooms* (ORAUT 2021d), identifies several parameters that can influence the BZ:GA ratios in a workroom (see Section 2.0 of that reference). As indicated in Section 11.0 of ORAUT-RPRT-0097, there are five key parameters that should be considered when justifying the application of BZ:GA ratios from other locations to the location of interest. Those parameters include (1) room size, (2) particle size distribution for the airborne radioactivity, (3) ventilation rate for the room, (4) room complexity, and (5) the presence of dominant particles. The following partial excerpts from Section 2.0 of ORAUT-RPRT-0097 briefly explain how each of those parameters can affect the air concentrations and/or BZ:GA ratios for a workroom. More detailed discussions can be found in ORAUT-RPRT-0097.

Total workroom or air space volume is a key parameter that usually influences how much an aerosol can or cannot be dispersed. However, with the exception of rooms where elevated releases above the typical BZ can occur, a room's height is usually less of a factor influencing the aerosol concentration than its other dimensions because of gravitational settling. Therefore, in most instances room sizes can be compared in terms of area for determining applicability of the BZ:GA ratio information in this report to a specific room (ORAUT 2021d). At ANL-W, the actinide-only areas were relatively small workrooms in comparison with the actinide handling and processing areas at other sites. Table 6-1 provides the room size information for each of the areas where unmonitored actinide intakes need to be assessed per the SEC-00224 evaluation report (NIOSH 2016). Note that Table 6-1 does not contain any area room size information for the Low Bay Area at the ITF, because no dimensions or scale drawings were found for that area. The only known size information for the

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ITF is for the size of the entire building. Section 5.1.2.5 of the SEC-00224 evaluation report indicates that the ITF building had a total square footage of 3,455 ft² (320 m²) (NIOSH 2016).

	Total area	Total area
Process area (actinide)	(ft²)	(m²)
FCF Room 20 (U)	672	62.4
FCF Room 22 (U)	672	62.4
FCF Room 23 (U)	528	49.1
FCF Room 25 (Th)	369	34.3
FCF Room 26 (U)	579	53.7
FCF Room 27 (Pu)	222	20.6
FCF Room 28 (Pu)	479	44.5
FCF Room 29 (Pu)	994	92.3
ITF Low Bay Area (U)	Unknown	Unknown
ZPR-III Workroom (U)	303	28.1
ZPPR Workroom (Pu)	1,662	154.4

Table 6-1. ANL-W actinide-only area room sizes (ORAUT 2019).

Because only respirable size particles (i.e., those particles with aerodynamic diameters smaller than about 20 µm) contribute to inhalation intakes, only the respirable fraction of an aerosol is of interest for assessing the inhalation intakes of workers. Therefore, the ideal air sample measurements for assessing a worker's inhalation exposure would discriminate against collecting nonrespirable particles. The presence of larger nonrespirable particles on any air sample being used to estimate a worker's inhalation intake, whether it is a BZ or GA air sample, could result in an unreasonable overestimate of the worker's intake (ORAUT 2021d).

The ventilation rate in terms of air changes (ACs) per unit time, usually in units of AC/hr, is a removal constant for the aerosol within a workroom. This removal constant is directly related to the depletion of the aerosol from an acute, constant, or variable release of aerosol and can be used to estimate the average air concentration at different points in time when the release quantity (instantaneous or puff releases) or release rate (longer term releases) along with several other parameters are known (ORAUT 2021d).

Factors contributing to the complexity of a workroom include room layout, obstructions, heat sources, room ventilation inlet and outlet locations, local exhaust locations, and general flow directions. All of these affect the airflow patterns and level of mixing within a room. These things tend to cause air streaming, dead air spaces, eddy currents, etc. within a room, which result in larger air concentration gradients. The more complex a room is, the more likely larger air concentration gradients are (ORAUT 2021d). To facilitate evaluating the complexity of the actinide-only areas at ANL-W, the available floorplans were compiled and provided in Attachment B.

Low numbers of airborne radioactive particles in a workroom are normally a good thing and a desirable condition for the workplace atmosphere. However, when those low-in-number radioactive particles have a relatively high specific activity and radiotoxicity (i.e., what this report refers to as "dominant particles"), that condition can become very problematic and have a very adverse effect on the ability to collect meaningful and reproducible air samples. When the numbers of the radioactive particles are too low, both BZ and GA air sampling becomes a stochastic process versus a deterministic process because there just are not enough radioactive particles to go around. Because this normally is not a significant issue of concern for lower specific activity and less radiotoxic particles, this situation is primarily relevant to when the low-in-number radioactive particles have a relatively high specific activity and radiotoxicity (ORAUT 2021d).

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6.1.2 <u>Air Concentration Levels</u>

In addition to the parameters above, higher maximum air concentrations can cause larger discrepancies between BZ and GA air sample results. For example, if the highest air concentration in a room is only 1 MPC, the potential air concentrations in the room can only range from zero to 1 MPC. Whereas, when the highest air concentration is only 10% of the MPC, the range is only from zero to 10% MPC, which means the potential differences between the GA and BZ air concentrations would also be smaller. The graphs presented in Caldwell, Potter, and Schnell (1967) and Brunskill and Holt (1967) for the Nuclear Materials and Equipment Corporation (NUMEC) plutonium laboratory and the Windscale Works show this effect. Those graphs are presented in this report as Figures 6-1 and 6-2. As indicated in those graphs, the BZ:GA ratios tend to be closer to unity when the air concentrations are low, even for larger workrooms. Given that the reported air concentrations for the ANL-W were generally low (ORAUT 2018a, 2018b, and 2018c), air sampling studies performed in facilities with significantly higher air concentrations would likely overestimate the BZ:GA ratios for the actinide-only workrooms at ANL-W. Summaries of the air concentrations for the ANL-W actinide-only areas and applicable periods are provided in Section 7.2.



Figure 6-1. BZ:GA ratio by BZ air concentration at the NUMEC plutonium laboratory (Caldwell, Potter, and Schnell 1967).



Figure 6-2. BZ:GA ratio by BZ air concentration at Windscale Works (Brunskill and Holt 1967).

6.2 STUDIES REFERENCED BY SCA-TR-2016-SEC009

SCA-TR-2016-SEC009 (SC&A and Salient 2016) made recommendations based on the information in the two studies Brunskill and Holt (1967) and Caldwell, Potter, and Schnell (1967). However, SCA-TR-2016-SEC009 does not provide a basis for why those two studies are applicable to the actinide-only areas at ANL-W.

6.2.1 Caldwell, Potter, and Schnell Study

The Caldwell, Potter, and Schnell (1967) study was performed at the Nuclear Materials and Equipment Corporation (NUMEC) uranium plant and plutonium laboratory. Based on ORAUT-TKBS-0041, *Site Profile for Nuclear Materials and Equipment Corporation, Apollo and Parks Township, Pennsylvania*, the uranium plant was at the NUMEC Apollo Site and the plutonium laboratory was at the Parks Township Site (ORAUT 2017). Additional information about the NUMEC uranium and plutonium facilities are summarized in the sections below. Note that, when possible, these summaries only provide information that is relevant in time to the 1967 study (i.e., around the years of 1966 and 1967).

6.2.1.1 Description of the NUMEC Apollo Site

By 1963, the majority of the Apollo Site was dedicated to the production of low-enriched uranium (LEU) and highly enriched uranium (HEU) fuels. The major activities at the Apollo Site included (1) the conversion of LEU hexafluoride (UF₆) to uranium oxide (UO₂) for use in light-water-moderated reactors, (2) the production of HEU nuclear fuel for use in the naval reactors program, and (3) the processing of unirradiated uranium scrap (including LEU and HEU) from the U.S. Atomic Energy Commission (AEC) in the 1960s (ORAUT 2017).

Production of UO₃ from UF₆ began with UF₆ being converted to UO₃ in the Chemical Conversion Room. The UF₆ gas was hydrolyzed to an aqueous solution of UO₂F₂ and HF. This solution was

reacted with NH₄OH to form a slurry of ammonium diuranate (ADU) $[(NH_4)_2U_2O_7]$. The slurry was then pumped through a hooded pressure filter. The filter cake was transferred to drying hoods where the ADU was decomposed to UO₃ at a controlled temperature. The UO₃ product was transferred in small polyethylene containers to the Ceramics Fabrication Area for further processing and conversion into UO₂. By August 1960, the filter cake was being dried by a rotary kiln rather than the earlier fry pan method. By June 1961, a calciner had been added for reduction of ADU to U₃O₈. The dried U₃O₈ cake was discharged directly from the kiln into a container, eliminating the manual transfer (ORAUT 2017).

Reduction of UO₃ to UO₂ was performed in the Ceramics Fabrication Area. UO₂ product was transferred to the Ceramic Laboratory for additional fabrication. The UO₃ was reduced to UO₂ in a rotary kiln rather than the reduction furnace that had been used earlier. Ceramics fabrication was performed in the Ceramics Fabrication Area where UO₂ was hammer-milled in a ventilated enclosure and then moved to the blender glovebox where Aerowax was added and the mixture was blended. The wax-UO₂ mixture was then pressed into a cake in a Drake press. The cake was placed in a glovebox where it was granulated by hand with screens to give the desired particle size. The UO₂ was loaded into shallow metal pans called "firing boats" and sintered. Sintered UO₂ was classified, weighed, and packaged (ORAUT 2017).

Uranium scrap was dissolved in two designated areas, CRP-2 and CRP-3. The product solutions from the dissolution methods were processed to generate insoluble UF₄ and ultimately converted into U_3O_8 or UO_2 as the final product (ORAUT 2017).

Table 6-2 provides a summary of the maximum possession limits for special nuclear material (SNM) for the Apollo Site during the 1972 timeframe. Note that these are the maximum possession limits, which could be much greater than the actual quantities that were used. The typical amounts of uranium in use in an area ranged from milligrams to hundreds of kilograms (ORAUT 2017).

		Maximum
Areas	Source/chemical or physical form	possession
Processing areas, laboratories, and vaults	U-235 enrichment >5%	5,000 kg
Processing areas, laboratories, and vaults	U-235 enrichment ≤5%	75,000 kg
Processing areas, laboratories, and vaults	Plutonium as fully clad or encapsulated material	500 kg
Mass Spectrometry Laboratory	Uranium in any enrichment	350 kg
Mass Spectrometry Laboratory	Plutonium in any form	0.5 g
Low-level radioactive waste storage areas	U-235 within fenced area in approved storage containers	35 g
Low-level radioactive waste storage areas	U-235 in buildings meeting safeguards and security requirements	50 kg
Nuclear Decontamination Corporation	Any byproduct material	20 mCi
Nuclear Decontamination Corporation	Any source material	20 g
Nuclear Decontamination Corporation	Any SNM	20 mCi

Table 6-2. SNM possession limits for NUMEC Apollo Site (ORAUT 2017).

6.2.1.2 Description of the NUMEC Parks Township Site

By 1967, the Parks Township Site primarily consisted of two processing buildings, Building A (the Plutonium Plant) and Building B (the Metals Complex). The original Building A was 20,000 ft². From 1961 through 1970, a major expansion of Building A was completed in five separate expansions that eventually increased the footprint of Building A to 61,000 ft² (ORAUT 2017). Caldwell, Potter, and Schnell (1967) indicated that the plutonium laboratory was 20,000 ft². Based on that, Caldwell, Potter, and Schnell (1967) appear to only have used Building A for their study. Therefore, only the applicable

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information for the portions of Building A that existed before and during the study (i.e., before 1968) are provided in this section.

Throughout its history, Building A has been referred to as the Plutonium Plant, Plutonium Laboratory, Plutonium Facility, Plutonium Processing Facility, Plutonium Building, and the NUMEC Advanced Material Center. The original portion of Building A was designed as a plutonium laboratory to perform research and development that led to plutonium-based products (ORAUT 2017). The main workrooms in Building A were called fabrication areas, which were also referred to as Fabs or FABs for short. Based on the information in a 1979 NRC licensing revision for SNM-414, FAB-1 through FAB-5 were the only plutonium production areas present at the time of the 1967 air sampling study. FAB-6 through FAB-9 were not added to the Plutonium Plant until 1968 and later (Austin 1979).

The initial functions of the Parks Township facilities included plutonium fuel fabrication, HEU fuel preparation, and the production of zirconium-hafnium bars. That work was performed under the AEC and later under NRC License SNM-414, which allowed the handling of plutonium already on the site. Before 1968, various plutonium conversion processes were performed in FAB-1 along with some repair and maintenance work on contaminated equipment. In FAB-2, plutonium fuel fabrication work was performed for the ZPR-III and ZPPR. Before 1968, FAB-3 was primarily used for metallography and some unspecified manufacturing operations that involved plutonium. The radioactive materials handled in FAB-1 through FAB-3 included plutonium metal, plutonium nitrates, plutonium oxalates, plutonium oxide (PuO₂), plutonium-beryllium compacted powder, americium metal, and depleted uranium oxide (UO₂) (ORAUT 2017). The chemical compound formulas were not provided for the plutonium nitrates and plutonium oxalates because they vary depending on the valence state of the plutonium.

Before 1968, FAB-4 was primarily used for alpha, beta, gamma, and neutron source fabrication involving various or unspecified forms of ⁷Be, ⁶⁰Co, ¹⁹²Ir, ²¹⁰Po, plutonium, and americium. FAB-5 was used for all forms of plutonium scrap recovery and analytical laboratory work (ORAUT 2017).

In the late 1960s, the Parks Township Site made an unspecified number of plutonium fuel wafers for ZPR-III and 11,500 plutonium fuel plates for the ZPPR, which were both at ANL-W. An additional 700 plutonium fuel plates were manufactured for ZPPR, but not until after 1967. The ZPPR fuel plates were plutonium-uranium-molybdenum ternary alloy plates encapsulated in stainless steel using fuel-grade plutonium (11.5% ²⁴⁰Pu) and depleted uranium. From 1961 through 1969, the SNM possession limit for Building A was any combination of plutonium and ²³⁵U up to 400 kg (ORAUT 2017). Based on Schedules A and C in the AEC plutonium supply contract with NUMEC, NUMEC was committed to processing between 100 and 160 kg of plutonium during most months for just the ZPPR fuel production effort (Shapiro 1967).

6.2.1.3 Applicability of NUMEC Sites to ANL-W Actinide-Only Areas

Caldwell, Potter, and Schnell (1967) indicated that the uranium plant was 40,000 ft² and the plutonium laboratory was 20,000 ft². Further research indicates that the 20,000 ft² and 40,000 ft² values likely represent the footprints for the entire processing areas during the time of the study versus the individual workrooms. As indicated above, the 20,000 ft² appears specifically to be the footprint for Building A at the Parks Township Site during that time. That further research also confirmed that the majority of the individual workroom sizes for the uranium and plutonium processing areas at the NUMEC facilities were significantly larger than the actinide-only areas at ANL-W. Table 6-3 summarizes the uranium processing room sizes at the Parks Township Site (ORAUT 2021e).

Process area ^a	Total area (ft ²)	Total area (m ²)
CP-1	6,652	618.0
CRP-1	2,500	232.3
PC-2	785	72.9
PC-3	1,329	123.4
CF-1	1,890	175.6
CRP-4	1,470	136.6
CRP-2/CP-2/CRP-3 (2nd floor) ^b	7,347	682.6
CP-1 (2nd floor)	1,920	178.4

Table 6-3. NUMEC Apollo facility room sizes (ORAUT 2021e).

a. All processing areas are on the first floor, unless indicated otherwise.

b. No drawings depicting walls between these three processing areas were found. Therefore, the available information indicated that they shared the same air space.

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2021e).				
I able 6-4.	NUMEC Parks	Iownship	facility room s	izes (ORAUT

Process area	Total area (ft ²)	Total area (m²)
FAB-1	4,951	459.9
FAB-2	1,934	179.6
FAB-3	3,355	311.7
FAB-4	806	74.9
FAB-5	2,182	202.7

As indicated in Table 6-1, the workroom sizes for the actinide-only areas at ANL-W were generally 92.3 m² (994 ft²) or less. The ZPPR Workroom at 154.4 m² (1,662 ft²) and potentially the ITF Low Bay Area were the only exceptions to that, but the air concentrations in those two areas were low. As indicated in Section 6.1.2, BZ:GA ratios tend to be closer to unity when the air concentrations are low, even for larger workrooms.

Based on the available information for the NUMEC workrooms involved in the Caldwell, Potter, and Schnell (1967) study, the data from that study is not applicable to the ANL-W actinide-only areas for the following reasons:

- 1. Most of the NUMEC workrooms were much larger than the ANL-W actinide-only areas.
- 2. The majority of reported air concentrations for the NUMEC workrooms were significantly higher than the air concentrations in the ANL-W actinide-only areas.
- 3. The processes at NUMEC involved relatively large quantities of dispersible uranium and plutonium; whereas, the processes at ANL-W worked with much smaller quantities. Because of that, much greater air concentration gradients were possible at NUMEC than at ANL-W. Note that at both NUMEC and ANL-W used engineering controls (e.g., gloveboxes, ventilation hood, etc...) to protect the workers from the dispersible forms of plutonium. Therefore, airborne plutonium in those workrooms was normally only possible when there was some degree of failure in those engineering controls.
- 4. The particle size distributions are unknown for the NUMEC workrooms. Therefore, it is possible that the air samples were influenced by nonrespirable radioactive particles.

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6.2.2 Brunskill and Holt Study

The Brunskill and Holt (1967) study was performed at the plutonium and uranium plants at the Windscale and Springfields Works in the United Kingdom. The study was performed in a number of plant and laboratory areas at those two sites. At the time of the study, both sites were under the United Kingdom Atomic Energy Authority. In 1981, Windscale was renamed Sellafield.

In general, there is insufficient information about the processes and workrooms at the Windscale and Springfields Works to assess the applicability of the Brunskill and Holt (1967) data to the ANL-W actinide-only areas. Sections 6.2.2.1 and 6.2.2.2 provide summaries of the available information on the processes and workrooms at those two sites.

6.2.2.1 Description of Windscale Works Site

The areas of operation for the Brunskill and Holt (1967) study included:

- 1. A plant for the recovery of plutonium from waste material.
- 2. A primary separation plant for the extraction of plutonium from irradiated fuel elements.
- 3. A plutonium finishing plant where pure metal was produced from plutonium nitrate solution.
- 4. A decontamination center.
- 5. An active laundry.
- 6. A facility employing "face standing" or "island" gloveboxes in an open laboratory and used for the manufacture of "plutonium oxide/uranium oxide ceramic fuel" (a.k.a. mixed oxide or MOX fuel) for reactors.

The airborne contamination at Windscale was entirely due to plutonium in one form or another (Brunskill and Holt 1967).

6.2.2.2 Description of Springfields Works Site

The areas of operation for the Brunskill and Holt (1967) study included:

- 1. A plant for the production of uranyl fluoride (UO_2F_2) from gaseous uranium hexafluoride (UF_6) .
- 2. A laboratory for the preparation of samples of uranium ore concentrates for chemical analysis.
- 3. A slag-crushing operation in which magnesium fluoride slag from the reduction of uranium tetrafluoride (UF₄) to metal was crushed in preparation for further treatment.
- 4. The open shop floor, which was used for the annealing and inspection of uranium metal rods.
- 5. The UF4 Reduction Plant where the particular operation considered was the weighing of UF₄-magnesium pellets after delivery from the pelleting press.

The airborne contamination at Springfields was entirely due to uranium and its compounds (Brunskill and Holt 1967).

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6.2.2.3 Applicability of United Kingdom Facilities to ANL-W Actinide-Only Areas

Based on the limited information for the air sampling locations at the Windscale and Springfields Works, the workrooms evaluated in Brunskill and Holt (1967) were full-scale plutonium and uranium production and processing areas. One of the plants at Windscale covered an area of about 300 by 300 ft (90,000 ft² or 8,361.3 m²) (Brunskill and Holt 1967). No other size information was found for the workrooms that were evaluated at the Windscale and Springfields Works. However, their room sizes are likely comparable to large-scale plutonium and uranium processing facilities at DOE sites like Hanford, Feed Materials Production Center, Savannah River Site, and Weldon Spring Plant. Based on the available information, the workrooms at the Windscale and Springfields Works would have had large quantities of dispersible plutonium and uranium present; the ANL-W actinide-only areas did not.

In general, there is insufficient information about the processes and workrooms at the Windscale and Springfields works to assess the applicability of the data in the Brunskill and Holt (1967) study to the actinide-only areas at ANL-W. In addition, there are significant issues with the Brunskill and Holt (1967) study that make all of its BZ:GA ratio information questionable.

A review of Brunskill and Holt (1967) indicates that the terms "static samplers" and "static environmental samplers" are used interchangeably and therefore mean the same thing. The terms "static samplers" and "static environmental samplers" have also been interpreted to represent GA air samplers. The terms "uranium areas" and "uranium-active areas" are also used interchangeably throughout the study, and are assumed to mean the same thing.

Brunskill and Holt (1967) contains contradicting statements regarding the correlation of BZ and GA air sample data. The following is stated in the first two sentences in Section 6.1:

An analysis of results from all uranium and plutonium areas investigated has indicated quite clearly that static environmental samplers are in themselves incapable of indicating the true conditions in the breathing zone of operators. Results from uranium-active areas have generally shown good correlation between the data from static and personal air samplers.

The following is stated in Section 4.1:

In uranium-active areas there is a significant correlation between the personnel and static data at the 99% confidence level. In plutonium-active areas there is virtually no positive correlation.

The following is stated in Section 6.2:

In the plutonium-active areas the general absence of correlation between the personal and static air-sampling data rules out any possibility of control using static sampler data.

The following is stated in Section 6.3:

In the plutonium-active areas, under normal conditions of operation, there is evidence that the nature of the airborne activity in the region of the static samplers is basically similar to that in the breathing zone.

Given the above, Brunskill and Holt (1967) had significant difficulty determining when there was not a good correlation between BZ and GA air sample results. Based on the majority of the statements, the correlation between the BZ and GA air sample results in the uranium areas was good. However, there seems to be confusion when it comes to the plutonium facilities. Based on the statement in
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Section 6.3 of that study, the correlation between the BZ and GA air sample results in the plutonium areas was good under normal conditions of operation; whereas, statements in Section 4.1 and 6.1 of that study indicated there was virtually no correlation.

In 1970, the results for another study that was performed at the Springfields Works were published. The purpose of that study was to investigate why air contamination measured with PAS devices is usually greater than indicated by fixed samplers in the working area. That study concluded that the dust released from protective clothing can contribute appreciably to the air contamination measured by PAS (Butterworth and Donoghue 1970).

In 1988, a review of the experience with plutonium exposure assessment methodologies at the Windscale/Sellafield site was completed (Strong 1988). The Discussion of Results section included the following statements (Strong 1988, pp.124-125):

- *i* there is no obvious correlation between PAS data, urine sample data, and faecal sample data
- *ii* biological sample results generally imply intakes smaller than indicated by personal air sampler...
- a the personal air sampler behaves as a statistical sampling device when operated in an environment having only a few to a few tens of particles per m^3 ,
- b trivial levels of surface contamination, once transported to the air sampler, can appear to indicate significant inhalation...

However, as is evident from the data presented, there is no clear relationship between significant PAS results and the evidence from biological sampling. Indeed this is a source of confusion.

These statements make it clear that through at least some of 1988 the Safety and Medical Services staff at Windscale/Sellafield were still unaware of the of the potentially significant sources of error with personal air sampling and were thoroughly confused as to why their bioassay data generally yielded intake estimates lower than the PAS data. The statements quoted above indicate that nonrespirable dominant particles were likely biasing the Windscale/Sellafield PAS results high, which would also cause the intakes based on those results to be overestimated. This is supported by the statement that the biological sample results generally imply intakes smaller than indicated by the PAS results.

Based on the information in Butterworth and Donoghue (1970) and Strong (1988), the BZ:GA air concentration ratios in Brunskill and Holt (1967) are likely biased high because of sampling resuspended from clothing and the presence of nonrespirable dominant particles on the samples. Therefore, the ORAU Team does not consider the Brunskill and Holt (1967) study to be a credible source of BZ:GA ratio information for any site.

6.2.3 Summary of Issues with Studies Referenced by SCA-TR-2016-SEC009

The determinations made from these two 1967 studies rest heavily on BZ air sample data that was collected using lapel air samplers (Brunskill and Holt 1967; Caldwell, Potter, and Schnell 1967). Both of these studies were completed before some of the potentially significant sources of error with lapel sampling were known.

In the years after those two studies were completed, subsequent air sampling studies indicated that a major cause of the discrepancies between GA or BZ air samples from SASs and BZ air samples from

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lapel air samplers are potentially due to the lapel air samplers sampling the contamination resuspended from a worker's protective clothing (Butterworth and Donoghue 1970; Cohen et al. 1982; Martinelli et al. 1983; Cohen, Harley, and Lippmann 1984; Bohne and Cohen 1985). In 1967, when that was unknown, steps were not likely taken during those two studies to prevent or minimize the sampling of contamination from the workers' clothing by the lapel air samplers.

In addition, subsequent air sampling studies indicated that the presence of larger nonrespirable size particles (i.e., >10 µm) and/or dominant radioactive particles on the filter media for a BZ air sample are also likely causes for the discrepancies between GA and BZ air samples. Because larger nonrespirable size particles do not travel as far as smaller size particles, BZ air concentrations can be higher than GA air concentrations due to the presence of those larger particles only being airborne within the BZ. The presence of nonrespirable radioactive particles in an air sample can result in the overestimate of a worker's intake, because only the respirable fraction of the airborne radioactivity needs to be accounted for and used to estimate internal exposures. When only smaller respirable particles are in the air, the discrepancies between GA and personal BZ air sample results tend to be smaller because the respirable particles are more readily spread throughout the room (Alvarez, Bennett, and Davidson 1994). Further, much of the disparity between air sample data and bioassay data is likely due to the nonrespirable fraction of airborne radioactivity in the air samples (Fischoff 1963; Alvarez, Bennett, and Davidson 1994). When the nonrespirable fraction of airborne radioactivity is significant, air sample data should overestimate a worker's intake. When that is the case, intakes based on air sample data often do not correlate well with bioassay data (Jones et al. 1983; NRC 1984). However, when the air samplers can discriminate against the nonrespirable particles, the air sample data and bioassay data correlate much better (Fischoff 1963).

The presence of dominant radioactive particles on the filter media for an air sample is also likely a cause of significant temporal and spatial variations between GA and BZ air concentrations. Even though this potential issue with BZ air samples was known before 1967, neither of the studies referenced in SCA-TR-2016-SEC009 appear to have considered this as a potential cause for the discrepancies between their GA and BZ air sample results. A 1967 paper on the development of air sampling technology by the United Kingdom Atomic Energy Research Establishment (UKAERE) stated that, in more than half of the air samples from PASs that were examined, much of the activity could be attributed to one or a few active particles (Lister 1967). A 1983 study at the UKAERE facilities indicated that about half of the significant air sample results from their PAS devices were attributable to a single hot particle on the filter media. In the cases that study investigated, there was little to no correlation between the elevated sample results and early fecal excretion data (Jones et al. 1983).

Because of the reasons presented above, the ORAU Team does not recommend using the information in Brunskill and Holt (1967) and Caldwell, Potter, and Schnell (1967) for the actinide-only areas at ANL-W or any other site.

6.3 STUDIES EVALUATED BY ORAU TEAM

The ORAU Team has evaluated five air sampling studies that it considers to be more applicable to the actinide-only areas at ANL-W than the studies cited in SCA-TR-2016-SEC009. Those studies were much more recent than the ones cited in SCA-TR-2016-SEC009. The results of the ORAU Team evaluation have been documented in ORAUT-RPRT-0097 (ORAUT 2021d). The studies evaluated for ORAUT-RPRT-0097 were limited to the air sampling studies that were completed in small workrooms or the mockups of small workrooms, which is more consistent with the sizes of the actinide-only workrooms at ANL-W.

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6.4 RECOMMENDED RESOLUTION TO THE LACK OF PARITY ISSUE

To address the potential lack of parity issue between BZ and GA air sampling results, the ORAU Team recommends using the recommendations and BZ:GA ratio information in ORAUT-RPRT-0097 (ORAUT 2021d).

ORAUT-RPRT-0097 recommends adjusting the GA air concentrations when the median of the BZ:GA ratio distribution becomes significantly greater than 1 or the GSD becomes large. Based on the evaluations in ORAUT-RPRT-0097, the GA air concentrations in most small workrooms should be adjusted to make them equivalent to BZ air concentrations (ORAUT 2021d).

6.4.1 Justifying Use of ORAUT-RPRT-0097 for ANL-W Actinide-Only Areas

As indicated in ORAUT-RPRT-0097, the use of the BZ:GA ratio information in that report should be justified on a case-by case basis (ORAUT 2021d). The following discussion is the justification for using the ORAUT-RPRT-0097 BZ:GA ratio information for the actinide-only areas at ANL-W.

As indicated in Section 11.0 of ORAUT-RPRT-0097, there are five key parameters that should be considered when justifying the application of BZ:GA ratios from other locations to the location of interest. Those parameters include (1) room size, (2) particle size distribution for the airborne radioactivity, (3) ventilation rate for the room, (4) room complexity, and (5) the presence of dominant particles (ORAUT 2021d). The following list addresses each of those five key parameters and provides the reasons why the ORAU Team considers the BZ:GA ratio information in ORAUT-RPRT-0097 to be applicable to the actinide-only areas at ANL-W:

- 1. <u>Room size</u>. The room sizes in the ORAUT-RPRT-0097 studies ranged from 24.0 to 105.0 m² (258 to 1,130 ft²), making them more comparable to the room sizes for the actinide-only areas at ANL-W (ORAUT 2019, ORAUT 2021d). As indicated in Table 6-1, the room sizes for the actinide-only areas at ANL-W were generally 92.3 m² (994 ft²) or less. The ZPPR Workroom was the only confirmed exception to that at 154.4 m² (1,662 ft²), but airborne releases at ZPPR were relatively rare. Because the room size information for the Low Bay Area at the ITF is unknown, the Low Bay Area might be another exception to that. Given that the ITF building had a total square footage of 320 m² (3,455 ft²), and given that the Low Bay Area likely occupied less than half of the ITF, the size of the Low Bay Area was likely less than 160 m² (1,727 ft²). In addition, only six air samples from the ITF Low Bay Area were used for unmonitored uranium intake calculations and all six had gross alpha radioactivity results that were <10% of the MPC (ORAUT 2018b). As indicated in Section 6.1.2, BZ:GA ratios tend to be closer to unity when the air concentrations are low, even for larger workrooms. Therefore, the ORAUT-RPRT-0097 BZ:GA ratio information is still applicable to the larger workroom at ZPPR and the potentially larger workroom at the ITF.</p>
- 2. <u>Particle size distribution</u>. The particle size distributions were known for all five of the ORAUT-RPRT-0097 studies, and the majority of the particles were in the respirable size range (ORAUT 2021d). Therefore, the BZ air sample results from those studies are not significantly biased high by nonrespirable particles. There was no indication of any particle sizing measurements being performed on the airborne particles in the ANL-W actinide-only areas, and there was no indication that the ANL-W air samplers were capable of discriminating against nonrespirable particles. Therefore, the ANL-W air sample results are likely biased high to some unknown degree because those air samples likely contained some nonrespirable radioactive particles. However, the presence of any nonrespirable particles on the ANL-W air samples would result in overestimates of the unmonitored actinide intakes, which is favorable to claimants.

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- 3. <u>Ventilation rate</u>. ORAUT-RPRT-0097 considers the BZ:GA ratio information in it to be applicable to rooms with all but the most extreme air change rates. The workrooms in the studies evaluated for ORAUT-RPRT-0097 had air change rates that ranged from 6 AC/hr to 90 AC/hr (ORAUT 2021d). Even though the air change rates for the actinide-only areas at ANL-W were not found, it is doubtful that any of the air change rates in those areas were significantly greater than 90 AC/hr.
- 4. <u>Room complexity</u>. With the exception of the Charuau (1987) study evaluated in ORAUT-RPRT-0097 (ORAUT 2021d), the complexity of the rooms and ventilation flow patterns evaluated in these studies were comparable to the actinide-only areas at ANL-W. Attachment B provides a compilation of the available floorplans for the actinide-only areas at ANL-W, some of which depict equipment and process locations for evaluating a room's complexity. Even though the room evaluated in the Charuau (1987) study had much more complex ventilation flow patterns than the ones at ANL-W, the use of the Charuau (1987) study data would likely result in a reasonable overadjustment of the ANL-W air sample data.
- 5. <u>Dominant particles</u>. As indicated in ORAUT-RPRT-0097, dominant particles were not present in the workrooms associated with the evaluated studies. For ORAUT-RPRT-0097, studies were excluded from being evaluated if there was any indication of the presence of dominant particles (ORAUT 2021d). Additionally, after several trips to capture data at the site, no records were identified that would have indicated that airborne dominant particles were present in the actinide-only areas at ANL-W. Therefore, the BZ:GA ratio information and air sample data being used for the unmonitored actinide intakes in this report was unaffected by dominant particles.

6.4.2 Applicable Scenarios from ORAUT-RPRT-0097

ORAUT-RPRT-0097 identified four common air sampling scenarios for small workrooms and provided BZ:GA ratio distributions that should be applied to the GA air concentrations to make them equivalent to BZ air concentrations and to account for the increased uncertainty in those air concentrations. The following are the two primary scenarios that were evaluated in ORAUT-RPRT-0097 (ORAUT 2021d):

- <u>Scenario 1</u>. Assumes the worker (BZ location) was always located at the same *X*, *Y* coordinates in the room as the release point (i.e., colocated worker and release point).
- <u>Scenario 2</u>. Assumes the worker (BZ location) was not necessarily located at the same *X*, *Y* coordinates in the room as the release point (i.e., variable worker location relative to the release point).

As indicated in ORAUT-RPRT-0097, Scenario 1 represents what is typically the worst-case scenario because it normally yields the highest BZ:GA ratios. In most instances, this scenario only applies to acute exposures because workers often move around and are not always located at the release point. Scenario 2 represents radiological processing areas that have multiple workstations and multiple workers moving around in each room rather than one worker being at a single stationary location for an entire work shift. For Scenario 2, it was assumed that the potential BZ locations had the same probability of being anywhere in the room (ORAUT 2021d).

Because the generated data for Scenario 1 also indicated there was a significant difference between the BZ:GA ratios when the workroom is generally open in the middle of the room rather than when the workroom has significant obstructions in the middle of the room, the Scenario 1 BZ:GA ratio distributions were divided into two subgroups and reevaluated. The first subgroup represented the scenario when the workroom is generally open in the middle of the room, and the second represented the scenario when the workroom has significant obstructions in the middle of the room. For

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Scenario 2, the generated data indicated that significant obstructions in the middle of the room had little effect on the BZ:GA ratio distribution. Therefore, no subgroups were evaluated for Scenario 2 for ORAUT-RPRT-0097 (ORAUT 2021d).

The BZ:GA ratio distributions associated with those scenarios are presented in Table 6-5. The information in Table 6-5 comes from Table 10-1 in ORAUT-RPRT-0097 (ORAUT 2021d).

Scenario	BZ:GA ratio GM	BZ:GA ratio GSD
Scenario 1	2.95	5.03
Scenario 1 – open in the middle	1.39	3.27
Scenario 1 – obstructions in the middle	12.0	4.11
Scenario 2	1.08	4.02

Table 6-5. Summary of the combined BZ:GA ratio distributions (ORAUT 2021d).

Section 10 of ORAUT-RPRT-0097 also provides the following guidance (ORAUT 2021d):

For acute exposure scenarios, the decision to use the results from Scenario 1 or Scenario 2 will be dependent on whether or not the worker was present at the release locations. Because it is unlikely that a worker would be located at the release location for every release event, the results from Scenario 1 would normally just be applicable to acute exposure scenarios, such as when the worker was at the release location during a radiological accident or excursion. Therefore, the results from Scenario 2 would usually be the more appropriate BZ:GA ratio distribution to use for assessing chronic exposure scenarios. However, in the rare instances when a chronic exposure scenario 1 should be used.

For the unmonitored intakes in the actinide-only areas at ANL-W, the BZ:GA ratio distribution associated with Scenario 2 was determined to be the most appropriate BZ:GA ratio distribution for the following reasons:

- 1. The unmonitored actinide intakes will be assessed as chronic exposures;
- 2. Most of the actinide-only workrooms included multiple workstations; and
- 3. Based on the work being performed in the actinide-only areas, it is unlikely that workers would have spent the majority of their workdays at a single workstation or location. This is especially true given the intermittent nature of most of the work in those areas. In most situations, those workers would have moved between multiple workstations during their workdays, including moving in and out of those workrooms.

Therefore, a BZ:GA ratio distribution with a GM of 1.08 and a GSD of 4.02 will be applied to the air concentrations being used to calculate the potential unmonitored actinide intakes in this report. The application of that BZ:GA ratio distribution should make the ANL-W actinide-only area air concentrations equivalent to BZ air concentrations and should adequately account for the increased uncertainty in those air concentrations.

7.0 UNMONITORED ACTINIDE INTAKES

This section describes the approaches used to calculate the unmonitored actinide intakes for ANL-W, summarizes the air concentrations used for those intake calculations, summarizes the resulting intake values, and describes how some of the application issues will be addressed.

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7.1 GENERAL APPROACHES USED FOR INTAKE CALCULATIONS

As indicated in ORAUT-RPRT-0097, the GA air concentrations in small workrooms should be adjusted using a BZ:GA ratio distribution to make them equivalent to the BZ air concentrations (ORAUT 2021d). Because the air samples at ANL-W were predominately collected using GA air samplers, the bounding and reported air concentrations needed to be converted into equivalent BZ air concentrations for the intake calculations.

The following assumptions and parameters were used for the intake calculations:

- Daily exposure time was 8 hours per workday;
- 5 days worked per week;
- 50 weeks worked per year;
- 250 workdays per year (D_{WrkYr}), based on prior assumptions; and
- 365 days per year ($D_{Ca|Yr}$) represents the average number of calendar days per year.

The 8 hr/d, 5 d/wk, 50 wk/yr is equivalent to a 2,000-hr exposure year. Even though some ANL-W workers might have worked more than 8 hours per workday, it is unlikely that they were in the actinide-only areas for more than 8 hours per workday, due to breaks and the intermittent nature of the work in the actinide-only workrooms. Table 6-1 lists the workrooms where actinide-only exposures were possible at ANL-W, and all of those are limited to specific workrooms in specific buildings. They also represent a very small fraction of the ANL-W site. Because of that, any time that a worker spent in the hallways or other rooms would be time that a worker was not in an actinide-only workroom. Therefore, on average, the potential for unmonitored actinide exposures did not likely exceed 8 hours per workday.

Based on the available information, process operations in the actinide-only areas only took place during the Monday through Friday work week. Activities that did not involve process operations did take place during some weekends, but those activities were even more intermittent than the process operations. Therefore, the 5 days worked per week assumption is reasonable for all types of work that might have taken place in the actinide-only areas.

The 50 weeks worked per year assumes that the ANL-W workers only took 2 weeks of vacation each year. Because this assumption does not factor in the observed holidays, it is favorable to claimants. For the intake periods addressed in this report, the number of observed holidays occurring during the workweek typically ranged from 8 to 10 days for a given calendar year. Accounting for the observed holidays that occurred during a weekday would reduce the weeks worked assumption by up to 2 weeks for a total of 48 weeks worked per year.

On average, there are actually 365.2465 days per calendar year, and 365 days represents a rounded value. For how it is used in the equations below, rounding that value to 365 days is favorable to claimants.

The following sections show how the unmonitored actinide intakes were calculated. When a specific value was used for one of the equation variables, that value was provided in parentheses at the end of the variable definition. For example in Equation 7-5, T_{DayExp} is defined as the "daily exposure period (8 hr/day)". The "(8 hr/day)" part of that definition indicates that 8 hr/day was always used for T_{DayExp} when using Equation 7-5.

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7.1.1 Intakes Based on Bounding Air Concentrations

The bounding air concentrations in the SEC-00224 evaluation report were only expressed in terms of percent of the applicable air concentration limits (NIOSH 2016). For the intake calculations, the following formula was used to convert those bounding air concentrations into maximizing GA air concentrations:

$$C_{\text{MaxGA}} = \frac{(CL_{\text{GA}})(P_{\text{CL}})}{CF}$$
(7-1)

where

C_{MaxGA}	=	maximizing GA air concentration (pCi/m ³)
CL _{GA}	=	the applicable air concentration limit value used during the applicable period, as
		measured by a GA sampler (dpm/m ³)
P_{CL}	=	percent of the <i>CL</i> maintained by the site during the applicable period (1-100%)
CF	=	conversion factor (2.22 dpm/pCi × 100 percent/fraction = 222)

Because the air concentrations being used for these calculations represent upper-bounds or maximizing air concentrations, the resulting maximizing GA air concentrations were treated as constants. To convert the maximizing GA air concentrations to maximizing BZ air concentrations, Equations 11-5 and 11-6 in ORAUT-RPRT-0097 were used in conjunction with the BZ:GA ratio distribution for Scenario 2 in ORAUT-RPRT-0097 (ORAUT 2021d):

$$C_{\rm BZ} = C_{\rm MaxBZ} = (C_{\rm MaxGA})({\rm BZ}:{\rm GA})$$
(7-2)

$$GSD_{BZ} = GSD_{MaxBZ} = GSD_{BZ:GA}$$
(7-3)

where

C _{BZ}	=	GM of the BZ air concentration distribution (pCi/m ³)
C _{MaxBZ}	=	GM of maximizing BZ air concentration distribution (pCi/m ³)
C _{MaxGA}	=	maximizing GA air concentration (pCi/m ³)
BZ:GA	=	GM of the applicable BZ:GA ratio distribution (1.08)
GSD _{BZ}	=	GSD of the BZ air concentration distribution
GSD _{MaxBZ}	=	GSD of the maximizing BZ air concentration distribution
GSD _{BZ:GA}	=	GSD of the applicable BZ:GA ratio distribution (4.02)

Equations 7-4 and 7-5 were then used for the intake calculations. Because air sampling data can only be used to estimate potential intakes due to inhalation, potential intakes due to ingestion were estimated based on the recommendations in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004). Equation 7-5 was derived from the information in OCAS-TIB-009. Because the Mode 2 ingestion intake factor in OCAS-TIB-009 was based on an assumed daily exposure period of 8 hours (i.e., a typical workday), the Mode 2 equation was modified to accommodate different exposure

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periods. Equations 7-5 and 7-6 also contain an adjustment to convert from an intake per workday to an intake per calendar day:

$$I_{\text{Inhal}} = (C_{\text{BZ}})(BR)(T_{\text{DayExp}})\left(\frac{D_{\text{WrkYr}}}{D_{\text{CalYr}}}\right)$$
(7-4)

where

 I_{Inhal} = daily inhalation intake (pCi/calendar day) C_{BZ} = GM of the BZ air concentration distribution (pCi/m³) BR = breathing rate (1.2 m³/hr) T_{DayExp} = daily exposure period (8 hr/d)

D_{WrkYr} D_{CalYr} = days per work year (250 d/yr) days per solandar year (265 d/yr)

= days per calendar year (365 d/yr)

$$I_{\text{lngest}} = (C_{\text{BZ}}) \left[(M2_{\text{lngest}}) \left(\frac{T_{\text{DayExp}}}{8 \,\text{hr}} \right) + M3_{\text{lngest}} \right] \left(\frac{D_{\text{WrkYr}}}{D_{\text{CalYr}}} \right)$$
(7-5)

where

I Ingest	=	daily ingestion intake (pCi/calendar day)
C _{BZ}	=	GM of the BZ air concentration distribution (pCi/m ³)
T_{DayExp}	=	daily exposure period (8 hr/day)
M2 _{Ingest}	=	Mode 2 ingestion intake factor based on an 8-hour workday (0.0985 m ³)
M3 _{Ingest}	=	Mode 3 ingestion intake factor based on a 24-hour day (0.1004 m ³)
DwrkYr	=	days per work year (250 days/yr)
D _{CalYr}	=	days per calendar year (365 days/yr)

7.1.2 Intakes Based on Air Sample Data

As indicated in Section 5.2, the evaluated sets of air sample concentrations in the ANL-W actinideonly areas can be represented by lognormal distributions. To convert the lognormally distributed timeweighted GA air concentrations to BZ air concentrations, Equations 7-6 and 7-7 were used in conjunction with the BZ:GA ratio distribution for Scenario 2 in ORAUT-RPRT-0097 (ORAUT 2021d). Equations 7-6 and 7-7 are based on Equations 11-1 and 11-2 in ORAUT-RPRT-0097 (ORAUT 2021d). Before using Equation 7-6, the time-weighted GMs for the GA air concentration distributions were converted from units of dpm/m³ to pCi/m³.

$$C_{\rm BZ} = \exp\left(\ln(C_{\rm GA}) + \ln({\rm BZ}:{\rm GA})\right)$$
(7-6)

$$GSD_{BZ} = \exp\left(\sqrt{\ln(GSD_{GA})^2 + \ln(GSD_{BZ:GA})^2}\right)$$
(7-7)

where

C _{BZ}	=	GM of the BZ air concentration distribution (pCi/m ³)
C _{GA}	=	time-weighted GM of the GA air concentration distribution (pCi/m ³)
BZ:GA	=	GM of the applicable BZ:GA ratio distribution (1.08)
GSD _{BZ}	=	GSD of the BZ air concentration distribution
GSD GA	=	GSD of the GA air concentration distribution
GSD _{BZ:GA}	=	GSD of the applicable BZ:GA ratio distribution (4.02)

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7.1.2.1 Intakes Based on Adjusted Air Sample Data

When the individual air sample results in the dataset were adjusted to address the air sample dilution concern, Equations 7-4 and 7-5 above were used, respectively, for the inhalation intake (I_{Inhal}) and ingestion intake (I_{Ingest}) calculations.

7.1.2.2 Intakes Based on Unadjusted Air Sample Data

When the individual air sample results in the dataset could not be adjusted to address the air sample dilution concern, the concern was addressed by modifying the intake calculations. Equation 7-8 is the modified inhalation intake calculation that addresses the sample dilution concern when the individual air sample results could not be adjusted. For the inhalation intake calculation, no adjustment was needed in Equation 7-8 to convert the intake into an intake per calendar day because the worker was assumed to be continuously exposed to the applicable air concentration during work and outside of work (i.e., for 24 hr/d and 365 d/yr). Note that this calculation is only equivalent to the approach taken for the adjusted air sample data when the air concentration is assumed to drop to zero during the nonoperational periods and when the daily operational period for the facility is the same as the daily exposure period assumed for the workers (i.e., no second or third work shifts at the facility).

$$I_{\text{Inhal}} = (C_{\text{BZ}})(BR)(T_{\text{DayExp}})$$
(7-8)

where

I _{Inhal}	=	daily inhalation intake (pCi/calendar day)
C _{BZ}	=	GM of the unadjusted BZ air concentration distribution (pCi/m ³)
BR	=	breathing rate (1.2 m ³ /hr)
T_{DayExp}	=	daily exposure period (24 hr/d)

As indicated above, potential intakes due to ingestion need to be estimated based on the recommendations in OCAS-TIB-009 (NIOSH 2004). Because OCAS-TIB-009 is not set up to account for ingestion intakes for a 24 hr/d exposure scenario and might underestimate the ingestion intakes if the BZ air concentration from Equation 7-6 is used, a modified calculation was performed, which included an additional calculation step. By adding a step to calculate <u>an equivalent to the adjusted BZ</u> <u>air concentration</u>, Equation 7-5 can then be used to calculate the ingestion intake. The equivalent to the adjusted BZ air concentration was calculated using Equation 7-9, which yields a BZ air concentration that is equivalent to a BZ air concentration that was adjusted to account for the air sample dilution concern.

$$C_{\text{AdjBZ}} = \frac{I_{\text{Inhal}}}{(BR)(T_{\text{DayExp}})\left(\frac{D_{\text{WrkYr}}}{D_{\text{CalYr}}}\right)}$$
(7-9)

where

C_{AdjBZ}	=	equivalent to the GM for the adjusted BZ air concentration distribution (pCi/m ³)
I Inhal	=	daily inhalation intake (pCi/d)
BR	=	breathing rate (1.2 m ³ /hr)
T _{DayExp}	=	daily exposure period (8 hr/d)
D _{WrkYr}	=	days per work year (250 d/yr)
D_{CalYr}	=	days per calendar year (365 d/yr)

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By using C_{AdjBZ} for C_{BZ} , Equation 7-5 was then used to calculate the ingestion intake. A daily exposure period of 8 hr/d was also used for that calculation.

7.2 SUMMARY OF AIR CONCENTRATIONS FOR INTAKE CALCULATIONS

Tables 7-1 through 7-4 summarize the air concentrations that were used for the unmonitored actinide intake calculations. The air concentrations in those tables are the GA air concentrations before the application of the BZ:GA ratio distribution. In all instances, the bounding air concentrations were based on 10% of the MPC values that ANL-W was using for those areas. The MPC values being used by ANL-W were obtained from the available Air Sample Data sheets for those areas. When 10% of the MPC might not have been bounding, the available air sample data for those areas were compiled and a statistical evaluation was performed. The details and results of that statistical evaluated air sample data could be represented by a lognormal distribution.

Table 7-1. EBR-I Complex uranium area air concentrations.^a

			Air	
	Applicable	Distribution	concentration ^b	
Actinide-only area	period	type	(dpm/m³)	GSD
ZPR-III Workroom	01/01/1958–07/31/1961	Constant	7.0	N/A
ZPR-III Workroom	08/01/1961-06/13/1975	Constant	13.2	N/A

a. N/A = not applicable.

b. When the distribution type is constant (i.e., no distribution), a bounding air concentration was used for the intake calculations. Per the SEC-00224 evaluation report, the bounding air concentrations for the uranium areas at the EBR-I Complex were 10% of the MPC values that ANL-W was using for those areas.

Table 7-2. EBR-II Complex uranium area air concentrations.

			Air	
	Applicable	Distribution	concentration ^a	
Actinide-only area	period	type	(dpm/m³)	GSD
Cold-Line Areas ^b	08/01/1967-12/31/1974	Lognormal	3.37	4.92
Cold-Line Areas ^b	01/01/1975-06/30/1976	Lognormal	2.28	5.20

a. When the distribution type is lognormal, this air concentration is the GM for that air concentration distribution.

b. In the EBR-II Complex, the uranium areas for the Cold-Line fuel work were in the FCF and the ITF buildings.

Table 7-3. EBR-II Complex thorium area air concentrations.

Applicable period	Distribution type	Air concentration ^a (dpm/m ³)	GSD
08/01/1963-11/30/1967	Lognormal	16.9	2.29
	Applicable period 08/01/1963–11/30/1967	Applicable periodDistribution type08/01/1963-11/30/1967Lognormal	Applicable periodDistribution typeAir concentrationa (dpm/m3)08/01/1963-11/30/1967Lognormal16.9

a. When the distribution type is lognormal, this air concentration is the GM for that air concentration distribution.

Table 7-4. EBR-II Complex plutonium area air concentrations.ª

Actinide-only area	Applicable period	Distribution type	Air concentration ^ь (dpm/m³)	GSD
ZPPR	09/01/1970-07/31/1975	Constant	0.44	N/A
FCF	04/01/1970-04/30/1973	Lognormal	1.35	5.73

a. N/A = not applicable.

b. When the distribution type is constant (i.e., no distribution), a bounding air concentration was used for the intake calculations. Per the SEC-00224 evaluation report, the bounding air concentration for the plutonium area at ZPPR was 10% of the MPC value that ANL-W was using for that area. When the distribution type is lognormal, this air concentration is the GM for that that air concentration distribution.

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7.3 CALCULATED UNMONITORED ACTINIDE INTAKES

Using the approaches described in Section 7.1 and the air concentration values and distributions in Section 7.2, the unmonitored actinide intakes were calculated for ANL-W. These intakes were calculated using the Excel workbook named "ANL-W - Unmonitored Actinide Intake Calculations" (ORAUT 2021f). The following sections describe the results.

7.3.1 EBR-I Complex Intakes

For the EBR-I Complex, uranium was the only type of unmonitored actinide exposure that was possible, and the potential unmonitored uranium intakes are presented in Table 7-5.

Applicable period	Inhalation intake (pCi/d)	Ingestion intake (pCi/d)	GSD
01/01/1958–07/31/1961	2.07E+01	4.30E-01	4.02
08/01/1961–06/13/1975	3.91E+01	8.10E-01	4.02

Table 7-5. Unmonitored uranium intakes at the EBR-I Complex.

7.3.2 EBR-II Complex Intakes

At the EBR-II Complex, the potential unmonitored actinide exposures included exposures to uranium, thorium, and plutonium. Tables 7-6 through 7-9 summarize the potential unmonitored actinide intakes at the EBR-II Complex and their applicable periods.

	Inhalation intake	Ingestion intake	
Period	(pCi/d)	(pCi/d)	GSD
08/01/1967–12/31/1974	1.08E+01	2.23E-01	8.29
01/01/1975–06/30/1976	3.19E+01	6.62E-01	8.65

Table 7-6. Unmonitored uranium intakes at the EBR-II Complex.

	Table 7-7.	Unmonitored thorium in	ntakes at the	EBR-II Com	olex.
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	Inhalation intake	Ingestion intake	
Period	(pCi/d)	(pCi/d)	GSD
08/01/1963-11/30/1967	5.41E+01	1.12E+00	5.05

Table 7-8. Unmonitored plutonium intakes for the EBR-II Complex.

	Inhalation intake	Ingestion intake	
Period	(pCi/d)	(pCi/d)	GSD
04/01/1970-04/30/1973	4.32E+00	8.28E-02	9.32

Table 7-9. Unmonitored plutonium intakes for the ZPPR.

	Inhalation	Ingestion		
	intake	intake		
Period	(pCi/d)	(pCi/d)	GSD	
09/01/1970-07/31/1975	1.4E+00	2.9E-02	4.02	

7.4 APPLICATION OF UNMONITORED ACTINIDE INTAKES

The following sections address some application issues that are unique to the unmonitored actinide intakes for ANL-W.

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7.4.1 Overlapping Periods of Exposure

Given that all workers at the EBR-I and EBR-II Complexes were required to wear dosimeters to monitor their external doses, work at each of those complexes can be identified by the location code information in their external dosimetry records. Therefore, for periods when a worker had both EBR-I and EBR-II Complex dosimeters, only the unmonitored intakes for the complex resulting in the highest dose would be assigned for that period.

At the EBR-II Complex, the actinide-only areas were distinctly separate air spaces, which means that a worker could not be simultaneously exposed to thorium, uranium, and plutonium. In addition, unmonitored actinide intakes for ZPPR workers and non-ZPPR workers will be handled separately. Because ZPPR workers can be identified by the available external dosimetry records in the SRDB and because it is unlikely that ZPPR workers would have entered the other actinide-only areas in the EBR-II Complex during the operational periods for the EBR-II fuel production work in those areas, only the unmonitored plutonium intakes specific to ZPPR will be assessed for the ZPPR workers (i.e., the intakes in Table 7-9). Additionally, because of security safeguards for the plutonium, it was unlikely that non-ZPPR workers would have been allowed in the ZPPR Workroom while the plutonium fuel was being handled. Therefore, the ZPPR plutonium intakes in Table 7-9 do not need to be assessed for non-ZPPR workers.

For the non-ZPPR workers in the EBR-II Complex, potential unmonitored actinide intakes will be assessed based on the intake information in Tables 7-6, 7-7, and 7-8. Because the unmonitored actinide intakes are based on gross alpha analysis results and because a non-ZPPR worker in the EBR-II Complex could not be simultaneously exposed to all three actinides, only the unmonitored actinide intake resulting in the highest dose would be assigned for periods when more than one type of actinide intake was possible.

7.4.2 Uncertainties

To account for uncertainties associated with the unmonitored actinide intakes, each unmonitored actinide intake would be assessed as a lognormal distribution with the appropriate GSD value, as indicated above in Tables 7-5 through 7-9.

8.0 CONCLUSIONS

The following sections provide the conclusions for each of the three main purposes of this report.

8.1 ISSUES RAISED IN SCA-TR-2016-SEC009

SCA-TR-2016-SEC009 indicates a concern that there are factors that caused the ANL-W air monitoring data to not be representative of the BZ air concentrations, and it concludes that NIOSH's proposed value of 10% MPC as a bounding value for internal dose assessment lacks credibility. However, SCA-TR-2016-SEC009 does not identify factors specific to or applicable to ANL-W that could cause those air monitoring results to be unrepresentative of the BZ air concentrations (SC&A and Salient 2016).

The dose reconstruction regulation (42 CFR Part 82) and Section 5.2 of OCAS-IG-002 (NIOSH 2002) still allow the use of workplace monitoring data such as BZ air samples, GA air samples, and surface contamination surveys to estimate an individual's internal dose when bioassay data is not adequate. However, Section 5.2 of OCAS-IG-002 requires that consideration be given to any factors that could create a difference between GA and BZ air concentrations when using GA air sampling data (NIOSH 2002). After an extensive data capture effort to find information on the ANL-W air monitoring program and find air sampling results for the actinide-only areas at ANL-W, those air monitoring records have

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been reviewed to identify potential factors that could create a difference between the GA and BZ air concentrations. There is no information that indicates that there might have been a significant difference between the GA and BZ air concentrations in the actinide-only areas at ANL-W. In addition, as indicated in Section 6.1, differences between the GA and BZ air concentrations would have been minimized by the relatively small size of the air spaces and normally low air concentrations associated with the ANL-W actinide-only areas. Therefore, the OCAS-IG-002 requirement to give consideration to those potential factors had been met by the SEC-00224 evaluation report (NIOSH 2016).

However, NIOSH has since decided to investigate the lack of parity issue further and tasked the ORAU Team to evaluate that issue for small workrooms. The results of that evaluation have been documented in ORAUT-RPRT-0097 (ORAUT 2021d). The studies evaluated for ORAUT-RPRT-0097 were limited to the air sampling studies that were completed in small workrooms or the mockups of small workrooms, which is more consistent with the sizes of the actinide-only workrooms at ANL-W. ORAUT-RPRT-0097 recommends adjusting the GA air concentrations when the median of the BZ:GA ratio distribution becomes significantly greater than 1 or the GSD becomes large. Based on the evaluations in ORAUT-RPRT-0097, the GA air concentrations in most small workrooms should be adjusted to make them equivalent to BZ air concentrations.

During the process of making the adjustments to correct for sample dilution, the ORAU Team determined that an air concentration equivalent to 10% MPC would not be bounding for all actinides and periods because of a few elevated air sample results for those periods. To address this, the time-weighted mean of the adjusted air concentrations will be used to bound the unmonitored internal actinide doses for those actinides and periods (for more details see Attachment A). As indicated in Section 5.2, it is still reasonable to bound the potential unmonitored uranium intakes at the EBR-I Complex and unmonitored plutonium intakes at ZPPR based on a concentration of 10% of the MPC.

The following are a few of the key factors that would ensure that the approaches described above and in Attachment A would not underestimate the unmonitored thorium, uranium, and plutonium intakes at ANL-W:

- A significant number of the elevated air sampling results were biased high because ANL-W did not allow enough time for all of the short-lived radon and thoron progeny to decay before performing the final counts on those air samples. As indicated in Section 4.0, some of the reported air concentrations could be as much a 171 times higher than what they actually were due to the final counts being performed on the air samples before all of the short-lived radon and thoron progeny could decay.
- 2. The actual occupancy in the actinide-only areas and intermittent nature of the operations in those areas are not accounted for. In general, radiological workers do not spend 100% of their workday in radiological areas for several reasons (e.g., breaks, lunches, meetings, training). At ANL-W, another factor that affects the occupancy is the intermittent nature of much of the work. Because the Cold-Line HEU fuel production work and RAS-TREAT sodium-loop experiment work with plutonium source terms were very intermittent, potential operational exposures to those source terms were also very intermittent. In addition, occupancy in the uranium areas (due to HEU) and plutonium areas was likely minimal during the nonoperational portions of the workdays due to accountability and security reasons associated with the protection of SNM. During all nonoperational periods, all of the HEU and plutonium source terms were probably locked up.
- 3. The respirable fraction of the airborne radioactivity was not accounted for. The unmonitored thorium intakes would be the most affected by this factor because all of the thoria was

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assumed to be respirable even though the majority of the thoria powder being used was likely nonrespirable.

- 4. A review of the radiological work permits for ANL-W indicated that the site often required respiratory protection when there was any potential for internal exposure. Not accounting for when a worker wore respirator protection would result in an overestimate of their actinide intakes by a factor of 10 or more.
- 5. The air sample results that were adjusted to account for potential sample dilution were adjusted based on the assumption that the concentrations of airborne radioactivity dropped to zero during the nonoperational periods. Because the concentrations of airborne radioactivity did not likely drop all of the way to zero outside of the normal work shift hours, those adjustments likely resulted in an overestimate of the air concentrations for the operational periods.

8.2 ADDITIONAL DETAILS ABOUT UNMONITORED ACTINIDE INTAKE CALCULATIONS

Section 7.0 provides the additional details about the unmonitored actinide intakes calculations that the SEC-00224 evaluation report did not go into. The resulting unmonitored actinide intakes are provided in Section 7.3. Section 7.4 provides some additional guidance on how the unmonitored actinide intakes in takes in Section 7.3 will be applied for ANL-W claims.

8.3 DEVIATIONS FROM SEC-00224 EVALUATION REPORT

As indicated in Section 1.0, one of the purposes of this report was to provide the basis for revising some of the unmonitored actinide intake and internal dose approaches proposed in the SEC-00224 evaluation report. This section provides a summary of those changes.

Rather than using the recommendation in the SEC-00224 evaluation report to bound all of the unmonitored intakes by using 10% of the MPC, the actual air concentration distribution information was used for four of the air sample datasets. As indicated in Sections 4.2 and 5.2, four of the air sample datasets were put through a formal statistical evaluation, because the numbers of elevated air sample results indicated that 10% of the MPC may not be bounding. The four air sample datasets included the air sample results for the EBR-II Complex thorium and uranium areas and the FCF plutonium areas, with the air sample results for the uranium areas being divided into two separate datasets.

The potential unmonitored actinide intakes for ZPPR and non-ZPPR workers in the EBR-II Complex are now being handled separately. Previously, it could not be determined which facility a worker in the EBR-II Complex worked at. Because of that, it had to be assumed that all EBR-II Complex could have received an unmonitored actinide intake in any of the actinide-only areas in the EBR-II Complex. As discussed in Section 7.4.1, ZPPR workers can now be identified using the available external dosimetry records in the SRDB. When the SEC-00224 evaluation report was issued, the external dosimetry records for some of years had not been captured yet.

As indicated in Section 4.2.2, the need to assess unmonitored uranium intakes at ZPPR has been eliminated. Only unmonitored plutonium intakes need to be assessed for ZPPR workers.

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The remaining deviations were either revisions to the unmonitored actinide intake date ranges in the SEC-00224 evaluation report or determined start and/or end dates when not provided in that evaluation report. The following is a summary of those revised dates.

- 1. For the unmonitored uranium intakes at the EBR-I Complex, June 13, 1975 was determined to be an appropriate end date for those intakes, as discussed in Section 4.1.
- 2. For the unmonitored uranium intakes for the Cold-Line areas at the EBR-II Complex, the intake end date was reduced from June 1983 to June 30, 1976, as discussed in Section 4.2.2.
- 3. For the unmonitored plutonium intakes at ZPPR, no start or end dates were provided in the SEC-00224 evaluation report for these potential intakes. As discussed in Section 4.2.3.1, the appropriate intake start and end dates were determined to be September 1, 1970, through July 31,1975.
- 4. For the unmonitored plutonium intakes at the FCF, the intake end date was extended from December 1972 to April 30, 1973, as discussed in Section 4.2.3.2.

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ATTACHMENT A STATISTICAL EVALUATION OF EBR-II COMPLEX AIR SAMPLE DATA

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ATTACHMENT A STATISTICAL EVALUATION OF EBR-II COMPLEX AIR SAMPLE DATA (continued)

A.1 PURPOSE

This attachment provides details on the statistical evaluations of several of the gross alpha radioactivity in air datasets for the ANL-W actinide-only areas. Those evaluations were only performed on the ANL-W air monitoring datasets for the actinide-only areas with the highest potential for internal exposures.

A.2 APPROACH USED

The regression on order statistic (ROS) is a method that can be used to calculate the GM and GSD of a lognormal distribution fit to a dataset (see ORAUT 2006 and ORAUT 2014b for technical details). ROS can handle datasets that have singly or multiply left-censored data and provides a graphic output that can be used to judge if a lognormal distribution indeed fits the dataset. In ROS the logs of the empirical quantiles (the observed air monitoring results) are plotted against the theoretical quantiles from a standard normal distribution (the *z*-scores or standard deviations). If the points fall approximately along a regression line, the data are taken to be lognormally distributed with the log of the GM given by the *y*-intercept of the line and the log of the GSD given by its slope.

The R code was used to perform the ROS calculations for this report. R is a language and environment for statistical computing and graphics. R is described as an integrated suite of software facilities for data manipulation, calculation, and graphical display. It is capable of performing a wide variety of statistical (linear and nonlinear modeling, classical statistical tests, time-series analysis, classification, clustering, and others) and graphical techniques, and is highly extensible.

ROS was performed on the ANL-W gross alpha radioactivity air monitoring datasets for the actinideonly areas with the highest potential for internal exposures. The air concentration data for those calculations was obtained from three spreadsheets (ORAUT 2018a, 2018b, and 2018c). Because the natural logarithm of zero cannot be taken, the ORAU Team evaluated results of 0 dpm/m³ with a single censoring level of <0.01 dpm/m³. Note that a portion of the reported air sample results also included several other censoring levels imposed by ANL-W (i.e., <1% of MPC, <10% of MPC, etc.). Except for the May 1975 through June 1976 uranium data (which had a better fit using an unweighted regression), a weighted linear regression was used to fit the data where the sampling time was the weight. Each plot lists the GM in units of dpm/m³, the GSD, the total number of results used in the regression, and the number of uncensored results used in the regression. Figures A-1 through A-4 contain quantile-quantile plots of those datasets along with the resulting GM and GSD values. The files associated with the ROS, which contain the details for those calculations, can be found in ORAUT (2021g).

Based on the plots in Figures A-1 through A-4, a lognormal distribution provides a reasonable fit for the four datasets. The FCF Cold-Line air data from August 1967 through March 1973, depicted in Figure A-2, has the worst fit for a lognormal distribution between the four datasets. The nonlinear shape depicted by the air concentration data is due to the much larger quantity of data in this dataset that was left-censored by ANL-W (e.g., only reported as <1% of MPC, <10% of MPC, etc.), and the fact that some of the censoring levels were greater than many of the other reported results. As a result, the distribution of that dataset has a bimodal component. However, a lognormal distribution is still considered to be a reasonable fit for the FCF Cold-Line air data from August 1967 through March 1973.

ATTACHMENT A STATISTICAL EVALUATION OF EBR-II COMPLEX AIR SAMPLE DATA (continued)



Figure A-1. FCF Room 25 (thoria room) air data, August 1963 to November 1967 (ORAUT 2021g).



Figure A-2. FCF Cold-Line air data, August 1967 to March 1973 (ORAUT 2021g).

ATTACHMENT A STATISTICAL EVALUATION OF EBR-II COMPLEX AIR SAMPLE DATA (continued)







Figure A-4. FCF RAS-TREAT sodium-loop air data, April 1970 to April 1973 (ORAUT 2021g).

ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS

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B-10	ZPR-III workroom floorplan with added dimensions71
B-11	ZPPR workroom and vault floorplan with added dimensions

B.1 PURPOSE

This attachment contains several figures associated with the ANL-W actinide-only areas. The purpose of this attachment is to consolidate these figures into one location, to provide a basis for some of the information in the main body of this report, and to facilitate review of this report.

B.2 SUMMARY OF FIGURES

Complete as-built room dimensions for the ANL-W actinide-only areas could not be found. However, a floorplan of the FCF from sometime between 1962 and 1964, which was potentially before construction of the FCF was completed, contained some dimensional information and is provided as Figure B-1 below (ANL-W 1962–1964). Figure B-2 is a floorplan of the FCF from before August 1967 that appears to be drawn to scale and also provides the room numbers. In August 1967, FCF Rooms 19 and 20 were combined into a single room that was identified as just Room 20. In addition, a number of dimensions were added to Figure B-2. The added dimensions were based on a combination of the dimensional values in Figure B-1 and dimensions that were scaled from Figure B-2. Using the dimensional information in Figure B-1 and scaling the room dimensions from objects in Figure B-2, reasonable estimates of the room sizes could be made. Figure B-3 is a late Cold-Line production era floorplan of the FCF that also provides the room numbers. Figure B-4 is a ventilation flow diagram for the FCF during the Cold-Line production era, which depicts where the ventilation air inlets and outlets were located.

Figures B-5 through B-8 depict specific FCF rooms and the equipment locations in those rooms. These figures were obtained from the various survey records for the FCF and might not be drawn to scale.

Figure B-9 is the only known depiction of the layout of the ITF. The figure was obtained from some 1976 survey records and might not be drawn to scale. Figure B-10 is a floorplan for ZPR-III. It also contains added dimensions for the Workroom, which was the actinide-only area at the EBR-I Complex. Figure B-11 is the floorplan for the Workroom and Vault at ZPPR, which also contains added dimensions. The actinide-only area at ZPPR was the Workroom.

The room sizes for the actinide-only areas in Figures B-1 through B-11 have been calculated based on the best available information and are documented in ORAUT (2019). Because no scale floorplan or room dimension information could be found for the ITF, no room sizes were calculated for the ITF.



Figure B-1. Early FCF floorplan with dimensions (ANL-W 1962–1964).



ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)

Figure B-2. FCF floorplan with room numbers and added dimensions, before August 1967 (ORAUT 2019).



Figure B-3. FCF late Cold-Line era floorplan (Forrester et al. 1989).

ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)



Figure B-4. FCF ventilation flow diagram during Cold-Line era (ERDA 1977).

ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)



Figure B-5. FCF Room 25 (thoria room) (ANL-W 1963).



Figure B-6. FCF Room 22 (sodium-loop glovebox room) (ANL-W 1974).

ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)



Figure B-7. FCF Room 20 (Cold-Line room) (ANL-W 1976).



Figure B-8. FCF Room 26 (Cold-Line room) (ANL-W 1976).



ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)

Figure B-9. ITF (ANL-W 1976).

Figure B-10. ZPR-III workroom floorplan with added dimensions (ORAUT 2019).



ATTACHMENT B FIGURES OF THE ANL-W ACTINIDE-ONLY AREAS (continued)

Figure B-11. ZPPR workroom and vault floorplan with added dimensions (ORAUT 2019).