

# ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities | Dade Moeller | MJW Technical Services

Page 1 of 188

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Total Rewrite

Revision

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#### **PUBLICATION RECORD**

EFFECTIVE	REVISION	
DATE	NUMBER	DESCRIPTION
05/28/2004	00	New Technical Basis Document for the Fernald Environmental Management Project (FEMP) – Occupational Internal Dose. Initiated by Bryce L. Rich. First approved issue. Incorporates comments from OCAS and Task 5 reviews. Initiated by Bryce L. Rich.
07/22/2016	01	Revised in response to Task 5 dose reconstruction needs for clarification. Added Purpose, Scope, and Special Exposure Cohort sections to Section 1.0. Revised to incorporate responses to comments and responses in various white papers that were consolidated into ORAUT-RPRT-0052. Incorporated changes in response to formal internal and NIOSH review comments and Sanford Cohen & Associates matrix issues. Revised to reflect changes resulting from Special Exposure Cohort designations. Revised recycled uranium mixture and default uranium enrichment assumptions. Added <sup>241</sup> Am and other radionuclides as recycled uranium contaminants. Added instructions for interpretation of in vivo data including methods for calculating thorium, thoron, unsupported radium, and raffinate exposures, including <sup>226</sup> Ra from radon breath analysis results. Incorporated supporting documents pertaining to rationale for 400 ppb U Pu, uranium (ORAUT-OTIB-0078) and thorium coworker intakes, assignment of thorium dose based on 10% of the derived air concentrations, thoron exposures, and effective derived air concentrations. Updated and added references, as appropriate. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Karen S. Kent.

Revision No. 01 Effective Date: 07/22/2016

Page 3 of 188

# TABLE OF CONTENTS

# **SECTION**

# <u>TITLE</u>

PAGE
------

Acrony	/ms and	l Abbrevia	tions		9
5.1	Introdu 5.1.1 5.1.2 5.1.3	iction Purpose . Scope Special E	xposure Co	hort	12 13 13 13
5.2	Backg	round			14
5.3	Radior 5.3.1 5.3.2 5.3.3 5.3.4	Q-11, K-6 5.3.4.2	Concern Uranium En Chemical Fo Airborne Du Uranium Mass Balan Uranium in Major Conta Tower Ash a Other Conta Summary D Thorium Op Thorium Op Thorium Op Thorium Op Thoron 5.3.3.3.1 5.3.3.3.2 5.3.3.3.3 5.3.3.3.4 55, Domestic Radon Brea Raffinate Ex	Arichments orms and Compounds ist Potential ce, Material Flow, and Plutonium-239 Levels in Recycled the Weapons Complex aminants in Recycled Uranium and Plutonium Out of Specification aminants in Recycled Uranium befault Values for Recycled Uranium operations, 1954 to 1979 berations, 1954 to 1979 perations, 1979 to 2006 Operations and Handling, 1954 to 1971 Long-Term DOE Repository Storage, 1972 to 1985 Passive Storage, 1986 to 1987 Remediation, 1988 to 2006 c Uranium, and Raffinates th Analysis cposures	16 17 18 18 21 21 21 22 22 25 26 26 26 26 26 28 29 29 29 29 29 30 31 33
5.4	Radiol 5.4.1 5.4.2 5.4.3 5.4.4 5.4.5 5.4.6	5.3.4.2 ogical Cor Air Monite Bioassay Uranium In Vitro B Gross Be In Vivo B	Raffinate Ex 5.3.4.2.1 5.3.4.2.2 5.3.4.2.3 htrols Progra oring Progra Program Urinalysis P ioassay Pro ioassay Pro ioassay	Kposures K-65 Raffinate Drum Transfer Workers, 1952 to 1956 Plant 2/3 Workers Handling or Processing Pitchblende Ores, 1954 to 1958. Plant 1, 2, 3, and 8 Workers Handling or Processing Yellowcake, 1959 to 1961 am Trogram Program cedures for Other Radionuclides mple Results, 1957	33 33 34 34 35 36 37 39 40 40 41
5.5	Dose F 5.5.1	Reconstrue Uranium 5.5.1.1	ction Dose Asses Bioassay Da	ssmentsata	47 47 47

	5.5.1.2	Uranium Br	eathing Zone Results	47
	5.5.1.3	Uranium Er	nrichment	
	5.5.1.4	Recycled U	ranium	
	5.5.1.5	Chemical F	orm	48
	5.5.1.6	Unmonitore	d Uranium Workers	48
5.5.2	Thorium	and Thoron	Dose Assessments	48
	5.5.2.1	Monitored 7	horium Assessment Methods, 1979 to 2006	49
		5.5.2.1.1	In Vivo Bioassay, 1979 to 1994	49
		5.5.2.1.2	Breathing Zone Air Samples, 1995 to 2006	51
		5.5.2.1.3	In Vivo Bioassay and BZ Data, 1995 to 2006	52
	5.5.2.2	Assignment	of Unsupported Radium	
	5.5.2.3	Assignment	of Unmonitored Thorium Dose	53
		5.5.2.3.1	Thorium Coworker Model, 1979 to 1989	53
		5.5.2.3.2	Airborne Thorium Concentration, 1990 to 1994	53
	5.5.2.4	Assignment	t of Thoron Dose, 1954 to 2006	54
5.5.3	Raffinate	e Dose Asse	ssments	54
	5.5.3.1	1952 to 195	54	
	5.5.3.2	1954 to 195	i8 (Pitchblende Ores)	
	5.5.3.3	1959 to 196	1 (Yellowcake)	
5.5.4	Radon E	xposure Ass	sessment	55
5.5.5	Site Clos	sure		
Attribut	tions and	Annotations		55
nces				
<b>F</b> 1 /				74
u y	•••••			
	5.5.2 5.5.3 5.5.4 5.5.5 Attribu	5.5.1.2 5.5.1.3 5.5.1.4 5.5.1.5 5.5.1.6 5.5.2 Thorium 5.5.2.1 5.5.2.2 5.5.2.3 5.5.2.4 5.5.3 5.5.3.1 5.5.3.2 5.5.3.3 5.5.4 Radon E 5.5.5 Site Close Attributions and ences	5.5.1.2 Uranium Br 5.5.1.3 Uranium Er 5.5.1.4 Recycled U 5.5.1.5 Chemical F 5.5.1.6 Unmonitore 5.5.2 Thorium and Thoron 5.5.2.1 Monitored T 5.5.2.1.1 5.5.2.1.2 5.5.2.1.3 5.5.2.2 Assignment 5.5.2.3 Assignment 5.5.2.3 Assignment 5.5.2.3 Assignment 5.5.2.4 Assignment 5.5.3 Raffinate Dose Asse 5.5.3.1 1952 to 195 5.5.3.2 1954 to 195 5.5.3 Site Closure Attributions and Annotations forces	<ul> <li>5.5.1.2 Uranium Breathing Zone Results</li> <li>5.5.1.3 Uranium Enrichment</li> <li>5.5.1.4 Recycled Uranium</li> <li>5.5.1.5 Chemical Form</li> <li>5.5.1.6 Unmonitored Uranium Workers</li> <li>5.5.2 Thorium and Thoron Dose Assessments</li> <li>5.5.2.1 Monitored Thorium Assessment Methods, 1979 to 2006</li> <li>5.5.2.1.1 In Vivo Bioassay, 1979 to 1994</li> <li>5.5.2.1.2 Breathing Zone Air Samples, 1995 to 2006</li> <li>5.5.2.1.3 In Vivo Bioassay and BZ Data, 1995 to 2006</li> <li>5.5.2.4 Assignment of Unsupported Radium</li> <li>5.5.2.3 Assignment of Unmonitored Thorium Dose</li> <li>5.5.2.3.1 Thorium Coworker Model, 1979 to 1989</li> <li>5.5.2.3.2 Airborne Thorium Concentration, 1990 to 1994</li> <li>5.5.2.3 Assignment of Thoron Dose, 1954 to 2006</li> <li>5.5.3.1 1952 to 1954</li> <li>5.5.3.1 1952 to 1954</li> <li>5.5.3.1 1952 to 1954</li> <li>5.5.3.3 1959 to 1961 (Yellowcake)</li> <li>5.5.4 Radon Exposure Assessment</li> <li>5.5.5 Site Closure</li> </ul>

ATTACHMENT A	RATIONALE FOR 400 PPB U PU FOR 10A PROCESS STREAM	75
ATTACHMENT B	ANALYSIS OF POTENTIAL THORON EXPOSURES	84
ATTACHMENT C	INTERNAL DOSIMETRY URANIUM COWORKER DATA	. 114
ATTACHMENT D	EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA	. 131
ATTACHMENT E	TECHNICAL BASIS FOR ASSIGNING DOSES FROM EFFECTIVE DERIVED AIR CONCENTRATION BREATHING ZONE RESULTS	. 169
ATTACHMENT F	THORIUM IN VIVO COWORKER STUDY, 1979 to 1989	. 175
ATTACHMENT G	CLASS W THORIUM-232 INHALATION INTAKE RATE BASED ON 10% OF THE DERIVED AIR CONCENTRATION	. 188

# LIST OF TABLES

#### TABLE PAGE TITLE 5-1 Uranium enrichments and associated isotopes ......19 5-2 5-3 Uranium chemical forms......19 5-4 5-5 5-6 5-7

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 5 of 188

5-8	RU enrichment	. 25
5-9	Age of plutonium mixture in RU	. 26
5-10	RU contaminant mass concentrations	. 26
5-11	Inventory of stored thorium, 1987	. 27
5-12	Post-SEC thorium activities and regulatory requirements, 1988 to 2006	. 28
5-13	Isotopic composition of K-65 silos 1, 2, and 3	. 31
5-14	Radon breath analysis statistical parameters	. 32
5-15	Radium intake rate based on radon breath analysis data	. 33
5-16	Ratios of silo 1 and silo 2 nuclides to <sup>226</sup> Ra	. 33
5-17	Ratios of silos 1, 2, and 3 nuclides to <sup>230</sup> Th	. 34
5-18	Air monitoring analysis sheet codes	. 37
5-19	Air monitoring methods with applicable MDLs	. 37
5-20	Chemical toxicity and associated limits	. 38
5-21	Urinary uranium averages summary	. 39
5-22	Routine uranium in vitro bioassay capabilities	. 40
5-23	Fernald uranium urinalysis sample type codes	. 41
5-24	Fernald sample type coding system and personnel assignment codes	. 42
5-25	MDAs for nonuranium radionuclides	.43
5-26	Typical reporting of internal dose determined by direct lung count	. 43
5-27	Typical MIVRML counting schedule at Fernald in the 1970s	. 44
5-28	In vivo bioassay MDAs for the lung	. 45
5-29	Summary of MIVRML chest count MDAs by uranium enrichment, 1968 to 1989	. 46
5-30	Summary of IVEC chest count MDAs by uranium enrichment, 1989 to 2002	. 46
5-31	Routine uranium urinalysis capabilities	. 47
5-32	Default uranium enrichment	. 47
5-33	RU contaminant intakes per unit mass of uranium	. 48
5-34	RU contaminant intakes per unit activity of uranium	. 48
5-35	Methods for thorium dose determination	. 49
5-36	Summary of bias corrections for <sup>228</sup> Ac and <sup>212</sup> Pb in chest counts	. 50
5-37	Thorium chest count MDAs (nCi), 1979 to 2001	. 50
5-38	Custom nuclides from EDAC BZ results	. 52
5-39	Thorium coworker intake rates, 1979 to 1989	. 53
5-40	<sup>232</sup> Th inhalation and ingestion rates based on 10% DAC	. 54
5-41	Thoron worker exposure	. 54
A-1	Timeline of PGDP shipments to Fernald	.78
A-2	Totals of recycled feeds to Fernald	.78
B-1	Historical thorium mass balance	. 86
B-2	Thorium storage inventory, 1987	. 90
B-3	PAEC for thoron and progeny	. 90
B-4	Building dimensions and ventilation systems	. 91
B-5	Summary of thoron exposure estimates	. 96
B-6	Thoron exposure	.96
B-7	Summary of the 1996 through 1998 thoron measurement data	100
B-8	Summary of WL measurements	103
B-9	Three high measurements near Plant 8 silo and bins in 1986	104
B-10	Locations and numbers of samples	109
B-11	Thoron and thoron daughter results	110
C-1	Criteria applied to extract for uranium bioassav data	116
C-2	Reporting level	117
C-3	Summary of uranium urinary excretion rate analyses, 1952 to 2006	118
C-4	Derived uranium intake rates for type F materials, 1952 to 2006	121
C-5	Derived uranium intake rates for type M materials, 1952 to 2006	121
C-6	Derived uranium intake rates for type S materials, 1952 to 2006	122

Document No. ORAUT-TKBS-0017-5 Revision No. 01 Effective Date: 07/22/2016 Page 6 of 188
---

D-1	Summary of bias and DL for <sup>228</sup> Ac and <sup>212</sup> Pb for each year	. 134
E-1	Count of EDAC results in HIS-20	. 170
E-2	Activity fractions for the BL-13 EDAC	. 171
E-3	Isotopic composition of K-65 silo 1	. 172
E-4	Isotopic composition of K-65 silo 2	. 172
E-5	Activity fractions for the RT-210 EDAC	. 174
F-1	Bias for <sup>212</sup> Pb in chest	. 183
F-2	In vivo lung counting statistics and <sup>232</sup> Th lung burdens	. 184
F-3	Derived thorium intakes, 1979 through 1989	. 185

# LIST OF FIGURES

# **FIGURE**

Γ

# <u>TITLE</u>

#### PAGE

B-1	<sup>232</sup> Th decay diagram, including the <sup>220</sup> Rn (thoron) decay chain	87
B-2	Decay and buildup of <sup>228</sup> Th and progeny after processing	88
B-3	Thoron comparison, 1996 to 1998.	106
B-4	Measurements and lognormal fits, 1996	107
B-5	WL comparisons, 1986 through 2003	108
B-6	GA center of rod storage area	111
B-7	GA north end of rod storage area	112
B-8	GA TNT and sump cake storage area	112
B-9	GA wet area, main floor	113
B-10	GA Wet area, top deck	113
C-1	Change in lognormal fit for various CLs	118
C-2	Predicted values versus measured results, 01/01/1952 to 12/31/1993, 50th percentile,	
	type F	123
C-3	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile,	
	type F	123
C-4	Predicted values versus measured results, 01/01/1952 to 12/31/1993, 84th percentile,	
	type F	123
C-5	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile,	
	type F	124
C-6	Predicted values versus measured results, 01/01/1952 to 12/31/1993, 50th percentile,	
	type M	124
C-7	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile,	
	type M	124
C-8	Predicted values versus measured results, 01/01/1952 to 12/31/1993, 84th percentile,	
	type M	125
C-9	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile,	
	type M	125
C-10	Predicted values versus measured results, 01/01/1952 to 12/31/1954, 50th percentile,	
	type S	125
C-11	Predicted values versus measured results, 01/01/1955 to 12/31/1955, 50th percentile,	
	type S	126
C-12	Predicted values versus measured results, 01/01/1956 to 12/31/1956, 50th percentile,	
	type S	126
C-13	Predicted values versus measured results, 01/01/1957 to 12/31/1961, 50th percentile,	
	type S	126
C-14	Predicted values versus measured results, 01/01/1962 to 12/31/1966, 50th percentile,	
	type S	127

C-15	Predicted values versus measured results, 01/01/1967 to 12/31/1993, 50th percentile, type S	127
C-16	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile, type S	127
C-17	Summarized results of the predicted values versus the excluded results, 01/01/1952 to 12/31/2006, 50th percentile, type S	128
C-18	Predicted values versus measured results, 01/01/1952 to 12/31/1956, 84th percentile, type S	128
C-19	Predicted values versus measured results, 01/01/1957 to 12/31/1961, 84th percentile, type S	128
C-20	Predicted values versus measured results, 01/01/1962 to 12/31/1966, 84th percentile, type S	129
C-21	Predicted values versus measured results, 01/01/1967 to 12/31/1980, 84th percentile, type S	129
C-22	Predicted values versus measured results, 01/01/1981 to 12/31/1993, 84th percentile, type S	129
C-23	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile, type S	130
C-24	Summarized results of the predicted values versus the excluded results, 01/01/1952 to 12/31/2006, 84th percentile, type S	130
D-1	Normal probability plot of <sup>212</sup> Pb chest count results for 1978	133
D-2	Normal probability plot of <sup>212</sup> Pb chest count results for 1978 after adjusting for bias	133
D-3	Normal probability plot of 212Pb chest count results for 1978 after adjusting for bias	134
D-4	Plot of 228Ac versus 212Pb for 1978 where the levels of both nuclides are above the DL	135
D-5	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1978	136
D-6	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1979	137
D-7	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1980	138
D-8	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1981	139
D-9	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1982	140
D-10	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1983	141
D-11	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1984	142
D-12	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1985	143
D-13	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1986	144
D-14	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1987	145
D-15	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1988	146
D-16	Both radionuclides above the DI 1978	147
D-17	Fither radionuclides above the DI 1978	148
D-18	Both radionuclides above the DL 1979	149
D-19	Fither radionuclides above the DI 1979	150
D-20	Both radionuclides above the DL 1980	151
D-21	Fither radionuclides above the DL 1980	152
D-21	Both radionuclides above the DL 1981	152
D-22	Either radionuclides above the DL 1081	157
D-23	Both radionuclides above the DL, 1981	154
D-24	Either radionuclides above the DL, 1992	155
D-20	Roth radionuclides above the DL, 1902	150
D-20	Either radionuclides above the DL, 1903	157
ע-21 סי ח	Path radionuclides above the DL, 1903	100
D-20	Either radionuclides above the DL, 1904	109
D-29	Reth radionuclides above the DL, 1904	100
D-30	Duit radionuclides above the DL, 1903	101
D-31	Either radionuclides above the DL, 1985	102
D-32 D-33	Either radionuclides above the DL, 1986	163 164

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 8 of 188

D-34	Both radionuclides above the DL, 1987	. 165
D-35	Either radionuclides above the DL, 1987	. 166
D-36	Both radionuclides above the DL, 1988	. 167
D-37	Either radionuclides above the DL, 1988	. 168
E-1	<sup>222</sup> Rn decay chain showing half-lives	. 174
F-1	Natural thorium decay chain showing isotopes and half-lives	. 177
F-2	Relative activity of the isotopes in the natural thorium decay chain as a function of time	
	after chemical purification	. 178
F-3	<sup>228</sup> Th activity after a single chemical purification	. 179
F-4	<sup>228</sup> Ra and <sup>224</sup> Ra ingrowth after thorium chemical separation	. 180
F-5	<sup>228</sup> Th activity after a second chemical purification at its lowest equilibration point	
	(4.55 years) after the first purification	. 180
F-6	<sup>224</sup> Ra and <sup>228</sup> Ra activity after a second chemical purification when <sup>228</sup> Th was at its lowest	
	equilibration point (4.55 years) after the first purification	. 181
F-7	<sup>228</sup> Th activity after a third chemical purification when at its lowest equilibration point	
	(7.09 years) after the first purification	. 181
F-8	<sup>224</sup> Ra and <sup>228</sup> Ra activity after the third chemical separation	. 182
F-9	50th- and 84th-percentile <sup>232</sup> Th chest burdens, 1979 to 1988	. 184
F-10	Predicted thorium lung burden calculated using the three-separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	50th percentile, Type M	. 185
F-11	Predicted thorium lung burden calculated using the three-separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	84th percentile, Type M	. 186
F-12	Predicted thorium lung burden calculated using the three separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	50th percentile, Type S	. 186
F-13	Predicted thorium lung burden calculated using the three separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	84th percentile, Type S	. 187

# ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
AMC	African Metal Company
AWE	Atomic Weapons Employer
Bq	becquerel
BZ	breathing zone
CEDE	committed effective dose equivalent
CFR	Code of Federal Regulations
Ci	curie
CL	censoring level
cm	centimeter
d	day
DAC	derived air concentration
DCAS	Division of Compensation Analysis and Support
DL	decision level
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
dpm	disintegrations per minute
DWE	daily weighted (average) exposure
EDAC	effective derived air concentration
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
ERDA	U.S. Energy Research and Development Administration
EU	enriched uranium
FEMP	Fernald Environmental Management Project
FERMCO	Fernald Environmental Restoration Management Corporation
FMPC	Feed Materials Production Center
ft	foot
g	gram
GA	general area
gal	gallon
GDP	gaseous diffusion plant
GM	geometric mean
GSD	geometric standard deviation
HASL hr	Health and Safety Laboratory hour
ICP-MS	inductively coupled plasma mass spectrometry
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
IREP	Interactive RadioEpidemiological Program
IVEC	In Vivo Examination Center
keV	kiloelectron-volt, 1,000 electron-volts
kg	kilogram
KPA	kinetic phosphorescence analysis

L	liter
Ib	pound
LEU	low-enriched uranium
LOOW	Lake Ontario Ordnance Works
m	meter
MAC	maximum allowable concentration
MCW	Mallinckrodt Chemical Works
MDA	minimum detectable activity
MDL	minimum detection level
MeV	megaelectron-volt, 1 million electron-volts
mg	milligram
MIVRML	Mobile In Vivo Radiation Monitoring Laboratory
mL	milliliter
mo	month
MPLB	maximum permissible lung burden
mrem	millirem
MT	metric ton
MTTh	metric ton of thorium
MTTh	metric ton of uranium
NCG	NLO Concentration Guide (same as MAC for NLO only)
nCi	nanocurie
ng	nanogram
NIOSH	National Institute for Occupational Safety and Health
NLO	National Lead Company of Ohio
NOCTS	NIOSH-DCAS Claims Tracking System
NTS	Nevada Test Site
NU	natural uranium
OPOS	one person–one sample
ORAU	Oak Ridge Associated Universities
OSDF	Onsite Disposal Facility
PAEC	potential alpha energy concentration
PAEE	potential alpha energy exposure
pCi	picocurie
PGDP	Paducah Gaseous Diffusion Plant
PID	Principal Internal Dosimetrist
POOS	plutonium out of specification
ppb	parts per billion
ppm	parts per million
PUREX	plutonium-uranium extraction
RCRA	Resource Conservation and Recovery Act of 1976
RCS	Radon Control System
RF	release fraction
ROS	regression on order statistic
RT	radon treatment
RU	recycled uranium
s	second
SC&A	Sanford Cohen & Associates

SEC	Special Exposure Cohort
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
TBD	technical basis document
TOP	Thorium Overpack Project
TRU	transuranic
TTA	Transfer Tank Area
TWOPOS	time-weighted one person–one sample
U.S.C.	United States Code
WEMCO	Westinghouse Environmental Management Company of Ohio
WL	working level
WLM	working level month
yr	year
μCi	microcurie
μg	microgram
α	alpha particle
§	section or sections

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 12 of 188
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#### 5.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular Department of Energy (DOE) or Atomic Weapons Employer (AWE) facilities or categories of DOE or AWE facilities. They will be revised in the event additional relevant information is obtained about the affected DOE or AWE facility(ies). These documents may be used to assist NIOSH staff in the evaluation of Special Exposure Cohort (SEC) petitions and the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used to refer to an area, building, or group of buildings that served a specific purpose at a DOE or AWE facility. It does not mean nor should it be equated to an "AWE facility" or a "DOE facility." The terms AWE and DOE facility are defined in sections 7384I(5) and (12) of the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA), respectively. An AWE facility means "a facility, owned by an atomic weapons employer, that is or was used to process or produce, for use by the United States, material that emitted radiation and was used in the production of an atomic weapon, excluding uranium mining or milling." 42 U.S.C. § 7384I(5). On the other hand, a DOE facility is defined as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the [DOE] (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program);" and with regard to which DOE has or had a proprietary interest, or "entered into a contract with an entity to provide management and operation, management and integration, environmental remediation services, construction, or maintenance services." 42 U.S.C. § 7384I(12). The Department of Energy (DOE) determines whether a site meets the statutory definition of an AWE facility and the Department of Labor (DOL) determines if a site is a DOE facility and, if it is, designates it as such.

Accordingly, a Part B claim for benefits must be based on an energy employee's eligible employment and occupational radiation exposure at a DOE or AWE facility during the facility's designated time period and location (i.e., covered employee). After DOL determines that a claim meets the eligibility requirements under EEOICPA, DOL transmits the claim to NIOSH for a dose reconstruction. EEOICPA provides, among other things, guidance on eligible employment and the types of radiation exposure to be included in an individual dose reconstruction. Under EEOICPA, eligible employment at a DOE facility includes individuals who are or were employed by DOE and its predecessor agencies, as well as their contractors and subcontractors at the facility. Unlike the abovementioned statutory provisions on DOE facility definitions that contain specific descriptions or exclusions on facility designation, the statutory provision governing types of exposure to be included in dose reconstructions for DOE covered employees only requires that such exposures be incurred in the performance of duty. As such, NIOSH broadly construes radiation exposures incurred in the performance of duty to include all radiation exposures received as a condition of employment at covered DOE facilities in its dose reconstructions for covered employees. For covered employees at DOE facilities, individual dose reconstructions may also include radiation exposures related to the Naval Nuclear Propulsion Program at DOE facilities, if applicable. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction.

NIOSH does not consider the following types of exposure as those incurred in the performance of duty as a condition of employment at a DOE facility. Therefore these exposures are not included in dose reconstructions for covered employees (NIOSH 2010):

- Background radiation, including radiation from naturally occurring radon present in conventional structures
- Radiation from X-rays received in the diagnosis of injuries or illnesses or for therapeutic reasons

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 13 of 188
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#### 5.1.1 Purpose

This technical basis document (TBD) is part of the overall site profile for the Feed Materials Production Center (FMPC) site in Fernald, Ohio, which describes plant facilities and processes, historical information, and internal exposure data in relation to dose reconstruction for Fernald site workers. This TBD provides a technical basis for evaluating the occupational internal dose for EEOICPA claimants who were employed at the site. Since operations ended in 1988, the Fernald site has been variously named the Fernald Environmental Management Project (FEMP), the Fernald Closure Project, and is now the Fernald Preserve. This document uses FMPC or Fernald for convenience.

Occupational internal dose refers to radiation exposures workers received as a result of intakes of radionuclides from site operations as well as radon exposures, which might have been enhanced due to the nature of those operations. FMPC employees who did not have bioassay data or who were not fully monitored for internal exposures may be assigned coworker intakes, in addition to environmental doses, as applicable.

#### 5.1.2 Scope

This TBD describes the methods for assessment of internal radiation dose to workers at Fernald. The radionuclides of concern are primarily the isotopes of uranium and their progeny. Information is included on the various enrichments of uranium that were handled or processed and on the contaminants in recycled uranium (RU). The various chemical and physical forms of these materials are also covered. Because FMPC also processed some natural thorium, this TBD includes information on thorium and its decay products, including thoron and radium, and radionuclides associated with raffinate exposure.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.6.

#### 5.1.3 Special Exposure Cohort

The Secretary of the U.S. Department of Health and Human Services has designated three classes of employees at Fernald as additions to the SEC:

#### Thorium, 1968 through 1978

All employees of DOE, its predecessor agencies, and their contractors, or subcontractors who worked at the Feed Materials Production Center in Fernald, Ohio, from January 1, 1968 through December 31, 1978, for a number of work days aggregating at least 250 work days, occurring either solely under this employment, or in combination with work days within the parameters established for one or more other classes of employees included in the SEC (Sebelius 2012).

It was determined that NIOSH lacked sufficient information to allow it to estimate with sufficient accuracy the potential internal doses from exposure to thorium, to which employees at this facility may have been subjected (Sebelius 2012).

#### Thorium, 1954 through 1967

All employees of the DOE, its predecessor agencies, and their contractors and subcontractors who worked at the Feed Materials Production Center in Fernald, Ohio, from January 1, 1954, through December 31, 1967, for a number of work days aggregating at least 250 work days, occurring either solely under this employment, or in

combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort (Sebelius 2013a).

It was determined that NIOSH lacked sufficient information to allow it to estimate with sufficient accuracy the potential internal doses from exposure to thorium, to which employees working at this facility may have been subjected.

#### Uranium, 1951 through 1983 [applies only to subcontractors]

All employees of the Feed Materials Production Center in Fernald, Ohio, who were not employed by National Lead of Ohio, NLO, or the Department of Energy or its predecessor agencies, who worked at FMPC from January 1, 1951, through December 31, 1983, for a number of work days aggregating at least 250 work days, occurring either solely under this employment, or in combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort (Sebelius 2013b).

It was determined that NIOSH lacked sufficient information to allow it to estimate with sufficient accuracy the potential internal doses from exposure to uranium, to which some employees of FMPC working at this facility may have been subjected.

This dose reconstruction infeasibility for the period from 1951 through 1983 applies only to subcontractors. NIOSH has access to an electronic dataset that contains the results of the uranium urinalysis bioassay program for all years of FMPC operations, and the overwhelming majority of employees of the prime contractor National Lead Company of Ohio (later named NLO) have results in the bioassay dataset. However, the dataset does not contain bioassay results for employees of companies other than the prime contractor (i.e., non-prime contractor employees). NIOSH has obtained a limited number of bioassay samples from non-prime contractor employees through data captures, but cannot be certain that all non-prime contractor employee bioassay data were retained by the site or captured by data capture efforts. In addition, there are some reasons to conclude that the prime contractor did not consistently evaluate whether non-prime contractor employees should be monitored for radiation exposure because of the transitory nature of their work (Sebelius 2013b).

Dose reconstruction guidance in this document for the period from January 1, 1954, to December 31, 1983, is presented to provide a technical basis for partial dose reconstructions for claims not compensated under the SEC (i.e., nonpresumptive cancers and SEC employment less than 250 days). Although it is not possible to completely reconstruct internal radiation doses for all workers for this period, NIOSH has determined, and the U.S. Department of Health and Human Services has concurred, that it is feasible to reconstruct external radiation doses for all FMPC workers for this period (Sebelius 2013b).

#### 5.2 BACKGROUND

Construction at FMPC began in 1951 on a 1,050-acre site near the small rural community of Fernald, Ohio. Table 5-1 lists the five contractors to DOE and its predecessor agencies [i.e., the U.S. Atomic Energy Commission (AEC) and the U.S. Energy and Research Development Administration (ERDA)] that operated Fernald during its operational history.

The primary missions at FMPC were the processing of uranium ores to high-quality finished metal products and the production of thorium metal parts and thorium feedstock for processes at other DOE sites. The 10 plants that constituted the Fernald facility processed a variety of natural uranium (NU) and low-enriched uranium (LEU) ores and scrap materials for the creation of high-purity uranium metal parts. The thorium production included metal parts and feedstock for the weapons production programs. Operations involved thousands of metric tons of ores, dry powder products, and corrosive

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 15 of 188
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Table 5-1.	Contractors	during the	operational	history	/ of the	Fernald site.

Contractors	Dates	Time
NLO	October 1951 to January 1986	34+ yr
Westinghouse Materials Company of Ohio (WEMCO)	January 1986 to December 1992	7 yr
Fernald Environmental Restoration Management Corporation	December 1992 to 2006	14+ yr
(FERMCO), <sup>a</sup> Fluor Daniel Fernald, and Fluor Fernald	(transferred to DOE Office of	
	Legacy Management)	

a. FERMCO received the first DOE Environmental Restoration Management contract for FEMP.

chemicals in processes that were inherently dusty, which produced an environment with internal intake potential.

Due to the inherent nature of the processes, the limitations of the ventilation and material confinement systems, and the volume (and mass) of the materials, significant environmental and in-plant releases of radioactive materials occurred during FMPC operations. The work environment included a chronic potential for internal exposure, as demonstrated by the comprehensive air monitoring program and the urine sampling program for uranium (documented by air samples and uranium urinalysis). During the early years, plant workers were routinely required to wear respiratory protection because of significant radioactive dust levels approaching or exceeding FMPC airborne alpha activity guidelines referred to as maximum allowable concentrations (MACs) [or NLO Concentration Guide (NCG)] depending on the period. In addition, over the operational history of Fernald, metric tons of uranium and thorium products were released from the ventilation stacks to the environment.

The number of personnel at Fernald peaked at 2,891 in 1956 and slowly declined to 538 in 1979; an estimated total of 7,300 personnel worked at Fernald from 1953 through 1990. Given the site conditions of routine measurable air activity in the workplace and significant routine releases to the surrounding areas, all of these personnel had some potential for internal exposure from either being in process or other areas associated with loose contamination, or as a result of environmental releases. Additional radionuclides were introduced from other feedstock sources: radium and thorium from pitchblende ores from the Belgian Congo, transuranic (TRU) elements and fission products from RU, and thorium.

Pitchblende ores from the Belgian Congo (known as Q-11 material), which had unusually high radium and thorium activities, were processed from 1953 to 1955 as part of the uranium ores processing program. The liquid extraction column raffinates (known as K-65 material) from uranium extraction processes were stored in two dedicated concrete silos (by agreement, they belonged to the Belgian government). Additional pitchblende chemical extraction residues from another AEC site, Mallinckrodt Chemical Works (MCW), were added to the silos starting from the time of completion of the silos in 1953 and continuing until 1958. The materials in these silos became the property and responsibility of the United States when the lease agreement expired in 1983. This material remained on the Fernald site in the K-65 silos until March 2005 (Fluor Fernald 2006, p. 104) and represented a continuing internal exposure potential (for any operations in which direct contact with these residues was required) from the unusually high concentrations of <sup>226</sup>Ra and its progeny, <sup>210</sup>Pb and <sup>210</sup>Po. These three radionuclides comprise approximately 90% of the total activity in the K-65 materials. The uranium progeny <sup>230</sup>Th was also present in significant quantities (7% to 10%) in this location (see Table 5-13 later in this document). Historical environmental releases from this location, with the resultant internal dose potential, occurred from the ingrown radon gases and the progeny.

Fernald processed thorium from 1954 to 1979 and was the national thorium materials repository for DOE starting in 1972. About two-thirds of the material in the repository was processed and stored at Fernald, while the remainder originated at other DOE facilities. The thorium was stored as metal and stable chemical compounds [e.g., thorium gel, Th(OH)<sub>4</sub>] containing primarily <sup>232</sup>Th and its long-lived progeny <sup>228</sup>Th with all of the progeny present, which included the short-lived <sup>220</sup>Rn (thoron gas). The

original thorium inventory was contained in approximately 15,000 storage containers of various sizes. The January 2001 inventory was verified to be approximately 500,000 lb stored in approximately 200 metal boxes (Tomes 2001). Internal thorium exposure potential is associated with all of the thorium processes and during handling and repackaging of the thorium in storage drums.

The initial uranium processing started in 1951 with virgin stock from uranium mines and mills. RU was received and processed at Fernald as early as 1961. RU is that which has been used in a reactor environment (such as fuel elements or TRU production targets) and then processed to separate the unspent uranium from fission products, activation products, and TRU elements. RU contains trace quantities of plutonium isotopes, <sup>237</sup>Np, and <sup>99</sup>Tc as the primary contaminants. Some waste materials from the uranium enrichment facilities in the form of tower ashes were sent to Fernald for uranium recovery and had higher than average TRU contaminant concentrations as a result of enriching RU material. The levels of contaminants in these materials containing RU were known before receipt and blended with other uranium feedstock for processing. Personnel who were exposed to uranium contamination could also be exposed to the RU contaminants, which could have contributed to unmonitored internal exposure.

The original health and safety program was conducted with an industrial hygiene emphasis, based on uranium heavy-metal toxicology, with awareness and utilization of technology from other AEC site programs (e.g., Y-12 programs). Basic changes in the radiological protection program occurred with the contractor change in 1986 with the addition of radiation safety staff and increased analyses of all radiological hazards.

#### 5.3 RADIONUCLIDES OF CONCERN

The following are the primary radionuclides that could have led to internal doses during the production history of Fernald:

- Uranium isotopes <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, and <sup>238</sup>U (although <sup>236</sup>U is listed as an expected uranium component, is found primarily in RU and represents <1% of the dose from exposure to RU);
- Uranium progeny <sup>230</sup>Th, <sup>226</sup>Ra, <sup>222</sup>Rn, <sup>231</sup>Pa, and <sup>227</sup>Ac (<sup>230</sup>Th was present primarily as a uranium decay product);
- RU contaminants <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>241</sup>Am, <sup>237</sup>Np, <sup>99</sup>Tc, <sup>232</sup>Th, <sup>228</sup>Th, <sup>90</sup>Sr, <sup>103/106</sup>Ru, and <sup>95</sup>Zr/<sup>95</sup>Nb;
- Thorium isotopes <sup>232</sup>Th and <sup>228</sup>Th; and <sup>230</sup>Th
- Thorium progeny <sup>226</sup>Ra, <sup>228</sup>Ac, <sup>224</sup>Ra, <sup>220</sup>Rn, and <sup>212</sup>Pb (<sup>228</sup>Ac and <sup>212</sup>Pb are listed because they were used in in vivo counting to quantify the parents <sup>232</sup>Th and <sup>228</sup>Th).

Other infrequent, and perhaps one-of-a-kind, operations might have exposed workers to additional radionuclides. One example was the changing of the ion-getter vacuum pump blades in a Cockcroft-Walton neutron generator, which was performed on June 24, 1966. An approved procedure was developed and followed. Supplied air and protective clothing were worn, and the pump was disassembled in a hood. The workers might have been exposed to tritium combined with titanium inside the pump. A series of tritium urine bioassay samples were collected and evaluated at Mound Laboratory and indicated no positive results (Ross and Starkey 1966). If unusual bioassay results are found in claimant files, the Site Lead and Principal Internal Dosimetrist (PID) should be consulted.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 17 of 188
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#### 5.3.1 Uranium

The FMPC production facilities consisted of 10 production plants, each of which produced a product for the next process in a series of steps, terminating in high-purity uranium metal stock or parts. Operations in each major facility are briefly described below.

The Pilot Plant, as an operational prototype of the entire production process, began operations in October 1951. The purpose was to develop performance data for designing large-scale equipment for the rest of the plant. At the same time, limited quantities of uranium metal were produced during the initial Pilot Plant operation. In addition, UF<sub>6</sub> was converted to UF<sub>4</sub>. On February 14, 1966, an accidental release from the Pilot Plant of approximately 1,200 kg of uranium occurred during conversion operations that resulted in elevated personnel exposures (Warner 1966). The enrichment is unknown, so 2% should be assumed for any worker identified as a subject of this incident. In addition, more than 70% of the thorium at FMPC was handled and processed from 1964 to 1979 in the Pilot Plant.

Plant 1, the Sampling Plant, started operations in December 1953 and was later designated the official AEC sampling station for uranium along with isotopic assays of uranium ores and concentrates (usually  $U_3O_8$ ). The sampling process frequently involved the dusty operations of mixing, blending, etc. of large quantities of uranium and thorium materials.

Plants 2 and 3, the Ore Refinery Plants, began operations in December 1953 and achieved full-scale production early in 1954. The chemical processing in these facilities involved a three-step operation that began with nitric acid leaching of uranium from dry solid feed materials. This was followed by solvent extraction processing using a modified plutonium-uranium extraction (PUREX)<sup>1</sup> process to produce a high-purity solution of uranyl nitrate (UNH). The final step was a high-temperature thermal decomposition of the UNH solution to produce UO<sub>3</sub> product in dry, solid form and nitric oxides as effluent. After startup, these two plants generally operated as a single plant due to the integral process of the operations. During operations, the Plant 2/3 complex was a single large plant (2A), which was surrounded by several small buildings and tank farms that performed various support functions. For example, the Hot Raffinate Building (3E) was used to filter insoluble material from UNH and to process raffinates.

Plant 4, the Green Salt Plant, which converted  $UO_3$  to  $UF_4$  (green salt), began operating in October 1953 and achieved full-scale operation in 1954. The two-step process involved hydrogen reduction of  $UO_3$  powder to  $UO_2$  solids and then to  $UF_4$  by the addition of anhydrous hydrofluoric acid. These processes mixed, agitated, and transferred metric tons of solid uranium and produced some of the higher levels of airborne radioactivity at the site. Some air sample data sheet information indicates that at least a limited amount of thorium might have been processed in Plant 4.

Plant 5, the Metals Production Plant, provided a chemical conversion of UF<sub>4</sub> powder to a uranium metal "derby" by a thermite furnace reduction process with magnesium metal. This facility began operations late in 1953. This process concentrated the very small concentrations of remaining TRU elements and uranium progeny in the MgF<sub>2</sub> slag. The transport and mixing of feed materials and MgF<sub>2</sub> slag led to inhalation exposure potentials that were also in the higher level categories at the site.

Plant 6, the Metals Fabrication Plant, produced metal parts in rolling mills and machining lathes (as did Plant 9). Plant 6 began operations in 1953. Uranium metal fires were common, resulting in elevated airborne uranium concentrations.

<sup>&</sup>lt;sup>1</sup> The PUREX process is a chemical extraction technology for the separation of radionuclides from uranium and was used at several DOE sites in a variety of applications.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 18 of 188
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Plant 7, the Hexafluoride Reduction Plant, was a chemical process facility that operated from 1954 to 1957 and provided an additional source of  $UF_4$  by hydrogen reduction of  $UF_6$ .  $UF_6$  was converted by temperature control to the gaseous phase, which increased the potential for uranium inhalation exposures.

Plant 8, the Scrap Recovery Plant, provided a scrap uranium recovery function. Plant 8 began operations in 1954 and was also ranked in the higher air activity level areas at the site. Thorium scraps and residues were processed in 1966, and from 1969 to 1971.

Plant 9, the Special Products Plant, included casting of uranium metal and high-purity recycled metal scraps into ingots. Operations began late in 1953. Thorium was processed as metal and briquettes in 1954 and 1955.

## 5.3.1.1 Uranium Enrichments

Late in 1964, FMPC provided the first production of 1.95% <sup>235</sup>U billets for the Hanford Site. During the following production years, uranium was processed in a variety of enrichments ranging from depleted to as high as 20% (DOE 1996). The quantities of enriched material above 2% were not documented, but were qualitatively reported to be small and/or insignificant in total mass. The reported highest enrichment level processed in quantity was 2%. As another point of reference, a 1961 to 1984 history (Dolan and Hill 1988) of the average uranium enrichments in dust collector stack discharges demonstrates that <sup>235</sup>U enrichments ranged from 0.20% to 1.68% with an average of 0.70%. Of the total quantity of uranium that was received and processed at FMPC, less than 25% was enriched above normal (60,181 MTU of the total 246,683 MTU). Approximately 95% (208 g of the total 218 g) of the <sup>239</sup>Pu that was received at the site came in the enriched uranium (EU) receipts (DOE 2000a).

Table 5-2 lists the primary assumptions for FMPC uranium enrichments and the isotopes associated with these enrichments.

In the absence of specific enrichment information, and considering the above available data on processing experience of uranium enrichments at FMPC, the default assumption for periods after 1964 is 2% enrichment for bioassay data in milligram quantities of uranium. A 1% uranium enrichment can be assumed through 1964 to ensure results are favorable to claimants (NIOSH 2008, NIOSH 2014a).

## 5.3.1.2 Chemical Forms and Compounds

There are approximately seven steps in the process of conversion of uranium ore or other scrap recovery materials to metallic uranium. Those steps produce a number of compounds, each of which has specific chemical characteristics that are associated with different internal exposure parameters. Each of the compounds in Table 5-3 was handled in metric ton quantities. Most of the compounds were dry powder or granular in form and represented a dust hazard potential as the material was processed, transferred, and otherwise handled. Because all solubilities of uranium were handled in large quantities, all must be considered when assessing an intake unless specific information is available.

# 5.3.1.3 Airborne Dust Potential

Production operations that involved handling dry uranium materials were generally equipped with engineered ventilation systems for controlling dusts. Standard operating procedures required the use of respiratory equipment when dusty conditions were anticipated. Good housekeeping involving the immediate clean-up of spilled uranium products was also a standing policy and practice. In spite of this emphasis on engineered and administrative contamination controls and policy to reduce the

DU					
Relative activity					
Isotopes	Mass %	(pCi/µg)	Activity %		
U-234	0.001	0.062	15.46		
U-235	0.20	0.004	1.07		
U-236	0.00031	0.0002	0.05		
U-238	99.80	0.3354	83.42		
Total	Not applicable	0.402	Not applicable		
NU					
		Relative activity			
Isotopes	Mass %	(pCi/µg)	Activity %		
U-234	0.0054	0.334	48.86		
U-235	0.7204	0.016	2.28		
U-236	0	0	0		
U-238	99.274	0.334	48.86		
Total	Not applicable	0.683	Not applicable		
1% EU					
		Relative activity			
Isotopes	Mass %	(pCi/µg)	Activity %		
U-234	0.01	0.62	63.7		
U-235	1.0	0.02	2.1		
U-236	0	0	0		
U-238	98.99	0.333	34.2		
Total	Not applicable	0.973	Not applicable		
	2% EU				

Table 5-2. Uranium enrichments and associated isotopes.<sup>a</sup>

		2% EU	
Isotopes	Mass %	Relative activity (pCi/µg)	Activity %
U-234	0.02	1.243	76.9
U-235	2.0	0.043	2.68
U-236	0	0	0
U-238	97.98	0.329	20.4
Total	Not applicable	1.616	Not applicable

a. U-236 is less than 1% mass activity in DU, NU, 1% EU, and 2% EU.

Table 5-3. Uranium chemical forms.

Uranium compound	Chemical formula	Lung absorption type
Uranium hexafluoride (gas)	$UF_6$	F
Uranyl fluoride	$UO_2F_2$	F
Uranyl nitrate	$UO_2(NO_3)_2$	F
Uranium trioxide (orange oxide)	UO₃	М
Uranium tetrafluoride (green salt)	UF <sub>4</sub>	М
Uranium dioxide (brown oxide)	UO <sub>2</sub>	S
Uranium tetraoxide	UO <sub>4</sub>	М
Uranium oxide (yellowcake)	U <sub>3</sub> O <sub>8</sub>	S
High fired uranium oxides	UO <sub>2</sub>	S

release of radioactive materials, spills and routine releases occurred. In addition to the routine releases at FMPC, there were frequent "upset" conditions (i.e., spills, effluent filter ruptures, etc.) that produced episodic airborne radioactivity in the work areas and plant effluents, and were of a magnitude such that the ventilation systems were unable to contain all of the releases.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 20 of 188
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In 2000, a Fernald team working on the DOE *Ohio Field Office Recycled Uranium Project Report* (DOE 2000a) qualitatively rated various plant processes in relation to the potential for producing airborne dusts in high, medium, or low categories, based on the criteria below. Though qualitative, this evaluation was consistent with historical site air activity measurements and recorded internal exposures. The evaluation involved the following ratings and the qualitative descriptions of "very large quantities," "small quantity, intermittent," and "highly reliable" are used as quoted in the reference:

- High ratings were assigned for operations that processed very large quantities of materials using processes or equipment susceptible to occasional events. Examples of such events include digestion tank fuming and denitration pot eruptions in Plant 2/3, failure of hydrofluorination banks in Plant 4, temperature excursions in Plant 8 furnace operations, and failure of ventilation system dust bags during both operation and maintenance. The ventilation systems were inadequate to contain the dust emissions from these events, and all plant workers were susceptible to exposure from these releases.
- Low ratings were assigned for small quantity, intermittent operations, those that did not involve appreciable amounts of dry materials, and those considered highly reliable.
- Medium ratings were assigned for operations which were neither of clearly high nor clearly low radioactive material release potential.

Table 5-4 lists the summary results of this evaluation including related information on the typical materials.

			Airborne dust potential
Plant	Process	Compound <sup>a</sup>	(qualitative)
5	Metal reduction and casting	$UF_4$ , U, U <sub>3</sub> O <sub>8</sub> , residues	High
8	Feed preparation furnaces	U <sub>3</sub> O <sub>8</sub> , U, residues	High
4	Banks 7–9, packing stations	UO <sub>3</sub> , UO <sub>2</sub> , UF <sub>4</sub>	High
2/3	Digestion, extraction denitration	Prepared feed, U <sub>3</sub> O <sub>8</sub> , UO <sub>2</sub> , UO <sub>3</sub>	Medium
9	Reduction, casting	UF <sub>4</sub> , U, U <sub>3</sub> O <sub>8</sub> , residues	Medium
Pilot	Hex reduction, metal operations	UF <sub>4</sub> , U, U <sub>3</sub> O <sub>8</sub> , residues	Medium
1	Milling	U, U <sub>3</sub> O <sub>8</sub> , residues	Low

Table 5-4. Plant processes, materials, and uranium dust release potential.

a. MgF2 (one of the residue products) in Plants 1, 2/3, 5, 9, and the Pilot Plant, contributed to the dust potential rating, but did not carry more than trace quantities of radionuclides. The hazard potential for MgF2 typically was associated with nonuranium radionuclides (i.e., uranium progeny and RU contaminants).

Accordingly, jobs at Fernald which contributed to higher airborne uranium exposures in routine operations included (Heatherton 1975, p. 159):

- Changing hoppers;
- Lidding and delidding drums;
- Handling contaminated drums;
- Adjusting weights at filling stations;
- Dumping drums of concentrate;
- Operating pot-filling machine in the Metals Plant;
- Breakout of furnace pots and molds;
- Cleaning uranium surfaces, graphite crucibles and molds;
- Reassembly of crucible and mold parts;
- Operating crushing and grinding equipment;
- Changing receiving drums at dust collectors;

- Cleaning out dust collector housings and furnace enclosures; and
- Breaking up clogged material in containers, conveyors, downcomers, and other equipment.

#### 5.3.2 Recycled Uranium

This section extracts some of the information on RU in ORAUT-RPRT-0052, *Feed Materials Production Center Internal Dose Topics* (ORAUT 2011a), which can be reviewed for additional details.

RU is uranium that has been recovered from irradiated production reactor fuel and plutonium production fuels. The RU was separated in the chemical processing plants at Hanford and the Savannah River Site (SRS), but was known to contain traces of TRU elements and fission product impurities. Most of the RU delivered to Fernald came from the Hanford Site, the Paducah Gaseous Diffusion Plant (PGDP), and SRS. The primary contaminants were <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>241</sup>Am, <sup>237</sup>Np, and a fission product, <sup>99</sup>Tc. The <sup>239</sup>Pu, <sup>237</sup>Np, and <sup>99</sup>Tc were the radionuclides of greatest concentrations and were tracked and documented for control purposes.

The first uranium feedstock introduced to the Fernald plant in 1953 was high-grade ores (50% to 80% uranium) (i.e., pitchblende ores from the Belgian Congo and other locations, which could be processed in a modified PUREX process directly to high-purity uranium). Other uranium feedstock came from low-grade ores, which was processed near the mines by mills that produced NU in the form of yellowcake ( $U_3O_8$ ) in the range of 60% uranium. The yellowcake required further processing by PUREX to a high-purity state. When RU from the chemical processing plants was introduced to the process feed streams at Fernald it was a high-purity oxide and was normally blended with NU or other existing feedstocks.

Relatively small quantities of RU (a recorded mass of 45 MT RU) from Hanford were received at FMPC as early as 1955 (DOE 2000a, 2000b) but did not constitute a feed stream component of significance. The first significant quantity of RU was received for processing on February 13, 1961, in a shipment of UF<sub>6</sub> that was to be converted to UF<sub>4</sub> (DOE 2000b) and introduced to the feed streams of the plant.

#### 5.3.2.1 <u>Mass Balance, Material Flow, and Plutonium-239 Levels in Recycled Uranium in the</u> <u>Weapons Complex</u>

From the beginning of operations, urinalyses for uranium were conducted extensively for purposes of controlling biological heavy-metal concerns. The presence of RU contaminants was recognized but was not specifically analyzed due to the anticipated low levels and related low-hazard contribution. The concentration of contaminants in RU changed with time, so default values for dose reconstruction are defined for discrete time intervals. Because this represents a bounding approach, it is not necessary to precisely quantify the mass balance and material flow of RU in the weapons complex to establish a bounding estimate of the contaminant concentration in RU at Fernald.

While DOE documents about mass flows of RU in the weapons complex include some discrepancies, these discrepancies in mass flow do not preclude use of the data to define bounding default assumptions. The defaults were chosen such that they bound the highest values of all the processes, with the exception of the concentrations in the gaseous diffusion plant (GDP) tails and wastes.

The concentration of the contaminants varies slightly with the source of the data. For example, the mass concentration of plutonium in DOE (2000b) is 0.9 ppb; for the complex-wide rollup, the mass concentration is 3.7 ppb; in DOE (2003), the mass concentration from the primary sites is 4.2 ppb. These values are essentially the same and all well below the 10-ppb control guideline at Hanford.

	Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 22 of 188
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#### 5.3.2.2 Major Contaminants in Recycled Uranium

Table 5-5 details DOE's estimate of the quantity and characteristics of RU and its major contaminants at Fernald. Approximately 218 g of <sup>239</sup>Pu, 25.7 kg of <sup>237</sup>Np, and 332 kg of <sup>99</sup>Tc were received in 246,683 MT of RU during the history of the plant (DOE 2000b). The historical average of contaminants in RU was approximately 1 ppb <sup>239</sup>Pu, 104 ppb <sup>237</sup>Np, and 1,346 ppb <sup>99</sup>Tc.

Uranium enrichment	MTU receipts	Percent uranium receipts	Total Pu- 239 (g)	Pu-239 (ppb)	Total Np-237 (g)	Np-237 (ppb)	Total Tc-99 (g)	Tc-99 (ppb)
EU	60,181	24.4	207.9	3.5	19,048	316	328,740	5,382
NU <sup>a</sup>	89,649	36.3	4.1	<0.1	3,026	34	1,197	13
DU	96,853	39.3	5.7	<0.1	3,669	38	2,061	21
Total or average	246,683 total	100.0 total	217.7 total	0.9 average	25,743 total	104 average	331,998 total	1,346 average

Table 5-5. Average characteristics of RU and its major contaminants.

a. Referenced as "normal uranium," but is natural uranium for the purposes of discussion.

A limit of 10 ppb total plutonium in RU was set by the nuclear weapons complex (primarily Oak Ridge) for use at Hanford (ORAUT 2015a; DOE 2000a). Initially no numeric limits were specified for <sup>237</sup>Np or <sup>99</sup>Tc. The plutonium limit of 10 ppb was set such that the radiological impurities did not exceed 0.1% of the alpha activity from NU.

#### 5.3.2.3 Tower Ash and Plutonium Out of Specification

The gaseous diffusion process concentrated the RU contaminants, primarily due to lack of volatility of the fluoride compounds in comparison with those of uranium, in tower ash and other residues in the  $UF_6$  conversion and handling processes. These ash and residues also contained significant amounts of uranium, which required that they be recovered according to economic discard criteria. In 1972, the decision was made by the AEC to process these materials from PGDP and the other GDP sites. Fernald was directed to process most of these materials. This decision introduced RU contaminants to Fernald with plutonium out of specification (POOS), meaning that it exceeded the 10 ppbU limit.

POOS feedstock from PGDP was sent to FMPC for reprocessing with the prior knowledge that the material was known to be above the established contamination limit. The 1970s and increased amounts in the 1980s GDP shipments accounted for a major portion of the total TRU inventory at FMPC, essentially doubling the plant inventory over this period (Bassett et al. 1989; DOE 2000b). The intent was to blend these materials with the inventories at the plant and still maintain the plantwide limits that had been determined to provide an adequate level of safety (i.e., staying below the average sitewide plutonium specification of 10 ppb). Workers handling POOS materials directly at Plant 1 and other locations were protected with airline respiratory equipment, particularly for the 1976 shipment of tower ashes from PGDP. Consequently, exposures to RU contaminants greater than the 10 ppbU could have increased significantly starting in 1973.

An examination of the statistical data sheets in Appendices C and F of DOE (2000b) indicates that maximum values in some subgroups (as determined by the 95th percentile of a lognormal distribution) ranged up to approximately 400 ppb Pu. These values are summarized in Table 5-6. A notable exception is the tower ash from PGDP, which had a mean value of 1,732 ppb Pu (range of 0.6 to 3,505). This maximum-level POOS material was handled as a recognized short-term risk during feed preparations and blending operations. Additional protective precautions were applied during these short-term tasks, and radiological procedures were implemented (WMCO 1986a; Jelinek and Thomas 1985). These precautions included airline respiratory protection and protective clothing over normal plant clothing that consisted of head covering, gloves, and shoe covers. Tape was applied at ankles and sleeves. Weekly urine bioassay was required of those involved (Author unknown undated a).

	Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 23 of 188
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Subgroup	Description	Pu ppb U at 95%	Np ppb U at 95%	Tc ppb U at 95%
1A	Miscellaneous	133	3,692	3,060
1B	Miscellaneous – minor offsite	1.6	179	1
2	UF <sub>6</sub> source UF <sub>4</sub> (GDP tails)	1.2	84.	171
3	UF <sub>6</sub> source metal and scrap	0.01	4.2	15
4	Normal U product, residues, and scrap	0.12	139	33
5	Enriched UF <sub>6</sub> source products and residues	3	109	15,410
6A	UO <sub>3</sub> PUREX source (A508)(unblended)	4.6	655	14,119
6B	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (low cross)	4.1	643	19,732
6C	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (high cross)	34	1,198	4,823
6D	A500 coded enriched residues	7.6	246	2,701
6E	SRS UNH	37.8	Not measured	Not measured
6F	SRS UO <sub>3</sub> – not shipped to FMPC	4.6	Not measured	Not measured
7A	A508 based derbies	17	513	2,870
7B	A508 based ingots and metal	1.7	433	133
8 <sup>a</sup>	Enriched MgF <sub>2</sub>	342	4,417	4,619
9	GDP incinerator ash and scrap residues	283	5,116	474
10A	Tower ash and decontamination residues	1,732	17,277	4,445
10B	UO <sub>3</sub> from tower ash	34.9	842	4,154
11 <sup>a</sup>	Waste residues	152	10,666	6,991

Table 5-6. RU contar	ninant levels b	y process	subgroups.
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a. Although the results in the table are reported in ppb U, this measure might appear less meaningful in subgroups in which there is a reduced amount of uranium (in one-digit percentage levels), such as subgroups 8 and 11, in which the MgF<sub>2</sub> and chemical plant raffinates accumulated some isotopes but were relatively low in uranium by design, yet still continued to be a process stream to recover uranium above the economic discharge levels. Note that even with decreased uranium levels the contamination default can still be a ratio of uranium.

The Fernald Health and Safety staff was aware that TRU and fission product contaminants were present in the RU, although they believed it to be less than 0.1% of the total alpha activity. Therefore, the biological concerns were thought to represent an increase of less than 10% to 15% in the hazard level. Radiological control was administered continually on the basis of both air analysis (gross alpha counts) and urine bioassay for uranium. Before 1989, no TRU analyses for radiological safety were performed on a routine basis for either airborne or urine activity, and exposure controls remained based on chemical toxicity under the assumption that these controls would be sufficient for all the radiological issues (Bassett et al. 1989). Although the alpha activity from the TRU alpha emitters would have been collected and detected on the air samples, the reported results were all considered to be uranium and compared to the MAC (WMCO 1986a).

Over time, much of the POOS material was blended with the NU feedstock and with RU of typical concentration. This was done to maintain the plantwide limits that had been determined to provide an adequate level of safety (i.e., staying near the average complexwide plutonium specification of 10 ppb). Workers handling the POOS materials directly before blending with other materials (Plant 1 and other locations) were provided with special protection. As a background comment, a small amount of this POOS material from the GDP was inadvertently stored for a period of years. The discovery of this material in 1985 was categorized as an unusual event and the material was subsequently processed and disposed.

After processing the PGDP POOS, the blended uranium oxide resulted in temporary storage of uranium with a maximum concentration of plutonium of 43 ppb U. In addition, several of the uranium purification processes resulted in concentrating the TRU contaminants in waste streams. Formal investigations were conducted that traced the TRU contaminants through the various plant processes. One study in 1975 defined contaminant affinity for the metal production furnace mold materials (some TRU studies are referenced in DOE 2000b). These analyses demonstrated that the extraction and

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 24 of 188
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metal conversion processes concentrated plutonium, neptunium, and uranium progeny contaminants in the raffinate waste of the liquid extraction columns and in the MgF<sub>2</sub> from the thermite metal conversion furnaces. MgF<sub>2</sub> slag from the thermite metal reduction process was also processed through a slag leach process and then through the chemical extraction plant. These concentration points or areas were identified by analytical methods in early years.

The plutonium contaminant mass concentrations are therefore assumed to be 400 ppb U after 1972. The basis for assuming 400 ppb U rather than the 1,732 ppb U for subgroup 10A is briefly described here. The high values in the activity distribution came from PGDP in a single shipment in 1980. In June of 1980 a single shipment of approximately 24 MTU PGDP process plant ash was shipped in 16 T-hoppers that contained 25.3 g of plutonium. This shipment was sampled and stored primarily to develop blending material inventories and processes to accommodate this unusual shipment. In 1982 after about 2 years, there is a record of repackaging five of the T-hoppers with the highest contaminant levels into large containers in Plant 4 to facilitate process needs in Plants 8 and 2/3. Based on the recorded times of the five hoppers with the greatest problems, 150 hours was conservatively estimated for repackaging those eight hoppers with concentrations above 400 ppb U plutonium. The next step (after another delay) was blending with sump cake in Plant 8 of appropriate content to create a calcium uranate feed for further dissolution and process in Plant 2/3, these being semi-remote operations. The final recorded processing resulted in a formal approval to process the Plant 2/3 UO<sub>3</sub> product, which was in the range of 20 to 42 ppb U plutonium). Therefore, the period of time material in excess of 400 ppb U was handled was relatively brief. Airline respirators and associated protective clothing were used, which ensures that 400 ppb is sufficiently conservative for dose reconstruction. Attachment A, Rationale for 400 ppb U Pu for 10A Process Stream, provides greater details.

From April 1986 to 1989, 675 bioassay samples from 441 workers were collected for plutonium analysis. "Those samples were collected primarily from workers who were expected to be and, in some cases, were actually involved in POOS processing and cleanup campaigns in Plants 4 and 8" (Bassett et al. 1989). Only 10 individuals exhibited quantities of plutonium in the urine above 0.02 dpm/sample. [This contractual urine minimum detectable activity (MDA) was determined to be unrealistically low and later adjusted to 0.13 dpm/sample for plutonium urinalyses.] Further investigation (follow-up samples) revealed that 3 of the 10 were below the specified detection limit of 0.02 dpm/sample. All of the 10 individuals were flown to Hanford, where in vivo counts were conducted for plutonium. All results were below the detection limit of the Hanford in vivo analyses. The conservative assumption was made by the site contractor that 7 of the 10 were technically at the urine MDA. Directions were given to "regard those results as valid indicators of exposure and to calculate the plutonium dose commitments to those workers based on these worst-case assumptions. The dose commitments are to be reported in each employee's permanent exposure record" (Bassett et al. 1989). Fernald radiation protection practices were adjusted to more rigorously account for those trace isotopes within the DOE radiation protection standards in effect at the time. The information above is taken from the report of a DOE team of investigators and documents an organized effort by an incoming contractor to verify the previous assumptions that TRU exposures were not a major exposure source.

In 1986 and later when <sup>238</sup>Pu and <sup>239/240</sup>Pu urine sample results are available for full-time POOS employees, the RU contaminants are based on the uranium intake. The purpose of these samples was for the site to address the issue of exposures from RU contaminants and that there was no indication of work with pure plutonium at Fernald. Therefore, because plutonium is addressed as part of the RU contaminants, missed dose associated with plutonium bioassay results are not assigned. The only exception is for a positive <sup>239/240</sup>Pu sample that is greater than the derived intake for uranium. As noted above, only 10 individuals exhibited quantities of plutonium in the urine above 0.02 dpm/sample (the initial MDA).

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 25 of 188
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Dose reconstructors should contact the Site Lead and PID for guidance if a claim file contains positive plutonium bioassay results.

#### 5.3.2.4 Other Contaminants in Recycled Uranium

Routine process chemical analysis at Hanford included gross beta and gamma measurements. These results were compared to the baseline measurements from aged NU (DOE 2000a). Limits on these other contaminants were set to 0.685 pCi/µg NU (beta/gamma), 50 µCi/lb U for <sup>103</sup>Ru or <sup>106</sup>Ru, 15  $\mu$ Ci/lb U for <sup>95</sup>Zr and <sup>95</sup>Nb, and 2  $\mu$ Ci/lb U for all other isotopes.

The mass balance reports established that most of the RU came from Hanford and was recycled from weapons-grade plutonium. Therefore, a 6% weapons-grade mixture that had been chemically purified in 1961 was selected for bounding calculations. For dose reconstruction, the plutonium alpha dose from the plutonium mixtures in RU is assumed to be from 100% <sup>239</sup>Pu. Plutonium mixture ratios for fresh, 10-, 20-, and 30-year aged intervals after purification were used to estimate the <sup>241</sup>Pu and <sup>241</sup>Am contaminant levels.

Specific analytical information for trace concentrations of other fission products as documented in a Hanford reference (DOE 2000a) are listed in Table 5-7.

Element/isotope	Observed range	
Plutonium (primarily Pu-239)	<1 to 2 ppb U	
Neptunium (primarily Np-237)	0.04 to 0.16 ppm U	
Thorium	8 to10 ppm U	
Technetium	3 to 4 ppm U	
Ru-103/106	<6 µCi/lb U	
Zr/Nb-95	<4 µCi/lb U	
Americium-241	40 to 320 Bq/g U	
Other gamma emitters excluding Tc-99	0.09 to 0.75 µCi/lb U	

Table 5-7. Other radioactive contaminants in RU from Hanior	able 5-7.	minants in RU from Hanford
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#### 5.3.2.5 Summary Default Values for Recycled Uranium

RU was first processed in significant amounts at FMPC beginning in 1961, so none of the associated contaminants were present before 1961. Therefore, uranium intakes in 1961 and later should include default factors to account for exposure to the RU contaminants. Table 5-8 lists the uranium enrichment assumptions during the RU period.

Table 5-8. RU enrichment.		
Period	Enrichment	
1961–1964	1%	
1965-2006	2%	

Table 5-9 lists the plutonium mixture ages assumed during the RU period.

Contaminant concentrations between 1961 and 1972 for plutonium alpha, <sup>237</sup>Np, and <sup>99</sup>Tc are based on the pre-POOS concentrations explained in Section 5.3.2.2. Contaminant concentrations from 1973 on for plutonium alpha, <sup>237</sup>Np and <sup>99</sup>Tc are based on bounding POOS concentrations explained Section 5.3.2.3. Contaminant concentrations for <sup>232</sup>Th, <sup>103/106</sup>Ru, <sup>95</sup>Zr/<sup>95</sup>Nb, and <sup>90</sup>Sr are based on the Hanford limits and observed concentrations explained Section 5.3.2.4. These are summarized in Table 5-10.

Table 5-9. Age of plutonium mixture in RU.

Period	Age
1961–1965	Fresh
1966–1975	10 Year
1976–1985	20 Year
1986-2006	30 Year

#### Table 5-10. RU contaminant mass concentrations.

RU contaminant	1961–1972	1973–present
Pu-alpha <sup>a</sup>	10 ppb U	400 ppb U
Np-237	400 ppb U	11,000 ppb U
Tc-99	6,000 ppb U	20,000 ppb U
Th-232⁵	10,000 ppb U	10,000 ppb U
Ru-103/106 <sup></sup>	50 µCi/lb	50 µCi/lb
Zr-95/Nb-95 <sup>d</sup>	15 µCi/lb	15 µCi/lb
Sr-90	2 µCi/lb	2 µCi/lb

a. The plutonium alpha mixture is assessed as 100% Pu-239. Am-241 and Pu-241 are assessed based on 6% weapons-grade plutonium mixture.

b. An intake of Th-228 should be added and assumed to be at 70% equilibrium with Th-232.

c. Ru-103/106 is assumed to be 100% Ru-106 due to its longer half-life.

d. Zr-95/Nb-95 pair is assumed to be in equilibrium with equal activities totaling the intake rate indicated in the table.

#### 5.3.3 Thorium

#### 5.3.3.1 Thorium Operations, 1954 to 1979

According to a timeline of thorium processing operations (ORAUT 2008a), thorium processing began at Fernald in 1954 and continued through the first quarter of 1979.

SEC classes have been designated based on inability to reconstruct thorium dose with sufficient accuracy for periods beginning in 1954 and ending in 1978 (Section 5.1.3). Therefore, no detailed information or process description for activities before 1979 is provided.

#### 5.3.3.2 Thorium Operations, 1979 to 2006

Thorium processing was completed in 1979, after which thorium exposure was due to repackaging and shipping operations.

In 1988, an environmental assessment indicated that the inventory of thorium materials at Fernald consisted of approximately 1,100 MT (as thorium) of various thorium compounds. In 1988, the Plant 8 silo and bins contained approximately 175 MT of bulk thorium oxide materials plus other inert materials. The remainder of the thorium inventory was stored in various sizes of container in warehouse Buildings 64, 65, 67, and 68 and as thorium nitrate in Tank 2 at the Pilot Plant. A small number of drums were in outside storage adjacent to Building 65 (DOE 1987a, p. 10). Table 5-11 summarizes Fernald's stored inventory of thorium in 1987 (Reafsnyder 1987, p. 3).

This includes a total of 13,329 drums and 2,062.2 MT of thorium material (Grumski 1987, p. 13), plus the Plant 8 silo and bins material and the thorium nitrate solution in the Pilot Plant tank. In 1985, NLO identified the need to repackage much of the thorium stored in 55-gal drums due to "serious

Location	Description or Quantity	Weight (MTTh)
Plant 8 silo and bins	Bulk thorium oxide material and other inert materials	175
Pilot Plant Tank 2	Thorium nitrate solution	9
Building 64 (Plant 9 warehouse)	181drums	64.2
Building 65 (Plant 5 warehouse)	5,599 drums	1,246.3
Building 67 (Plant 1 storage building)	5,992 drums	213.5
Building 68 (Pilot Plant warehouse)	1,317 drums	485.7
Pad west of Building 65	212 (240 overpacked) drums	52.5
Totals	Not applicable	2246.2

Table 5-11. Inventory of stored thorium, 1987.

deterioration" (Adams 1985). Some repackaging was done in 1985 involving only a few individuals, during which breathing zone (BZ) samples were taken (Mengel 1985). In 1986, possible overstressed conditions in the Plant 8 silo and bins used to store thorium oxide were identified (WMCO 1986b).

In 1988, Fernald initiated a "three Project plan" for improving the temporary storage conditions for thorium (WMCO 1992a, p. 203; Reafsnyder 1987). During the execution of the plan, approximately 1,094 MTTh were declared waste (WMCO 1992a). The "three Project plan" was followed by two additional phases which resulted in the removal of all thorium from the site.

- <u>Project 1</u>. June 1988 (anticipated schedule, see DOE 1987b, p. 19) to March 1989, addressed the bulk thorium materials in the Plant 8 silo and bins. As the bulk thorium was removed from the silo and bins, it was placed in double-containment drums called overpacks (a 48-gal drum was packaged inside a 55-gal drum), inventoried, and monitored. The drums were then stored in an onsite warehouse along the northern edge of the production area, away from daily plant operations. The silo and bins were then decontaminated and demolished (WMCO 1992a). This work was subcontracted to International Technology (IT) Corporation, which participated in the site's radiation safety program, and the employees assigned to the project were required to receive in vivo counts at 6-month intervals (see whole-body counts in February 1989 in Walker 1989).
- <u>Project 2</u>. January to March 1990, addressed overpacking of the 241 containers stored outdoors; 212 of the containers were drums. A remote system to handle, identify, and overpack the thorium drums and containers was designed. Each container was inventoried, weighed, overpacked, and then placed in temporary indoor storage (WMCO 1990a). For Project 2, isotopic fecal sampling was recognized as the only method capable of monitoring intakes below the annual limit on intake (WMCO 1991, p. 4). The detection limit of this analysis was 1.0 dpm (WMCO 1991, p. 6 and others). As a result, starting with this project, baseline fecal samples were taken from the directly exposed workers (chemical operators, forklift operators, etc.). These results are included in the site's HIS-20 database. General area (GA) samples were taken and each operator was required to wear a BZ sampler when inside the radiological barrier (Walker 1990, p. 3). These samples are not in the HIS-20 database and are therefore unlikely to be reported to NIOSH by DOE.
- <u>Project 3</u>. 1991 to 1995, involved overpacking 13,000 containers of thorium in warehouses. This project included overpacking in the Thorium Warehouse, the Plant 1 Thorium Warehouse, the Pilot Plant Warehouse, and the (old) Plant 5 Warehouse (WMCO 1992a). Radiological controls similar to those in Project 2 were in place during these operations. The first thorium was shipped to the Nevada Test Site (NTS) in 1992 (FERMCO 1993a), and shipping continued each year thereafter. Almost 6,000 gal of thorium nitrate, a contaminated acid waste stream, were treated and solidified in 1995 (FERMCO 1996).

- <u>Thorium Overpack Project (TOP)</u>. Project 3 was succeeded by the TOP in 1996, which completed the overpacking of the drums stored in warehouses in 1997. This did not include some 8,500 drums of thorium waste that required Resource Conservation and Recovery Act of 1976 (RCRA) classification (FDF 1998, p. 54).
- <u>RCRA classification and shipping for treatment or disposal, 1997 to 2006</u>. Characterization of the remaining 8,500 containers of thorium legacy waste was initiated in 1997. Through the end of 2003, over 8,400 of these containers were shipped off the site for treatment, with subsequent disposal at NTS. The remaining thorium inventory of approximately 100 containers had been evaluated. Of these, approximately 90 containers were non-RCRA, low-level radioactive waste and 10 were RCRA hazardous waste. From 2004 to 2006, the remainder of the mixed thorium waste was shipped to Envirocare of Utah for treatment and disposal; the nonhazardous thorium waste was shipped to NTS for disposal. At the end of 2004, there were only a few remaining containers of thorium waste. The last shipment of hazardous waste occurred October 2, 2006, ending hazardous waste management activities (FDF 1998, 1999; Fluor Fernald 2000, 2001, 2002, 2003a, 2004, 2005, 2006; DOE 2007).

The major Fernald thorium work and regulatory requirements for thorium after the SEC periods (that is, for 1979 to 2006) are summarized below in Table 5-12.

Project	Project Description	Timeframe	Regulatory requirements
1	Overpacking Plant 8 and silo	November 1988–	DOE 5480.11 (DOE 1988)
	thorium material	March 1989	DOE 5480.1A
2	Overpacking 241 containers	January 1990–	DOE 5480.11 (DOE 1988)
	(212 were drums)	May 1990	
3	Overpacking 13,000 containers	1991–2006	DOE 5480.11 (DOE 1988);
			Radiological Control
			Manual (Fluor Fernald
			2003b);
			10 CFR Part 835

Table 5-12. Post-SEC thorium activities and regulatory requirements, 1988 to 2006.

#### 5.3.3.3 <u>Thoron</u>

Thoron was present at Fernald and a portion was released during processing, repackaging, and other tasks, as well as during storage, of the metric tons of a variety of forms of thorium.

Because the amount of time workers spent in a particular location varied in terms of the operating and handling, and because passive storage and remediation categories varied, the determination of working level months (WLMs) per year incorporated occupancy factors for the fraction of time spent by workers in these areas. Attachment B, Analysis of Potential Thoron Exposures, provides additional information on thoron exposures. It does not provide precise estimates because of the lack of site-specific thoron data by time and location, but it provides a reasonably conservative estimate of thoron exposures. The attachment indicates that the nature of storage facilities results in a minimal occupancy time for required functions. The time of occupancy during production periods is assumed to be 1,750 hr/yr, based on an average of a continuous 1-MTTh production rate assumption. At a 1-MTTh daily rate, the recorded annual quantities processed would be completed in just a fraction of a year. For long-term storage of high-integrity storage containers, 1 mo/yr is assumed. For routine storage conditions, including required repackaging, 500 hr/yr (or 3 mo/yr) is assumed.

Doses for thoron potential were separated into four categories: (1) various operating and handling period (2) long-term storage period, (3) passive storage period, and (4) remediation period. The highest WLM per yr values for 1954 to 1985 from Attachment B, Tables B-5 and B-6, were used to provide a bounding thoron dose. Passive storage occurred from 1986 to 1987, when the potential for

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 29 of 188
	,		

no significant thoron dose occurred. Remediation occurred from 1988 to 2006, along with increased radiological controls. The potential for thoron exposure was greatly reduced in comparison with the earlier years at the site, as discussed below.

#### 5.3.3.3.1 Operations and Handling, 1954 to 1971

Thorium processing began in 1954 (ORAUT 2008a), and there were at least potential exposures from levels of thoron during processing and storage of thorium. The thoron exposure potential varied between plant operations and other handling areas, and for some years and areas was insignificant. There were various storage sites that had the highest potential for thoron exposure. Although Fernald did not store large quantities of thorium until 1972 when it became the DOE repository, because work locations are unknown, the maximum WLM per year will be assumed.

#### 5.3.3.3.2 Long-Term DOE Repository Storage, 1972 to 1985

Fernald became the national thorium repository for DOE starting in 1972. An inventory of thorium production orders shows that there were no orders after 1985 and only a few from 1979 to 1985 (Bonfer 1988). It is likely that most or all of these production orders involved only taking stored material out of a warehouse and shipping it to a customer. The occasional thorium repackaging effort (which typically involved placing eroding containers into larger containers) would have been of short duration and would have involved very few individuals.

#### 5.3.3.3.3 Passive Storage, 1986 to 1987

A period of passive storage occurred at the site from 1986 through 1987. This was a time for planning for the disposition and off-site shipping of the stored thorium inventory (Reafsnyder 1987). In 1985, Lockwood Greene was under contract to DOE to perform a conceptual design for environmental, health, and safety improvements for a number of FMPC facilities. Included in this contract was the development of a conceptual design for a thorium handling system to remove the materials from the Plant 8 storage vessels. As part of the design process, Lockwood Greene performed a preliminary structural analysis of the storage vessels. On June 9, 1986, a Thorium Silo Remediation/Thorium Materials Disposition Task Force was convened (WMCO 1986b). Structural remediation of the Plant 8 silo occurred from June 1986 to September 1986 (Cook 1987, p. 7). In September 1987, Fernald began the design and construction of the handling system necessary to remove and package the bulk thorium materials. After construction, the removing, handling, and packaging of thorium began in November 1988 (Aas et al. 1987, p. 451).

#### 5.3.3.3.4 Remediation, 1988 to 2006

The majority of efforts at the site involved remediation and cleanup activities from 1988 through October 29, 2006. In 1988, Fernald initiated a "three Project plan" for improving the temporary storage conditions for thorium (WMCO 1992a, p. 203; Reafsnyder 1987). The removal, handling, and packaging of thorium began in November 1988 (Aas et al. 1987, p. 451). Remediation was completed at Fernald on October 29, 2006 (Stoller 2007, p. 20). By the late 1980s, the respiratory protection program was well established. Those who worked on thorium remediation wore respirators. Even allowing a respiratory protection factor of only 10 is equivalent to reducing the thorium workers' approximate full-time occupancy to about 1 mo/yr occupancy. Therefore, the value for 1988 to 2006 is appropriate for all radiological workers, especially since the WLMs derived in Attachment B took no credit for respiratory protection. Within the remediation period, a two-tiered approach should be taken: (1) radiological workers and (2) nonradiological workers. Nonradiological workers are those who clearly did not work in the process area (cafeteria, accounting, legal staff, etc.) and should not be assigned thoron dose. Radiological workers are those who could have worked in the process area, and are assumed to have the potential for thoron exposure.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 30 of 188
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#### 5.3.4 Q-11, K-65, Domestic Uranium, and Raffinates

A detailed processing history of pitchblende and domestic uranium feedstock, as well as the available monitoring data, can be found in ORAUT-RPRT-0052 (ORAUT 2011a). A brief summary is contained here.

During the Manhattan Project, extremely high-grade uranium ore (50% to 80%) was imported from African Metal Company (AMC) in the Belgian Congo and was designated with the code "Q-11" (Robinson 1973). At this level of uranium content, the ore could be processed directly in the modified PUREX process at the Fernald site. From 1946 until 1953, MCW in St. Louis, Missouri, produced uranium from the Q-11 ore. The Q-11 ore contained all of the uranium progeny, including <sup>226</sup>Ra. When the ore was refined, <sup>226</sup>Ra and other progeny were concentrated in the waste stream, which was identified with the code "K-65." The term raffinate was used to describe the waste stream. The K-65 residues were returned to Belgium until March 1949 (NLO 1979; Heatherton 1949). In 1949, an agreement was reached wherein AEC would store the K-65 residues for later return to AMC. Beginning on March 22, 1949, the K-65 residues from MCW were sent to Lake Ontario Ordnance Works (LOOW) to be stored (Heatherton 1949). Construction of FMPC began in 1951 and MCW began shipping K-65 material to FMPC rather than LOOW (Heatherton 1952; Walden 1952; Vogel 1989).

When construction was completed, storage of all K-65 material was assigned to FMPC and two large ground-level silos were built for this purpose. Most U.S. and Canadian ores were low grade and required processing near the mine sites. The product from these mills was yellowcake ( $U_3O_8$ ) of approximately 60% uranium content. After 1958, when processing of the Q-11 and other pitchblende ores at FMPC had been completed, the source of the uranium for processing at FMPC was this yellowcake. The yellowcake had most of the radium and other impurities removed before being shipped to FMPC. This raffinate was called cold raffinate because the waste stream had much lower concentrations of <sup>226</sup>Ra and progeny in comparison with the K-65 raffinate. The cold raffinate was a calcined dry, dispersible powder that was transferred to Silo 3 using an enclosed airlift (ORAUT 2008b).

The silo waste became the property of the United States in 1983 and was a storage problem. The concrete silos cracked, leaked, and were the object of several upgrade efforts to reduce both the liquid leaks and the release of radon and its progeny. A dirt berm was added around the silos (primarily as silo wall support, but it also provides shielding), as were roof sealants and a semipermeable cap on the waste itself, all of which reduced the routine releases of radon and progeny from the silos.

The operation of handling the large number of drums of K-65 waste materials and dumping them into the silos in the period from July 1953 to September 1958 involved external and internal exposure to the radium and other associated contaminants. In addition, there were several operations to improve the confinement of the silos for the radon gases and associated progeny, although the waste materials have not been directly disturbed since they were put into storage. Air sample data sheets from around1953 provide insight into the operation and exposure levels. Bioassay information about internal exposures to radium or the associated contaminants consists of over 600 radon breath samples. In vivo counts were periodically performed by the Y-12 mobile counting laboratory, but those did not begin until 1968, well after completion of the handling and dumping of the drums of waste.

The isotopic contents of the three silos are shown in Table 5-13.

Silos 1 and 2 contained the K-65 (hot) raffinates and included relatively high concentrations of <sup>226</sup>Ra. Silo 3 contained cold raffinate with very little <sup>226</sup>Ra. Because uranium was the extraction product, the

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 31 of 188
			5

Isotope	Silo 1 activity concentration (nCi/g raffinate)	Silo 1 activity (%)	Silo 2 activity concentration (nCi/g raffinate)	Silo2 activity (%)	Silo 3 activity concentration (nCi/g raffinate)	Silo 3 activity (%)
Uranium	1.68	0.2	2.37	0.3	3.63	4.8
Ac-227	7.67	0.8	6.64	0.8	0.93	1.2
Pa-231	Not applicable	0.0	4.04	0.5	0.63	0.8
Pb-210	202	20.0	190	24.3	3.48	4.6
Po-210	281	27.8	231	29.6	Not applicable	0.0
Ra-224	Not applicable	0.0	Not applicable	0.0	0.37	0.5
Ra-226	477	44.2	263	33.6	3.87	5.2
Ra-228	2.28	0.2	Not applicable	0.0	0.41	0.5
Th-228	Not applicable	0.0	7.36	0.9	0.75	1.0
Th-230	68.9	6.8	76.2	9.7	60.2	80.1
Th-232	1.11	0.1	0.99	0.1	0.84	1.1
Total	1,041.64	Not	781.6	Not	75.11	Not
		applicable		applicable		applicable

Table 5-13. Isotopic composition of K-65 silos 1, 2, and 3 (from Table 1 of ORAUT-RPRT-0052 (ORAUT 2011a)).

raffinates in the silos contained low levels of uranium. Thorium-230 was present in each silo in similar concentrations, indicating that the uranium mills effectively removed radium but were not effective in removing thorium from the yellowcake. The total activity concentration in silo 3 was approximately 10% of the activity concentration in silos 1 and 2. However, <sup>230</sup>Th is a dominant radionuclide in Silo 3 and requires consideration for dose evaluation purposes. The concentration of <sup>230</sup>Th is essentially the same in all three silos. The <sup>230</sup>Th in Silo 3 constitutes a high fraction of the total activity in comparison with the fractions in silos 1 and 2, due not to high <sup>230</sup>Th levels, but due to the absence of radium and its progeny. If the <sup>226</sup>Ra and progeny were removed from silos 1 and 2, the <sup>230</sup>Th would constitute about 80% of the remaining relative hazard, as it does in the Silo 3 material.

RAC (1998) provides estimates of the annual emission of <sup>222</sup>Rn from the K-65 silos for the period from 1952 through 1988. The 95th-percentile value of these estimates, which defines upper estimates of emission rates, has been used in the analysis to calculate the annual environmental intake rates shown in Table 4-2 in ORAUT-TKBS-0017-4, *Feed Materials Production Center – Environmental Dose* (ORAUT 2015b).

## 5.3.4.1 Radon Breath Analysis

During 1952 through 1954, to infer radium intakes, 609 breath samples were taken for radon at the site. The samples were sent for analysis to the AEC Health and Safety Laboratory (HASL) in New York City. At this time, no specific information is available about the measurement method; the assumption is that the ambient radon concentration present during the sampling was included in the result. Therefore, any dose reconstruction based on these uncorrected results would include a bias favorable to the claimant.

A typical radon breath analysis result identifies the individual and provides the radon breath concentration measurement results in curies per liter. Annual and composite parametric values for the data are shown in Table 5-14. In all years, the minimum reported value is  $1 \times 10^{-13}$  Ci/L, which is assumed to be the reporting limit for the analysis. When the reported result is "< $1 \times 10^{-13}$  Ci/L" a value half of that ( $5 \times 10^{-14}$  Ci/L) was used in calculations. This method is endorsed in NIOSH (1993), which is incorporated by reference in the implementing rules for EEOICPA [42 CFR Part 82.16(a)]. Approximately 25% of the values were imputed by this method. In addition, approximately 26% of the submitted samples were not successfully analyzed due to processing errors or equipment failures.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 32 of 188
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Descenter	4050	4050	4054	Composite 1952
Parameter	1952	1953	1954	through 1954
Number of attempted measurements	140	238	231	609
N, number of valid measurements	84	183	182	449
Number of valid measurements less than the	55	54	46	155
reporting limit				
Minimum, Ci/L (assumed to be the reporting limit)	5.00E-14	5.00E-14	5.00E-14	5.00E-14
Maximum, Ci/L	9.00E-13	3.30E-12	1.20E-12	3.30E-12
Mean, Ci/L	2.66E-13	2.84E-13	2.47E-13	2.66E-13
Standard deviation	2.01E-13	3.26E-13	1.79E–13	2.53E-13
Coefficient of variation (standard deviation ÷ mean)	0.76	1.14	0.72	0.95
GM, Ci/L	2.01E-13	2.07E-13	1.91E–13	1.99E–13
GSD	2.18	2.13	2.11	2.13
Median, Ci/L	2.00E-13	2.00E-13	2.00E-13	2.00E-13
84th percentile, Ci/L	4.00E-13	4.00E-13	4.00E-13	4.00E-13
95th percentile, Ci/L	6.85E-13	6.90E-13	6.00E-13	6.00E-13

Table 5-14.	Radon breath	analysis	statistical	parameters.
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Lacking data, these were removed from consideration, resulting in a total of 449 valid samples over the period from 1952 through 1954.

ORAUT-OTIB-0025, *Estimation of Radium-226 Activity in the Body from Breath Radon-222 Measurements*, provides information on partitioning of the whole-body radium activity among bone, lung, and soft tissue compartments (ORAUT 2005a). The radiation dose to various organs from <sup>226</sup>Ra in the body can be derived by selecting an appropriate intake scenario and applying appropriate dose factors to the derived <sup>226</sup>Ra activity level. ORAUT-OTIB-0025 defines a whole-body <sup>226</sup>Ra activity conversion factor for radon breath data as 2.52 × 10<sup>5</sup> pCi of <sup>226</sup>Ra per pCi/L of exhaled <sup>222</sup>Rn.

The median whole-body estimate developed from the radon breath analysis is 0.05  $\mu$ Ci of <sup>226</sup>Ra (assuming a 1-year chronic inhalation).

Fernald Plant 2/3 workers were also exposed to pitchblende ores and their raffinates from about 1954 through 1958 (Q-11 through Australian pitchblende). However, processing was done in the Fernald plant with the resultant raffinates going directly to the K-65 silos. The raffinate handling and transfer operation in Plant 2/3 was of a slurry form, and the raffinates were transferred in process lines directly to the silos. This operation was quite different from the drum handling described above. The bounding process is taken to be the opening of the drums in Plant 2. At this point in the process, the uranium, its progeny, and the associated contaminants are all present. Therefore, uranium bioassay was adequate and the other components contributing to the dose can be assigned by using ratios to uranium.

After the processing of the pitchblende ores ended in 1958, Plant 2/3 workers processed only ore concentrates (yellowcake) from ore processing plants (uranium "mills") with the resulting "cold" raffinates. These were transferred in calcined, dry, dispersible powder form to Silo 3, using an enclosed air lift. Again, the bounding process is taken to be the opening of the drums in Plant 2. At this point in the process, the uranium, its progeny, and the associated contaminants are all present. Therefore, uranium bioassay was adequate and the other components contributing to the dose can be assigned by using ratios to uranium.

#### 5.3.4.2 Raffinate Exposures

#### 5.3.4.2.1 K-65 Raffinate Drum Transfer Workers, 1952 to 1956

Radon breath analysis results exist for some workers at Fernald (NLO 1952, 1953a). Radon breath analysis results, titled "Personnel Radon Breath Analysis Measurement Results," are evaluated and included in the dose reconstruction as follows:

- 1. Convert the radon breath analysis result to picocuries per liter.
- 2. Apply the ORAUT-OTIB-0025 conversion factor for the whole body of 2.52 × 10<sup>5</sup> pCi/(pCi/L) to get a whole-body content of <sup>226</sup>Ra in picocuries (ORAUT 2005a).
- 3. Use the whole-body <sup>226</sup>Ra content in IMBA to derive a chronic intake of type M <sup>226</sup>Ra from January 1, 1952, through the date of the last radon breath analysis result.
- 4. In addition to the <sup>226</sup>Ra intake estimate, apply the derived intake to the isotopes and ratios in Table 5-16.

If there are no radon breath analysis results, dose reconstructors should use the <sup>226</sup>Ra default intake rate from Table 5-15. This is based on the assumption of the median whole-body estimate developed from the radon breath analysis (0.05  $\mu$ Ci of <sup>226</sup>Ra) and assuming a 1-year chronic inhalation intake. The intake rate of <sup>226</sup>Ra is then used with activity ratios of <sup>226</sup>Ra to other raffinate nuclides to assign intakes of the other radionuclides in Table 5-16.

Table 5-15. Radium intake rate based of	on radon breath anal	ysis data (pCi/d).
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Years	Ra-226 intake estimate <sup>a</sup>	IREP <sup>b</sup> distribution
1952–1954	5,184	Lognormal GSD=3

- a. Ra-226 is solubility type M.
- b. Interactive RadioEpidemiological Program.

	Activity ratio of nuclide to	Activity ratio of nuclide to
Isotope	Ra-226 in silo 1 material	Ra-226 in silo 2 material
Ac-227	0.016	0.025
Pa-231	Not applicable	0.015
Pb-210	0.423	0.722
Po-210	0.589	0.878
Ra-224	Not applicable	Not applicable
Ra-228	0.005	Not applicable
Th-228	Not applicable	0.028
Th-230	0.144	0.290
Th-232	0.002	0.004

Table 5-16. Ratios of silos 1 and 2 nuclides to <sup>226</sup>Ra.

Table 5-16 shows the ratio of the isotopes in the silo 1 and 2 materials to <sup>226</sup>Ra. These ratios are based on data in Table 5-13. The ratios from both silos 1 and 2 are similar, but the ratios to the radium isotopes from the silo 1 and 2 data should be compared for dose reconstruction, and the most favorable to the claimant should be used to assign dose because it is not possible to know which material was included with the radium intake. It is not necessary to consider the ratio from silo 3, since the processing of the yellowcake (slurry) did not begin in Silo 3 until 1959 (ORAUT 2011a).

These estimates establish an upper bound of intake for workers involved in the transfer operation of the 13,000 barrels of the stored MCW raffinates to the K-65 silos. An examination of external penetrating radiation dose for workers who were known to have worked with and handled these

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 34 of 188

drums of raffinate wastes show significant penetrating dose accumulation (several hundreds of millirem per week). Therefore, a criterion to determine or verify that a worker had indeed been exposed to internal intake from raffinate dusts would be a record of penetrating external dose (i.e., no detectable dose would clearly indicate little direct contact or work with the barrels of waste). However, the assumption that the population of K-65 raffinate workers could be limited by external dose because the high <sup>226</sup>Ra content would have produced high dose rates did not prove to be true [1]. Because a method for identifying workers on this project could not be identified, all workers should be assigned an intake.

Workers opening drums of the K-65 raffinates would have also been exposed to the radon escaping from the drums. Natural ventilation outdoors would have decreased the concentrations in a few minutes. No radon beyond the environmental doses need be assigned.

#### 5.3.4.2.2 Plant 2/3 Workers Handling or Processing Pitchblende Ores, 1954 to 1958

Dose reconstructors should assign uranium intakes from uranium urine bioassay. Unmonitored exposure to the impurities in pitchblende might have occurred to workers who sampled or handled these materials. Assume that the <sup>230</sup>Th in silos 1, 2, and 3 is in equilibrium with the total uranium activity and assign an intake of <sup>230</sup>Th that is equivalent to the derived uranium intake.

In addition, use the ratio to <sup>230</sup>Th activity of the isotopes in silos 1 and 2 materials from Table 5-17. These ratios are based on the activity concentration of raffinate data in Table 5-13. The ratios from the silo 1 and silo 2 data should be compared for dose reconstruction, and the most favorable to the claimant used to assign dose, if it is unknown to which silo a worker was exposed. Because the activity ratios in Silo 3 are lower (and not as favorable to the claimant), a comparison to Silo 3 is not necessary. For dose reconstructions with overlapping periods in 1955-1956, a comparison between Table 5-16 and Table 5-17 doses are made, and the most favorable to the claimant should be selected for assigning dose.

	Activity ratio of nuclide to	Activity ratio of nuclide to	Activity ratio of nuclide to
Isotope	Th-230 in silo 1 material	Th-230 in silo 2 material	Th-230 in silo 3 material
Ac-227	0.111	0.087	0.015
Pa-231	Not applicable	0.053	0.010
Pb-210	2.932	2.493	0.058
Po-210	4.078	3.031	Not applicable
Ra-224	Not applicable	Not applicable	0.006
Ra-226	6.923	3.451	0.064
Ra-228	0.033	Not applicable	0.007
Th-228	Not applicable	0.097	0.012
Th-232	0.016	0.013	0.014

Table 5-17. Ratios of silos 1, 2, and 3 nuclides to <sup>230</sup>Th.

#### 5.3.4.2.3 Plant 1, 2, 3, and 8 Workers Handling or Processing Yellowcake, 1959 to 1961

Unmonitored exposure to the impurities in yellowcake might have occurred to workers who sampled or handled these materials. Workers in Plants 1, 2, 3, and 8 might have been exposed. The intake rates from these exposures can be bounded by adding an intake from these impurities in proportion to the uranium intake as measured by bioassay. The ratio from silo 3 is used for dose reconstruction because the processing of the pitchblende ores (whose residues were stored in Silos 1 and 2) ended in 1958 (ORAUT 2011a).

Assume that the <sup>230</sup>Th in silo 3 is in equilibrium with the calculated uranium activity and ratio the rest of the isotopes to <sup>230</sup>Th, as given in Table 5-17. These ratios are based on the activity concentration of raffinate data in Table 5-13. Beginning in 1961, RU contaminants must also be included.

Air monitoring results show that no significant exposure to radon from cold raffinates occurred. The milling process removed nearly all of the radium from the ore concentrates. Therefore, the concentrate materials presented no significant hazard from penetrating radiation or from the accumulation of radon gas during drum opening and sampling operations (Christofano and Harris 1960).

### 5.4 RADIOLOGICAL CONTROLS PROGRAM

The initial health and safety organization (the Industrial Hygiene and Radiation Division) at Fernald was organized and directed by an occupational medical physician and staffed primarily with industrial hygienists. The Fernald staff worked with the Y-12 Plant staff because of similar radiation safety issues associated with their uranium processing, although Y-12 typically had a more highly enriched uranium source term. Fernald also used Y-12's Mobile In Vivo Radiation Monitoring Laboratory (MIVRML) for over 20 years. A review of the Y-12 radiation protection program and internal dosimetry technology (Cofield 1959; Scott and West 1964; Steckel and West 1966; West, Scott, and Schultz 1979) indicated that the internal dose technology, techniques, procedures, and philosophy similar to Y-12's were used at Fernald. The fact that the Y-12 MIVRML provided routine service to the Fernald in vivo internal dosimetry program is an indicator of the close working relationship the Fernald staff had with the Y-12 program.

The safety organization's staff was aware that TRU elements and fission product contaminants were present in the RU. The biological concerns were thought to represent an increase of less than 10% in hazard level in comparison with uranium that had never been irradiated. Radiological controls were determined and adjusted based on routine air sample analysis and urine bioassay for uranium.

Before 1986, routine TRU analyses were not performed for either airborne or urine activity, although a few examples of specific analyses of plutonium on air samples were recorded and indicated that the uranium concentrations were controlling. Exposure controls were set at levels that prevented chemical toxicity of uranium, assuming that these controls were sufficient for all the radiological hazards (Bassett et al. 1989). Although the alpha activity from the TRU alpha emitters would have been collected and detected in the air samples, the reported results were all considered to be primarily uranium and compared to the MAC for uranium (Wing and Halcomb 1959a, 1959b). As a consequence, bioassay measurements were not routinely performed on workers for the contaminants associated with RU until 1986. Heatherton (1975, p. 159) includes a report that discusses the results of the FMPC uranium monitoring program.

Radioactive contamination was routinely measured in occupied areas of the plant, and there were significant radioactive material releases to the environment. It was necessary for every facility at the Fernald site to address issues involving airborne contamination, even though engineered confinement barriers were used in conjunction with process and work area ventilation. Elevated airborne radioactivity resulted from processing thousands of metric tons of dispersible radioactive materials with a variety of chemical and physical characteristics. Throughout the process history of the site there were high potentials for intakes of uranium, thorium, and their radioactive progeny.

A radiological controls program was in place from the beginning of FMPC operations. The internal dose control program consisted of:

- An air sampling program in all processing areas to evaluate internal exposure potential via inhalation,
- Urine samples submitted after at least a 2-day work break to allow elimination of uranium cleared rapidly via the gastrointestinal tract (this material causes relatively little dose), and

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 36 of 188

• In vivo analysis once a month for workers with high exposure potential on a frequent urinalysis program and once a year for workers with a low potential for internal intake.

Other elements of the protection program included routine monitoring of the workplace and personnel for radiation (NLO 1953b) and contamination, personnel protection in the form of protective clothing and respiratory protection in all of the operational areas as needed, and restricting workers from workplaces with elevated airborne radioactivity concentrations when the level of uranium in the urine or in vivo counting results exceeded specified plant action limits.

#### 5.4.1 Air Monitoring Program

The large quantity of data in the archives shows that Fernald maintained an aggressive air monitoring program from the beginning of operations (AEC 1955, Halcomb and Huesing 1957). Both high and [primarily] low volume GA and BZ air samples were collected (most for 3 to 30 minutes) and counted for alpha contamination in the 1950s. A few sample records and claim file records indicate that some beta counts were performed. In the 1960s the samples were counted for both alpha and beta activity. The results were compared to the NLO MAC guidelines (a.k.a. the NCG) of 100 dpm/m<sup>3</sup> [70 dpm/m<sup>3</sup> was used as the MAC (NCG) until the 1970s]. The 100 dpm/m<sup>3</sup> equates to about 4.5 x 10<sup>-11</sup> µCi/cm<sup>3</sup>.

Routine air samples were taken in every plant and operational area. This program was the primary means of controlling intakes. Workers were directed to use respiratory protection in the form of dust masks or supplied air respirators depending on the anticipated or measured airborne radioactivity concentration. From a historical viewpoint, extensive, long-term air activity summary sheets that covered 15 or more years were periodically prepared that indicated routine detectable air activity in all working areas of each plant. These summaries detailed annual average exposures to workers without respiratory protection and average air activities associated with job assignments that required respirators. The air activity ranged from a fraction of the MAC levels to hundreds of times those levels.

From 1953 to 1986, the air monitoring program was used primarily to establish work controls, such as respiratory protection requirements for workers. However, the measured air concentration levels from the routine sampling program typically were not used to establish worker intakes, and workers were required to submit routine urine samples for uranium analysis only for the purpose of verification of site controls based on the air sample. The MDL for routine air sampling was in the 0.02 to 0.2 MAC range. When multiplied by an 8-hour workday, an MDL of approximately 1.5 MAC-hr/d is derived (based on informal calculations conducted from data taken from recorded sample sheet volume and radiation counting data). The most typical sample volume was a 3-minute sample (0.06 m<sup>3</sup>) in dusty areas, which equates to the 1.6 MAC-hr MDL, and up to a 30-minute sample (0.6 m<sup>3</sup>) with an MDL of 0.2 MAC-hr. BZ samples were generally of the same flow rate but shorter duration. Long-term averages at 0.1-MAC levels were reported, but should be interpreted as numerical averages only.

Beginning in 1989, BZ samples were converted to DAC-hours in the case of thorium, uranium, and custom nuclide exposures. On occasion, these air sample analyses were converted to intake, dose, or both. The calculated doses observed in the records were low [i.e., tens of millirem committed effective dose equivalent (CEDE)]. The air sample results are used with the isotope and a conversion to a percentage of the DAC, then converted directly to dose in CEDE.

For reference, Table 5-18 lists the abbreviations and codes on the sample and dose calculation datasheets.
Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 37 of 188
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Code	Description
MAC	NLO MAC (NCG) –100 dpm/m <sup>3</sup> and 70 dpm/m <sup>3</sup> before 1970
NCG	NLO concentration guide–used interchangeably with MAC
R	Sample collection rate in m <sup>3</sup> /min
Т	Sample time in minutes
Q	Sample volume in $m^3 Q = R \times T$
GA	Sample collected in a GA
BZ	Sample collected as close to the BZ as practicable
Analyzed for–alpha	Gross alpha count
Analyzed for-beta	Gross beta count
Analyzed for–Ra	Alpha count on radium sample separation
Analyzed for–Th (#33)	Alpha count on thorium sample separation
Analyzed for-all others	Chemical analysis for nonradiological samples

Table 5-18. Air monitoring analysis sheet codes.

Later (1993 to 2006) air monitoring was performed with the intent of calculating intake and dose if necessary for low dose cases in addition to maintaining site control of personnel exposures. The air sample data were used to calculate a specific concentration of a specific nuclide in terms of microcuries per cubic centimeter and conversion to a specific DAC percentage. This value was directly converted to intake and organ dose, as appropriate.

Table 5-19 cross-references Fernald air monitoring methods with the applicable MDLs.

Type of				
analysis	Method	Period	Frequency	MDL <sup>a,c</sup>
Air sampling	BZ sampling	1952–1993	Variable and frequent,	0.02–0.2 MAC depending on
			depending on work	sample volume
Air sampling	BZ lapel	1993–2006	Job specific, when Th-230 or	1–2 DAC-hr with a minimum
	samplers		Th-232 exposure potential	detectable intake <sup>b</sup> of 0.001–
			exists. BZ sampling for	0.002 corrected DAC-hr for
			uranium is also performed for	thorium
			work controls.	

Table 5-19. Air monitoring methods with applicable MDLs.

a. From 1952 to 1993, BZ samples were taken for 5 to 30 minutes at 20 L/min (0.1 to 0.6 m<sup>3</sup> of air); after 1993, the lapel sampler was typically worn continuously at about 4 L/min for 8 hr/d (approximately 2 m<sup>3</sup> of air collected), then counted for longer times with more sensitive laboratory counters.

b. Detectable Intake is lower due to the application of a respiratory protection factor.

c. MDLs reported in worker's records take precedence over the default values in this table.

Fernald's HIS-20 database was implemented at the site in the mid-1990s and is the source of BZ air monitoring data. Some of the data in HIS-20 were migrated from legacy health and safety databases. DOE has also provided data extracted from HIS-20 to NIOSH in the form of database tables. Thorium BZ results begin in 1993 in the HIS-20 database. Beginning in 1995 (HIS-20 database), BZ air monitoring data are consistently available.

# 5.4.2 Bioassay Program

A uranium urinalysis program was administered at Fernald starting in 1953 or possibly earlier. Uranium is an unusual radioactive material in that its chemical toxicity can be the dominant hazard in cases of readily or moderately soluble compounds of depleted uranium (DU), NU, and LEU. Uranium is a heavy metal and a sufficient heavy metal exposure can lead to kidney damage.

In 1959, a chemical toxicity threshold concentration of 3 µg U/g of kidney tissue (based on animal studies and some human data) was established by the International Commission on Radiation Protection (ICRP), and has been used since that time as the reference limit. Concentrations in

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 38 of 188
--------------------------------	-----------------	----------------------------	----------------

excess of 3  $\mu$ g U/g of kidney tissue have been shown to result in renal tubule damage. The no-effect level of uranium in the kidney has been calculated to be 1.1  $\mu$ g U/g of kidney tissue, which averages to a kidney burden of 0.337 mg (Rich et al. 1988). Table 5-20 lists a summary of the Fernald chemical toxicity limits with their associated radiological limits.

Solubility and enrichment	OSHA limit for toxicity (mg/m³ )	OSHA limit for equivalent radiation exposure (µCi/cm <sup>3</sup> )	Limit based on radiation exposure (µCi/cm³)
Soluble DU	0.05	1.8E–11	6.0E–10
Soluble NU	0.05	3.5E–11	3.0E–10
Soluble 2% EU <sup>b</sup>	0.05	8.1E–11	2.0E–11
Insoluble DU	0.25	9.2E–11	6.0E–10
Insoluble NU	0.25	1.8E–10	3.0E–10
Insoluble 2% EU <sup>b</sup>	0.25	1.8E–10	3.0E-10

Table 5-20. Chemical toxicity and associated limits.<sup>a,b</sup>

a. A 1-milligram intake is approximately 0.337 of the kidney toxicity limit. The intakes are assumed to be 7 mg for type F, 37 mg for type M, and 696 mg for type S.

b. Italicized entries, and the type S intake of 696 mg, indicate limits that are controlled by radiological concerns.

The early basis for conducting routine urine analysis was to ensure that uranium exposure controls were adequate to prevent chemical toxicity. In addition, the urinalysis results were used to restrict further worker exposure when control levels were exceeded. Urine samples were taken as frequently as weekly (or more frequently as indicated by unusual events). Internal radiation doses, which were derived from urine sample results, were not calculated until the mid to late 1980s.

Although the primary exposure control during the early years of operation was based on toxicity concerns, some radiological exposures were reported in addition to a few work restrictions based on radiological dose limits.

Nearly all employees provided urine samples for uranium analysis at the time of their annual physicals. Workers with work assignments such that exposure could be expected on a routine basis were sampled weekly, monthly, or (at the least) bimonthly. The frequency for routine sampling for operations personnel changed in the early years from quarterly to a variable frequency depending on previous urinary uranium results, knowledge of plant operations, and possible exposure. This frequency might have varied from 3 times per year to once per month for any particular worker. Routine samples were collected at the beginning of the shift, but not necessarily at the beginning of the workweek. In the early years, any routine result that exceeded 40  $\mu$ g/L of uranium was followed up by obtaining start-of-shift urine samples for 3 successive workdays. If one of these results were greater than 40  $\mu$ g/L, the individual's dose was investigated for cause (Heatherton 1975, p. 160)

As early as 1958, FMPC reported internal dose exposure to the AEC in an annual report. Table 5-21 summarizes the data from 1958 to 1966. The data provide indication of recorded annual urinary results in summary form.

The value of fecal samples was recognized even in the early years and has been well understood since 1986. As an example, fecal sampling for uranium was performed on several workers in 1968 as a part of a DOE HASL-NLO study (as recorded on analytical data sheets of October 4, 1968). However, because it was recognized that the natural environmental levels vary considerably, fecal samples were determined to be less reliable so were not a part of the routine bioassay program at the Fernald site. Fecal samples were collected in special cases and beginning in the mid-1990s, they were analyzed under contract with a specified maximum MDA of 0.1 pCi/sample. If lower sample MDAs are found in records associated with contracted fecal uranium analyses, these lower MDAs should be used.

Document No. ORAUT-TKBS-0017-	Revision No. 01	Effective Date: 07/22/2016	Page 39 of 188
-------------------------------	-----------------	----------------------------	----------------

Urine average		Т	otal numb	per of exp	osed work	ers in eac	h categor	У <sup>ь</sup>	
(µg/L) <sup>a</sup>	1958	1959	1960	1961	1962	1963	1964	1965	1966
25–30	22	51	125	75	25	26	18	5	3
31–35	18	37	44	41	5	9	9	1	1
36–40	17	21	25	18	5	1	6	0	0
41–45	9	5	15	6	1	1	0	0	0
46–50	7	4	6	3	1	0	0	1	0
51–55	5	3	7	1	0	0	0	0	0
56–60	2	1	3	1	0	0	0	0	0
61–70	3	4	4	0	0	0	0	0	0
71–90	3	5	0	0	0	0	0	0	0
91–110	3	3	0	0	0	0	0	0	0
111–155	45	2	0	0	0	0	0	0	0
Total	94	136	229	145	37	37	33	7	4

Table 5-21. Urinary uranium averages summary.

a. Various annual reports reported the units in mg/L, which is an obvious typographical error. The permissible urine concentration, averaged throughout the year, was 0.05 mg/L, (which is equivalent to 50 µg/L).

In vivo lung counts were routine from 1968 to 1989 using a MIVRML from the Y-12 Plant in Oak Ridge, Tennessee. Typical results of the MIVRML were uranium lung burdens, although a few thorium analyses results were reported at specific times. The results were reported in micrograms of <sup>235</sup>U and milligrams of total uranium (mg U) and were recorded in individual dosimetry files and summarized in formal reports. TRU analyses were attempted; however, the limitations of the MIVRML, which included limited detection sensitivities for TRU isotopes and infrequent counts, and lacked the ability to detect the anticipated levels at Fernald or in fact could not detect levels that met regulatory limits for TRU isotopes.

From 1989 to 2002, the lung counts took place in the Fernald In Vivo Examination Center (IVEC) (Waligora 2003). Results from the IVEC were reported in nCi if a nuclide was detected.

# 5.4.3 Uranium Urinalysis Program

The urinalysis procedure in place until 1993 was conducted using the fluorometric fusion process, which fused uranium from raw urine with sodium fluoride and measured the fluorescence created by ultraviolet light. This method provided a measure of the total amount of elemental uranium in the sample. Comparison studies between the other laboratories provided assurance that the analytical process was consistent with industry standards. The MDA was not formally established in the early periods (as evidenced by a records search). The values listed in the bioassay data sheets generally range from 0.003 mg/L to 0.008 mg/L in urine. The effective MDA is now understood to have been near 0.014 mg/L.

In early 1993, other DOE laboratories were changing to kinetic phosphorescence analysis (KPA), with detection capabilities as low as 0.02  $\mu$ g/L, which is far below the natural environmental background in most people. The MDA of 0.17  $\mu$ g/L was established consistent with other laboratories. A decision level (DL) of 0.8  $\mu$ g/L at Fernald since 1993 represents the best estimate of the nonoccupational excretion of uranium, although values below the DL are reported in the dose history records. Specifically, when Fernald changed to the KPA, 0.8  $\mu$ g/L became the reporting level until 1997, after which all calculated results were reported as derived. Some database printouts for samples after 1997 include less-than symbols (<) for results that are less than 0.17  $\mu$ g/L (e.g., <0.06  $\mu$ g U/L). These results should be interpreted as less than 0.17  $\mu$ g/L. This represents a database formatting issue rather than a lowering of detection limits.

The method used at Fernald from 2002 to 2006 for urinalysis was inductively coupled plasma mass spectrometry (ICP-MS), which has an a priori MDA of 0.15  $\mu$ g/L.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 40 of 188
--------------------------------	-----------------	----------------------------	----------------

Table 5-22 lists the MDLs, which remained essentially unchanged throughout the history of the site.

Type and method of analysis	Period	Frequency	MDL
Urine, uranium,	1952–1993	Weekly to annual-	14 μg/L <sup>(a)</sup>
fluorophotometry		job specific	
Urine, uranium, Chemchek KPA	1993-09/2002	Bimonthly	0.17 μg/L (total U)
Urine, uranium, ICP-MS	09/2002–2006	As requested	0.15 μg/L (total U)
Feces, <sup>b</sup> fluorophotometry	Various	No routine	Unknown; assume environmental
		schedule	levels of 2 µg/sample
Feces, <sup>b</sup> contract laboratory	Occasional	No routine	0.1 pCi/sample
		schedule	

Table 5-22. Routine uranium in vitro bioassay capabilities.

a. Y-12 listed a sensitivity of 1.6 µg/L in 1973 using the fluorometric process for 0.7% U-235. Fernald frequently listed less than 0.003 mg/L in the bioassay data reports. Several blank samples on intercomparison studies also list results as 0.003 mg/L. A value of 0.008 mg/L has also been quoted in the records as the MDL. However, a formal response on January 21, 1993 (Blalock and Neton 1993), to a deficiency in the ability to detect 100 mrem CEDE with the existing 0.014-mg/L MDA is accepted as the most reliable representation for historical MDAs for this analytical procedure.

b. Fecal sampling was performed as a part of special studies or specific incident investigations on occasion. Records of this activity are generally not present in the dosimetry records for individuals.

To be favorable to the claimant, intakes of uranium due to environmental background are not considered. That is, any detectable uranium is assumed to be due to occupational intakes. All uranium bioassay results (whether urine or fecal) that are greater than the MDA are assumed to be indicative of occupational exposure, regardless of any larger value that the site might have used.

Table 5-23 lists the uranium urinalysis sample type codes, and Table 5-24 lists sample type coding system and personnel assignment codes.

Note that Code 30 urine samples are believed to have been spot samples rather than 24-hour samples. All uranium bioassay samples should be considered to be spot samples, unless there is something in the record to indicate otherwise. Because the results are almost always reported in micrograms per liter rather than micrograms per sample, this will typically not make a difference when evaluating the results. However, this needs to be considered for incidents and for days when there are multiple results.

# 5.4.4 In Vitro Bioassay Procedures for Other Radionuclides

The fundamental and primary bioassay for the first 35 years (1951 through 1986) of Fernald operational experience was urine analysis for uranium metal, reported in milligrams per liter. Radionuclides other than uranium have been analyzed on occasion through the years, predominantly by contract laboratories. Even for those special cases, they have been so few in number that the review of records for this TBD did not reveal a dose record with nonuranium urinalysis results. There are records of special studies, but no documented intent to analyze for radionuclides other than uranium. The primary contract laboratory for FMPC in vitro analyses was United States Testing Company in Richland, Washington. Table 5-25 lists MDAs for various nonuranium radionuclides.

# 5.4.5 Gross Beta Urine Sample Results, 1957

In 1957, there appeared to be a concern for workers in Plant 5 who were suspected of unusual intakes due to the presence of recorded gross beta urine results. These urine samples were analyzed for uranium, and an aliquot from the sample was then counted for gross beta. An expressed concern was noted in a 1957 monthly report of "unusual uranium intakes." FMPC likely counted an aliquot for beta due to their sensitivity for "other" isotopes, although it is doubtful that the concern was specifically for thorium – perhaps a concern for fission products, considering that Plant 5 had not been involved in thorium processing, as far as is recorded.

Table 5-25. Fernalu uranium unnaiysis sample type codes	Table 5-23.	Fernald	uranium	urinalysis	sample t	ype codes
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Code	Description
00	No Code
10	Pre-Employment Sample – collected during pre-employment physical exam or before beginning work on first day. Establishes individual U background
20	Annual Sample – collected as a part of each employee's annual physical exam
30	Routine Sample – samples from plant workers who are on a routine schedule for the purpose of insuring that airborne levels of uranium in the work place are being controlled within safe limits
40	Incident – Follow-up Sample – samples from employees involved in an event or circumstances which presents a potential for elevated exposure
49	An incident sample left at the end of the shift on the day of the incident.
50	Special Sample – samples collected as a part of a study to provide data related to uranium exposure and/or excretion characteristics of specific areas and/or conditions. Also includes samples taken from subcontractors. In general, results for subcontractors are not in the HIS-20 database before 1986.
5C	Special Correlation Sample
60	Termination/Retirement Sample – sample obtained from employee during post-employment physical exam.
70	Rehire – sample obtained during a former employee's physical exam before being re-employed
R	Recall Sample (example: 3R – Routine Sample Recall) – samples taken to verify positive sample results and/or to follow elimination pattern
VF	Visitor First Sample
VR	Visitor Routine Sample
VE	Visitor Exit Sample
BDL	Below Detection Level
NA	Not applicable/available

These beta counts are recorded as "gross" beta, which could contain <sup>40</sup>K and a variety of other "natural" contaminants (because they were in the height of the atmospheric weapons testing fallout, the generation of which started to decline in 1962 to 1963). Claimant files typically show that those who worked beyond 1968 had routine in vivo lung counts with reported thorium levels. Therefore, the gross beta counts are not well-enough defined to be of value. There are hundreds of uranium urine analyses, a defined RU contaminant default approach, thorium lung counts, and a defined intake analyses based on DWE air sampling analyses before 1968. This appears to be a justifiable basis for ignoring the 2 to 4 gross beta counts, which resulted from a single occurrence in Plant 5 [2].

## 5.4.6 In Vivo Bioassay

Lung counting became available to FMPC in 1968 in the form of the MIVRML. The mobile van visited the Fernald plant on a routine schedule and counted the workers on a schedule based on their internal exposure potential and their urine sampling results. When lung counting became available, the annual reports to AEC listed the number of workers who exceeded 50% of the maximum permissible lung burden (MPLB) and the calculated annual doses to the lung in rem. The uranium MPLB was 0.0175  $\mu$ Ci, assuming 1% enrichment. Table 5-26 presents the typical reporting of internal dose as determined by direct lung counting.

Table 5-24. Fernald sample type coding system and personnel assignment codes.

Code	Definition
1	During 1st hour of shift
2	During 2nd hour of shift
2	During 3rd hour of shift
3	During 5td hour of shift
4	During 5th hour of shift
5	During 6th hour of shift
7	During 7th hour of shift
0	During 8th hour of shift
0	After completing work
9	Somple type poding system _ further definition
Codo	Sample type could system – further definition
5 A	Definition
5D	Off the job, overnight individual encoimen
	On the job, overnight individual specifien
50	Sample to test for possible correlation with abnormal clinical lab
<i>5</i> D	Informers 24 hour individual completions confined actionts
50	24-hour individual sample from confined patients
55	24-nour composite sample from contined patients
5F	24-nour individual sample from unconfined patients
5G	24-nour composite sample from unconfined patients
5H	On-the-job individual sample collected in the work area
0.1	Personnel assignment codes
Lode	Definition
150	Plant 6 Inspection
200	General Project (Plant 8)
210	Plant 1
220	Plant 2 and 3
240	Plant 4
250	Plant 5 and 9
261	Plant 6 Rolling Mill
262	Plant 6 and 9
2623	Plant 6 and 9 Chemical
2625	
070	Plant 6 and 9 Machining
270	Plant 6 and 9 Machining Pilot Plant
270	Plant 6 and 9 Machining Pilot Plant Plant 8
270 280 430	Plant 6 and 9 Machining Pilot Plant Plant 8 Boiler Plant
270 280 430 436	Plant 6 and 9 Machining Pilot Plant Plant 8 Boiler Plant Mechanical Department
270 280 430 436 452	Plant 6 and 9 Machining      Pilot Plant      Plant 8      Boiler Plant      Mechanical Department      Cafeteria
270 280 430 436 452 455	Plant 6 and 9 Machining      Pilot Plant      Plant 8      Boiler Plant      Mechanical Department      Cafeteria      Security Department
270 280 430 436 452 455 462	Plant 6 and 9 Machining      Pilot Plant      Plant 8      Boiler Plant      Mechanical Department      Cafeteria      Security Department      Stores
270 280 430 436 452 455 462 463	Plant 6 and 9 Machining      Pilot Plant      Plant 8      Boiler Plant      Mechanical Department      Cafeteria      Security Department      Stores      Transportation Department
270 280 430 436 452 455 462 463 465	Plant 6 and 9 Machining      Pilot Plant      Plant 8      Boiler Plant      Mechanical Department      Cafeteria      Security Department      Stores      Transportation Department      Garage

The Industrial Hygiene & Radiation Department had Internal Deposition Action Levels procedures from about 1970 that indicate actions related to the determination of percent MPLB to either uranium or thorium. Uranium-235 was detected primarily by the emission of its 186-keV photon. Uranium-238 was calculated from measurement of the <sup>234</sup>Th progeny assumed to be in equilibrium with the <sup>238</sup>U. Thorium-232 and <sup>228</sup>Th activities were determined based on equilibrium assumptions and detection of their progeny, most likely <sup>228</sup>Ac for <sup>232</sup>Th, but <sup>212</sup>Pb might have been used for assessment of both thorium isotopes. Thorium-230 is not readily detectable by in vivo measurements. There appeared to be no attempt to detect TRU contaminants with the MIVRML. The only quantification routinely made

Sample type coding system – specific

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 43 of 188
--------------------------------	-----------------	----------------------------	----------------

Type of analysis	Method	Period	Sample frequency	MDA
Urine–plutonium (Pu-239/240)	Chemical extraction and gross count	Before 1988	No routine schedule- unknown	~0.1 pCi/sample
Urine–plutonium (Pu-239/240)	Extraction and alpha spectroscopy count	1990s-2006	Special study samples only	0.1 pCi/sample
Urine-plutonium (Pu-238/239/240)	Extraction and alpha spectroscopy count	1986–1990	Workers potentially exposed to POOS in Plants 4 and 8	0.02 dpm/sample (0.22 pCi/sample) <sup>b</sup>
Fecal–plutonium (Pu-239/240)	Extraction and alpha spectroscopy count	1990s-2006	Special study samples only	0.1 pCi/sample
Urine–thorium Th-230 or Th-232	Extraction alpha spectroscopy count	1990s-2006	Not performed	0.1 pCi/sample
Fecal–thorium Th-230 or Th-232	Extraction and alpha spectroscopy count	1990s-2006	Only in cases of significant exposure	0.1 pCi/sample
Urine–radium <sup>a</sup> Ra-226	Unknown	1952–1953	Special study samples	30 dpm/sample
Urine–radium <sup>a</sup> Ra-226	Unknown	1954–1976	Special study samples	1.3 dpm/sample
Urine-radium <sup>a</sup> Ra-226	Unknown	1971–1989	Special study samples	0.07 dpm/sample
Urine or fecal – radium (Ra-226)	Unknown	1997–2006	Contingency only	0.1 pCi/sample

Table 5-25.	MDAs for	nonuranium	radionuclides. <sup>c</sup>

Contractor for radium samples not known. The MDAs listed are typical of existing industry standards. a.

b. Initial contractual MDA; this MDA could not be maintained and was increased to 0.13 dpm/sample (Bassett et al., p. 76). Suspected positive urine samples were followed with additional urine samples, in vivo counts, and fecal samples. c. MDAs for the methods are found in Tomes (2001). For plutonium in urine and in feces, by extraction and alpha

spectroscopy count, for the period from the 1990s to 2006, the value is on p. 57. That for thorium in feces is also on p. 57. For radium-226 in urine or feces, the value is on p. 93.

Year	Number above 50% MPLB
1974	21
1975	21
1976	9
1977	8
1978	10
1979	13
1980	6

dose determined by direct lung count.	Table 5-26.	Typical reporting of internal	
	dose determ	ined by direct lung count.	

with the MIVRML was uranium lung burden in units of micrograms of uranium and occasionally thorium activity. Uranium quantification required an assumption about uranium enrichment. The results from the MIVRML were reported in micrograms of <sup>235</sup>U and milligrams of total uranium in the lung, which was translated to MPLB based on the assumed enrichment (generally 1%). The percent lung burden was then multiplied by 15 rem to obtain the assigned annual lung dose in rem.

Workers who had known exposures to high air concentrations, had high urine results, or were involved in an incident were counted on first priority each time the MIVRML visited the site. Other workers were counted based on their job exposure potentials, as shown in Table 5-27.

The IVEC operated at the Fernald site from 1989 to 2002. A subject with a 2.5-cm chest wall thickness had the MDAs listed in Table 5-28 at the 95% confidence interval for a 3.600-second count. The previous mobile counting system, which serviced Fernald from 1968 to 1989, provided reports to the site. However, no system performance characteristics have been discovered to date. Table 5-28 lists the MDAs for Fernald in vivo analyses.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 44 of 188
--------------------------------	-----------------	----------------------------	----------------

Labor category description	In vivo counting schedule
All chemical operators	Once per year
Members of Project Labor Pool	Once per year
Mechanical Department crafts,	During each MIVRML visit, 25% of the employees in these
Mechanical Department laborer,	classifications were scheduled to be counted, and each worker
Laundry group,	would be counted at least once during a 2-year period.
Industrial truck operator,	
Locomotive operator,	
Switchman,	
Graphite shop machinist,	
Machine tool operator,	
Degreaser,	
Crane operator,	
Stamper,	
Plant 6 laborer,	
Furnace operator heater,	
Mill man,	
Decontaminator,	
Transportation laborer	-
Cafeteria,	Salaried personnel and workers in these classifications were
Water treatment group,	not routinely counted because of low chronic exposure and low
Power plant group,	potential for unobserved acute exposures.
Heavy equipment operator,	
Motor vehicle operator,	
Stores Warehouse attendant,	
Checker,	
Industrial mechanic,	
Security Police officer,	
Porter,	
Toolmaker,	
Machine set-up,	
Tool room machinist,	
Gauge set-up,	
Inspector	

Table 5-27.	Typical MIVRML	counting schedule a	t Fernald in the 1970s.
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Summaries of the chest count MDAs by uranium enrichment and mode of counting are shown in Tables 5-29 and 5-30 for the MIVRML and IVEC facilities, respectively.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 45 of 188
--------------------------------	-----------------	----------------------------	----------------

Radionuclide	Facility <sup>a</sup>	MDA (nCi)	DU MDA (mg)	NU MDA (mg)	2% EU MDA (mg)
U-235	MIVRML 1968–1989	Not available	Not available	Not available	100 µg (20%)
U-238	MIVRML 1968–1989	Not available	Not available	Not available	6.5 <sup>ь</sup> (6 nCi for 1% EU)
U-238 (Th-234)	FMPC IVEC <sup>c</sup> 1989–2001	2.5	7.4 (3 nCi)	7.5 (5 nCi)	7.6 (12 nCi)
U-235	FMPC IVEC <sup>c</sup> 1989–2001	0.18	36 (15 nCi)	11.3 (8 nCi)	4.2 (7 nCi)
Pu-239	FMPC IVEC <sup>c</sup> 1989–2001	190	Not available	Not available	Not available
Am-241	FMPC IVEC <sup>c</sup> 1989–2001	0.25	Not available	Not available	Not available
Ra-226	FMPC IVEC <sup>c</sup> 1989–2001	~3.0	Not available	Not available	Not available

Table 5-28. In vivo bioassay MDAs for the lung.

a. The values for the FMPC IVEC MDAs are taken from Soldano (1997). The U-235 and U-238 values are on p. 45. Values for Pu-239 and Am-241 are on p. 63. Values for Ac-228 and Pb-212 are on p. 81 and are based on an assumption of equilibrium with Th-232. The MDA for Ra-226 is on p. 92.

b. The recorded MDA in mass quantities were reported in Scott et al. (1969, p. 7) on the basis of the enrichments assumption shown - and converted to activity units in this table. Values reported in a worker's record were below 10% MPLB, which implies an MDA of less than 1 nCi total 1% (assumed) U-235.

c. IVEC MDAs are from site internal dosimetry TBDs (Tomes 2001; Soldano 1997; WMCO 1990b).

Page 46 of 188

Enrichment	Isotopes	Mass %	Activity %	Total U SA	Isotopic MDA	Isotopic MDA	Total U MDA (pCi)	Total U MDA (mg)
DU	U-235	0.20%	1.07%	0.402	100	215	20.100	50.00
DU	U-238	99.80%	83.42%	0.402	6,500	2,184	2,618	6.51
NU	U-235	0.72%	2.28%	0.683	100	216	9,481	13.88
NU	U-238	99.27%	48.86%	0.683	6,500	2,185	4,472	6.55
1% EU	U-235	1.00%	2.10%	0.973	100	204	9,730	10.00
1% EU	U-238	98.99%	34.20%	0.973	6,500	2,185	6,389	6.57
2% EU	U-235	2.00%	2.68%	1.616	100	217	8,080	5.00
2% EU	U-238	97.98%	20.40%	1.616	6,500	2,187	10,721	6.63
Table 5-30. Summary of IVEC chest count MDAs by uranium enrichment, 1989 to 2002.								
							Total U	Total U

Enrichment	Isotopes	Mass %	Activity %	Total U SA (pCi/µg)	Isotopic MDA (nCi)	Total U MDA (nCi)	Total U MDA (pCi)	Total U MDA (mg)
DU	U-235	0.20%	1.07%	0.402	0.18	17	16,822	41.85
DU	U-238	99.80%	83.42%	0.402	2.5	3	2,997	7.45
NU	U-235	0.72%	2.28%	0.683	0.18	8	7,895	11.56
NU	U-238	99.27%	48.86%	0.683	2.5	5	5,117	7.49
1% EU	U-235	1.00%	2.10%	0.973	0.18	9	8,571	8.81
1% EU	U-238	98.99%	34.20%	0.973	2.5	7	7,310	7.51
2% EU	U-235	2.00%	2.68%	1.616	0.18	7	6,716	4.16
2% EU	U-238	97.98%	20.40%	1.616	2.5	12	12,255	7.58

# 5.5 DOSE RECONSTRUCTION

## 5.5.1 Uranium Dose Assessments

Note that employees at the FMPC who were not employed by NLO (or by DOE or its predecessor agencies), and who worked at FMPC from January 1, 1951, through December 31, 1983, are included in the SEC due to the fact that their uranium urinalysis results might not have been included in the database used for the coworker study. While internal uranium doses for members of this class cannot be completely reconstructed, NIOSH intends to use any internal monitoring data that may become available for an individual claim (and that can be interpreted using existing NIOSH dose reconstruction processes or procedures).

## 5.5.1.1 Bioassay Data

A uranium urinalysis program was administered at Fernald starting in 1952. Data should be available for exposed workers because of the extensive urine bioassay program at Fernald. Table 5-31 contains a summary of the MDAs for routine urinalysis samples. See Section 5.4.3 and Table 5-29 for additional information.

Table 5-31. Routine tranium unnalysis capabilities.				
Method of analysis	Period	MDA		
Fluorophotometry	1952–1993	14 µg/L		
Chemchek KPA	1993-09/2002	0.17 µg/L (total U)		
ICP-MS	09/2002–2006	0.15 µg/L (total U)		

Table 5.24 Deutine une since unional usia approbilities

In vivo bioassay, in the form of lung counts, was also performed. From 1968 to 1989, measurements from the Y-12 MIVRML are available. From 1989 to 2002, the lung counts took place in the Fernald IVEC. Refer to Tables 5-29 and 5-30 for the MDAs for each of these counters.

## 5.5.1.2 <u>Uranium Breathing Zone Results</u>

Uranium urine results and chest count measurements take precedence over air data, and uranium BZ results do not have to be evaluated if there are positive urine or chest count measurements. However, when those results are all negative and there are positive uranium BZ results, include the intake based on BZ sample results in addition to the missed dose based on urine or chest count measurements. Ingestion intakes are also assigned when only BZ data are used to assign inhalation intakes (i.e. there are no in vitro or in vivo bioassay), as described in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004).

## 5.5.1.3 Uranium Enrichment

For converting uranium mass to activity, use the values in Table 5-2. Table 5-32 lists the default assumptions for cases where the uranium enrichment is not known (this will be most, if not all, cases).

Years	Enrichment			
1952–1964	1%			
1965–2006	2%			

Table 5-32. Default uranium enrichment.

# 5.5.1.4 Recycled Uranium

Starting with 1961, RU components should be added to uranium intakes. Intakes are assigned by applying the Table 5-33 (ratio to uranium mass) or Table 5-34 (ratio to activity) contaminants and ratios to the calculated uranium intake. The selected material types for RU contaminants are

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 48 of 188
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assigned using the direction in ORAUT (2014a). <u>These contaminants are added to all uranium</u> intakes regardless of monitoring method (urine, chest counts, BZ air samples, etc.).

Isotope	1961–1964 (Ba/a U)	1965 (Bq/q U)	1966–1972 (Ba/a U)	1973–1975 (Ba/a U)	1976–1985 (Ba/a U)	1986 on (Bg/g U)
U	3.61E+04	5.98E+04	5.98E+04	5.98E+04	5.98E+04	5.98E+04
Pu-239	2.96E+01	2.96E+01	2.93E+01	1.17E+03	1.16E+03	1.15E+03
Pu-241	3.05E+02	3.05E+02	1.88E+02	7.53E+03	4.66E+03	2.89E+03
Am-241	1.96E–02	1.96E–02	3.85E+00	1.54E+02	2.46E+02	3.00E+02
Np-237	1.02E+01	1.02E+01	1.02E+01	2.81E+02	2.81E+02	2.81E+02
Tc-99	3.77E+03	3.77E+03	3.77E+03	1.26E+04	1.26E+04	1.26E+04
Th-232	4.07E-02	4.07E-02	4.07E-02	4.07E-02	4.07E-02	4.07E-02
Th-228	2.85E-02	2.85E-02	2.85E-02	2.85E-02	2.85E-02	2.85E-02
Ru-106	4.08E+03	4.08E+03	4.08E+03	4.08E+03	4.08E+03	4.08E+03
Zr-95	6.12E+02	6.12E+02	6.12E+02	6.12E+02	6.12E+02	6.12E+02
Nb-95	6.12E+02	6.12E+02	6.12E+02	6.12E+02	6.12E+02	6.12E+02
Sr-90	1.63E+02	1.63E+02	1.63E+02	1.63E+02	1.63E+02	1.63E+02

Table 5-33. RU contaminant intakes per unit mass of uranium.

Table 5-34. RU contaminant intakes per unit activity of uranium.

Isotope	1961–1964 (Bq/Bq U)	1965 (Bq/Bq U)	1966–1972 (Bq/Bq U)	1973–1975 (Bq/Bq U)	1976–1985 (Bq/Bq U)	1986 on (Bq/Bq U)
U	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00	1.00E+00
Pu-alpha <sup>a</sup>	8.19E–04	8.19E–04	8.11E–04	3.25E-02	3.22E-02	3.20E-02
Pu-241	8.44E–03	8.44E-03	5.22E-03	2.09E-01	1.29E–01	7.99E–02
Am-241	5.43E–07	5.43E–07	1.07E–04	4.26E-03	6.80E-03	8.32E-03
Np-237	2.83E-04	2.83E-04	2.83E-04	7.78E–03	7.78E-03	7.78E–03
Tc-99	1.05E–01	1.05E–01	1.05E-01	3.48E–01	3.48E-01	3.48E–01
Th-232	1.13E–06	1.13E–06	1.13E-06	1.13E–06	1.13E-06	1.13E–06
Th-228	7.89E–07	7.89E–07	7.89E–07	7.89E–07	7.89E–07	7.89E–07
Ru-106	1.13E–01	1.13E–01	1.13E–01	1.13E–01	1.13E–01	1.13E–01
Zr-95	1.69E–02	1.69E–02	1.69E-02	1.69E–02	1.69E-02	1.69E–02
Nb-95	1.69E-02	1.69E-02	1.69E-02	1.69E-02	1.69E-02	1.69E-02
Sr-90	4.52E-03	4.52E-03	4.52E-03	4.52E-03	4.52E-03	4.52E-03

 The plutonium alpha mixture is assessed as 100% Pu-239. Am-241 and Pu-241 are assessed based on 6% weaponsgrade plutonium mixture.

# 5.5.1.5 <u>Chemical Form</u>

If the chemical compound is known for a particular exposure scenario, then Table 5-3 can be consulted for uranium chemical forms. However, dose reconstructors must typically evaluate each absorption type to determine the type most favorable to the claimant.

# 5.5.1.6 Unmonitored Uranium Workers

For unmonitored uranium workers, dose reconstructors should use the uranium intake rates from Attachment C, Internal Dosimetry Uranium Coworker Data, in combination with the special instructions (e.g., the addition of RU contaminants) in this section and subsections.

# 5.5.2 Thorium and Thoron Dose Assessments

Thorium processing did not occur in 1953; therefore thorium dose reconstruction is not required for 1953. There have been two SEC classes approved because of the infeasibility of constructing the internal dose due to thorium. Therefore, thorium doses for 1954 through 1978 are also not reconstructed.

The method for assigning thoron (<sup>220</sup>Rn) does not rely on those used for thorium assessment, so although thoron exposures may be associated with thorium, thoron dose is assigned during the thorium SEC periods.

The dose reconstruction method varies depending on the available data and timeframe. In many instances thorium workers were routinely monitored and chest count data are available. In other instances BZ air sampling data are available, and in other times the demonstrated effectiveness of radiological controls makes it possible to assume that no worker was exposed to concentrations in excess of 10% of the derived air concentration (DAC). Sustained airborne activity levels in excess of 10% of the DAC triggered airborne area radiological control posting.

Table 5-35 shows the dose reconstruction method for various times and different situations of data availability.

	In vivo data		
Timeframe	exists (yes or no)	Evaluation method	Section
1979–1994	Yes	In vivo results	5.5.2.1.1
1979–1989	No	Coworker data	5.5.2.3.1
1990–1994	No	Assume full-time occupancy in areas at 10% class W $^{\rm 232}{\rm Th}~{\rm DAC}$	5.5.2.3.2
1995–2006	Yes	Evaluate in vivo data to discover previous intakes. If a previous intake is discovered, use BZ data. For new intakes, use positive in vivo data instead of negative BZ data. If in vivo results are below MDA, use BZ data.	5.5.2.1.3
1995–2006	No	Use BZ data	5.5.2.1.2

## Table 5-35. Methods for thorium dose determination.

## 5.5.2.1 Monitored Thorium Assessment Methods, 1979 to 2006

# 5.5.2.1.1 In Vivo Bioassay, 1979 to 1994

As previously discussed, *in vivo* lung counts were performed from 1968 to 1989 using the Y-12 Plant MIVRML. From 1989 to 2002, the counts took place in the Fernald IVEC. Thorium-232 and its progeny <sup>228</sup>Th cannot be detected when inside the body, but the gamma emissions of the subsequent <sup>228</sup>Ac and <sup>212</sup>Pb progeny, are of sufficient energy to be detected. For this reason, in vivo bioassay for <sup>232</sup>Th is often performed by quantifying the activity of the <sup>228</sup>Ac or <sup>212</sup>Pb in the body and then calculating the amount of <sup>232</sup>Th (and relatively long-lived progeny <sup>228</sup>Ra and <sup>228</sup>Th) present by making assumptions about the relative ratios of the nuclides.

The FMPC <sup>228</sup>Ac and <sup>212</sup>Pb chest count results appeared to exhibit a bias, so an analysis of the data was performed and bias correction value calculated. MDAs were also generated. Attachment D, Evaluation of Fernald Actinium-228/Lead-212 Chest Count Data, contains the details of this analysis. Table 5-36 summarizes the bias corrections while Table 5-37 summarizes the MDA values for <sup>228</sup>Ac and <sup>212</sup>Pb. MDAs from Table 5-37 should be used to evaluate chest count measurements unless there are claim-specific chest count MDAs. Positive chest count results (i.e., those >MDA) are adjusted by <u>subtracting</u> the bias values in Table 5-36 before the results are used.

Document No. ORAUT-TKBS-0017-5 Revision No. 01 Effective Date: 07/22/2016 Page 50 of 188

Table 5-36.	Summary of bias corrections for
<sup>228</sup> Ac and <sup>212</sup>	Pb (nCi) in chest counts. <sup>a</sup>

Year	<sup>212</sup> Pb bias	<sup>228</sup> Ac bias				
1979	-0.014	0.031				
1980	-0.045	0.025				
1981	-0.056	0.022				
1982	-0.058	0.010				
1983	-0.101	-0.008				
1984	-0.089	0.001				
1985	-0.086	0.017				
1986	-0.085	0.020				
1987	-0.047	0.008				
1988	0.002	0.058				
1989–2001 <sup>b</sup>	0.072	0.182				

a. These values are to be subtracted from a positive (i.e., >MDA) result.

b. ORAUT (2014d) provides the basis for these values.

Table 5-37. Thorium chest count MDAs (nCi), 1979 to 2001.

Counter	Years	<sup>212</sup> Pb MDA	<sup>228</sup> Ac MDA
MIVRML	1979–1988	0.24	0.24
IVEC	1989–2001	0.42	1.10

Because IMBA is not designed to be used with multiple-nuclide chains, it cannot be used for an exact assessment when progeny are used to determine the parent intake. However, an approximation can be made that can be used for many cases. ORAUT-OTIB-0076, *Guiding Reconstruction of Intakes of Thorium Resulting from Nuclear Weapons Programs* (ORAUT 2014b), derives the basis and provides examples of how to evaluate thorium chest count data using IMBA. Assumptions about the process history of the material are made such that the derived intakes are favorable to the claimant. In most cases, the <sup>212</sup>Pb activity is a more reliable indicator of a thorium intake, so it is used for the thorium for dose reconstruction. Triple-separated thorium is assumed because it results in intakes that are favorable to the claimant.

Because IMBA does not correctly calculate <sup>232</sup>Th and <sup>228</sup>Th doses, the following steps apply only to the <u>intake</u> assessment. Alternate methods must be used for the dose assessment.

Given a <sup>212</sup>Pb chest count result for a chronic intake longer than 1 year or a chest count more than 30 days after an acute intake:

Evaluate the chest burden using the <sup>228</sup>Th biokinetic model in IMBA (i.e., use the <sup>212</sup>Pb result to model the <sup>228</sup>Th intake in IMBA).

Multiply the intake rate obtained in step 1 by a factor of 1.1 and assign it as the intake rate of <sup>228</sup>Th.

Divide the <sup>228</sup>Th intake rate by 0.19 to obtain the <sup>232</sup>Th intake rate.

If the conditions above (chronic intake longer than 1 year or chest count more than 30 days after an acute intake) are not met and an over- or underestimate cannot be performed, contact the PID for a best estimate.

Document No. ORAOTTIRDS-0017-51 Revision No. 01 [Effective Date. 07/22/2010] Fage 51 01 1	Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 51 of 188
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## 5.5.2.1.2 Breathing Zone Air Samples, 1995 to 2006

Thorium BZ samples are more sensitive than chest count measurements. Therefore, for 1995 through 2006, thorium doses are typically based on BZ data. Ingestion intakes are also assigned when only BZ data are used to assign inhalation intakes (i.e. there are no in vitro or in vivo bioassay), as described in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004). See Section 5.5.2.1.3 for a discussion of how to handle cases with both chest counts and BZ sample results.

When calculating intakes based on BZ data, include the missed dose [see Table 5-19 for minimum detection levels (MDLs)]. Use the highest value in the range of MDLs in Table 5-19 to assess the missed dose. Reported BZ values less than the MDL are set equal to the MDL (corrected for any respiratory protection, to be consistent with the reported measured results). Results are summed and assessed as an annual chronic intake rather than being assessed individually.

The solubility class (W or Y) that is in the claimant's records for each radionuclide and year is used for converting reported DAC-hr to activity. The DAC hours "with respirator" column (there are no totals in the "without resp" column) should be used, and the DAC-hr for each combination of radionuclide and class for each year must be summed before calculating the intake. The solubility type that is most favorable to the claimant should then be selected for calculating the dose [3].

For thorium, the BZ results are isotopic for <sup>228</sup>Th, <sup>230</sup>Th, and <sup>232</sup>Th, although there are few <sup>228</sup>Th results and these were always analyzed in conjunction with <sup>232</sup>Th. In addition to isotopic thorium results in HIS-20, a review in August 2013 indicated five custom nuclides identified as BL-13, BL-65, CELL 8, KS-65 and RT-210 [4]. Fernald used these custom nuclides for known mixtures of nuclides by calculating an effective DAC (EDAC) for the mixture to assign a more accurate DAC-hr exposure total for BZ air monitoring. The likely definitions for BL-13, BL-65, and KS-65 have been located. These and potential definitions for CELL-8 and RT-210 are described in Attachment E, Technical Basis for Assigning Doses from Effective Derived Air Concentration Breathing Zone Results. The attachment also provides a method of calculating intakes from the EDAC data for dose reconstruction. A summary of the custom nuclides is shown in Table 5-38.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 52 of 188
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Custom		Isotope or element,
nuclide	EDAC (µCi/mL)	activity fraction
BL-13	3.0E-12	Th-232, 0.0504;
		Th-230, 0.248;
		Th-228, 0.0565;
		U-total <sup>c</sup> , 0.645
BL-65	2.22E-12	Th-232, 0.20;
		Ra-228, 0.20;
		Ac-228, 0.20;
		Th-228, 0.20;
		Ra-224, 0.20
Cell 8 <sup>a</sup>	Reserved	Reserved
KS-65 silo 1 <sup>b</sup>	3.2E–11	K-65 silo 1,
		see Table 5-13
KS-65 silo 2 <sup>b</sup>	2.0E-11	K-65 silo 2,
		see Table 5-13
RT-210	2.25E-10	Pb-210, 0.333;
		Bi-210, 0.333;
		Po-210, 0.333;

Table 5-38. Custom nuclides from EDAC BZ results.

a. Contact Site Lead for guidance.

b. Dose reconstructors should use the mixture of either silo 1, 2 or 3 that is most favorable to the claimant, if unknown.

c. Because BL-13 primarily consists of uranium, it is not a replacement for thorium bioassay.

Dose reconstructors should apply the activity fraction for each isotope or element, then multiply the reported DAC-hr by the EDAC and the breathing rate ( $1.2 \times s10^6$  mL/hr) to get the total intake per year.

# 5.5.2.1.3 In Vivo Bioassay and BZ Data, 1995 to 2006

There might be chest count data and BZ air monitoring results for a thorium worker from 1995 to 2006. If all chest count data are below the MDA, then only the BZ air monitoring results should be used to evaluate thorium dose. If thorium BZ air monitoring data are negative, but there are positive chest count results, then chest counts should be reviewed to determine if there were thorium intakes before 1995. An evaluation of the previous in vivo counts is performed to determine if the in vivo results reflect a lung burden that had been previously identified from earlier exposure. If a previous intake is discovered, then use the BZ data. For new intakes, use positive in vivo data instead of negative BZ data.

# 5.5.2.2 Assignment of Unsupported Radium

For triple-separated thorium, there is no <sup>228</sup>Ra intake associated with the thorium intake. However, in some unusual cases, there might have been an intake of <sup>228</sup>Ra that was not associated with a thorium intake. This is called an intake of unsupported radium (and not part of the thorium mixture), and is evaluated separately, as discussed below. When a thorium intake occurs, the activity ratio between <sup>228</sup>Ac and <sup>212</sup>Pb can be up to 1:1. If the ratio exceeds 1.5:1, this is an indicator of an intake of unsupported radium and the <sup>228</sup>Ra intake rate is estimated from the <sup>228</sup>Ac activity.

When there was an intake of unsupported radium, the following approach is taken.

Because <sup>228</sup>Ac has a 6.15-hr half-life, it quickly (about 2 days) comes to equilibrium with its <sup>228</sup>Ra parent. It can therefore be used to directly assess the <sup>228</sup>Ra-228 intake. Determine the <sup>228</sup>Ra intake

by assuming a <sup>228</sup>Ra chest count equal to the <sup>228</sup>Ac measured value. Assume a type M material because this is not associated with a thorium intake.

## 5.5.2.3 Assignment of Unmonitored Thorium Dose

## 5.5.2.3.1 Thorium Coworker Model, 1979 to 1989

The derivation of the thorium intakes is described in Attachment F, Thorium In Vivo Coworker Study, 1979 to 1989. As noted in the attachment, as well as discussed in Section 5.5.2.1.1, triple-separated thorium is assumed because it results in intakes that are favorable to the claimant. The coworker thorium intake rates are summarized in Table 5-39.

Intake solubility type	50th percentile (pCi/d)	GSD	95th percentile (pCi/d)
<sup>232</sup> Th Type M	35.3	3.00	215
<sup>228</sup> Th Type M	6.71	3.00	40.85
<sup>232</sup> Th Type S	5.46	3.00	33.3
<sup>228</sup> Th Type S	1.04	3.00	6.33

Table 5-39. Thorium coworker intake rates, 1979 to 1989.

Thorium coworker doses are assigned to all unmonitored workers, unless there is a reason why they should be excluded (e.g., a secretary or administrative individual who worked only in nonradiological areas). The 95th-percentile intake rate, with a constant distribution, is assigned to those with the highest potential for exposure. Workers with a baseline thorium fecal sample are included in this group, as well as subcontractors from IT Corporation working during 1988 and 1989. All others are assigned the 50th-percentile intake rate with a lognormal distribution. The same material type is assigned for both isotopes of thorium (i.e., both are assigned as type M or both are assigned as type S).

An unsupported radium intake is not part of the coworker model and is not assigned.

# 5.5.2.3.2 Airborne Thorium Concentration, 1990 to 1994

The implementation of DOE 5480.11 (DOE 1988) requirements by January 1990 was readily apparent from the site documentation of the thorium overpacking work (access control, respiratory protection requirements, air monitoring with GA and BZ samples, and pre-job and follow-up fecal samples if required.) This provides confidence that no thorium workers were exposed to greater than 10% of a DAC (with respiratory protection being taken into account) on a sustained or average basis. Routine workplace air monitoring in nonairborne radioactivity areas was a site requirement, which provides a general assurance that workers were not chronically exposed to airborne levels greater than or equivalent to 10% of a DAC.

Because the thorium overpacking procedures did not appear to change much over time, it was considered useful to compare the thorium BZ results from the later periods to the 10% of a DAC assumption proposed for 1990 to 1994. The total annual thorium exposures in DAC-hr from 1993 to 2006 in the HIS-20 database were fit to a lognormal distribution. The GM of the results without the respiratory protection factor did not exceed 17 DAC-hr/yr for any of the three data-weighting methods. This is much less than 0.1 DAC times 2,000 hr/yr (200 DAC-hr/yr), which would be the upper bound. With respiratory protection factors applied, the 95th percentile of the BZ results did not exceed 14 DAC-hr/yr for any of the three data-weighting methods [5].

Therefore, the use of 10% of the thorium DAC air limit is appropriate. The thorium dose is based on the assumption of 100% <sup>232</sup>Th as favorable to claimants. Airborne areas would have been controlled to the lower Class W DAC. It is a standard industry practice for the lowest DAC value to be used

because, for a given air concentration, this results in a higher exposure in DAC-hours. An inhalation and ingestion intake of <sup>232</sup>Th is assumed. Although the thorium exposure in 1990 could be limited to January to May 1990 (due to 5 months of Project 2 duration), a full year of exposure should be assumed as favorable to claimants. Attachment G, Class W Thorium-232 Inhalation Intake Rate Based on 10% of the Derived Air Concentration, provides the assumptions used to derive these intake rates. Ingestion doses are assigned as described in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004), where the amount of activity ingested on a daily basis can be approximated assuming it to be 0.2 times the activity per cubic meter of air.

For 1990 through 1994, unless case-specific in vivo measurements are available, assume that each thorium worker was a full-time occupant of an area where the airborne thorium activity concentration was 10% of the DAC for Class W <sup>232</sup>Th, which is  $5 \times 10^{-14} \,\mu$ Ci/mL.

The derived inhalation and ingestion intake rates are shown in Table 5-40. Dose reconstructors should assign these intake rates as a constant distribution in IREP.

Table 5-40. <sup>232</sup>Th inhalation and ingestion rates based on 10% DAC.

Years	Inhalation rate (dpm/yr)	Ingestion rate (dpm/yr)
1990–1994	266.4	5.55

# 5.5.2.4 Assignment of Thoron Dose, 1954 to 2006

As previously discussed, thoron was present at Fernald. The primary dose from thoron is to the respiratory tract organs and is delivered primarily by the short-lived progeny (NIOSH 2013).

Thoron doses are assigned to all workers, unless there is a reason why they should be excluded (e.g., a cafeteria worker, accounting, legal staff individual, etc. who worked only in nonradiological areas). Thoron doses are summarized in Table 5-41. A constant distribution is assumed in IREP.

Table 5-41.	Thoron worker exposure.	
Years	Category	WLM/yr
1954–1971	Operations and Handling	1.6
1972-1985	Long-term DOE repository storage	1.6
1986–1987	Passive storage	No significant dose
1988–2006	Remediation (radiological workers) <sup>a</sup>	0.53
1988–2006	Remediation (non-radiological workers) <sup>b</sup>	No significant dose

a. Radiological workers are those that could have worked in a process area and are assigned thoron dose.

b. Nonradiological workers are not assigned thoron exposures because they would not have received any significant dose.

Table 1 of DCAS-TIB-011, *Lung Dose Conversion Factor for Thoron WLM* (NIOSH 2013), provides dose conversion factors for converting WLMs to dose in rem for the lung, ET1, and ET2. An equilibrium of 80% <sup>212</sup>Bi/<sup>212</sup>Pb is assumed.

# 5.5.3 Raffinate Dose Assessments

## 5.5.3.1 <u>1952 to 1954</u>

Assess radon breath measurements in this timeframe as follows:

1. Convert the radon breath analysis result to units of picocuries per liter.

- Convert the value in step 1 to <sup>226</sup>Ra whole-body content in pCi by multiplying by 2.52 × 10<sup>5</sup> pCi/(pCi/L).
- 3. Use the whole-body <sup>226</sup>Ra content in IMBA to calculate a chronic intake of type M <sup>226</sup>Ra beginning on January 1, 1952 (or date of employment, if later), through the date of the last radon breath analysis result.
- 4. In addition to the <sup>226</sup>Ra intake estimate, apply the derived intake to the isotopes and ratios in Table 5-16.

If there are no radon breath analysis results for a worker, assign the <sup>226</sup>Ra intake rate from Table 5-15. This intake rate is then used with the activity ratios in Table 5-16 to assign intakes of associated raffinate nuclides.

# 5.5.3.2 <u>1954 to 1958 (Pitchblende Ores)</u>

To account for the impurities in pitchblende, assume a <sup>230</sup>Th intake equal to the calculated uranium (total uranium activity) intake. Assign the additional impurities by applying the ratios in Table 5-17. Dose reconstructions with overlapping periods in 1955 and 1956 require a comparison between Table 5-16 and Table 5-17 doses for silos 1 and 2, and the most favorable to the claimant selected for assigning dose.

# 5.5.3.3 <u>1959 to 1961 (Yellowcake)</u>

To account for the handling or processing of yellowcake, assume a <sup>230</sup>Th intake equal to the calculated uranium (total uranium activity) intake. Assign the additional impurities by applying the ratios in Table 5-17 for silo 3. Include the RU contaminants beginning in 1961.

# 5.5.4 Radon Exposure Assessment

These records typically appear in Personnel Exposure files, separate from the DOE Response file in the NIOSH-DCAS Claims Tracking System (NOCTS) and are radon exposure records in WLMs. Follow ORAUT-TKBS-0017-4 (ORAUT 2015b) guidance when evaluating these records.

# 5.5.5 Site Closure

In 2006, the site remediation was complete, and it was transferred to the DOE Office of Legacy Management. This ended the site bioassay and BZ measurements. Internal doses after that date should be assessed using the current version of ORAUT-TKBS-0017-4 (ORAUT 2015b).

# 5.6 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database.

[1] Potter, Eugene. ORAUT Team. Principal Health Physicist. November 2014. An investigation was conducted into the possibility of limiting the workers to which <sup>226</sup>Ra intakes would apply by external dose. NLO (1952) includes radon breath results for 1952. Assuming that workers doing the K-65 drum handling were subject to high external dose rates,

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 56 of 188

the HIS-20 database was queried for 1952 external dose results. A comparison was made of the groups with 1952 radon breath results with the other individuals monitored for external dose in 1952. A comparison was made of the list of individuals in 1952 with breath data (65 total) to claimant files, and showed 10 workers with breath data. DOE files in NOCTS were examined; none included breath data. [redacted] of the individuals did have film badge investigations in the DOE file, which listed work with K-65 drums. The distributions were different with the breath sample group being higher, but the two distributions overlapped significantly. In addition, there were [redacted] workers with radon breath samples but no external data for 1952 in HIS-20. From this, it was concluded that an external dose trigger could not be used to assign <sup>226</sup>Ra intakes.

- [2] Rich, Bryce. ORAUT Team. Principal Health Physicist. March 2008. A review conducted on gross beta urine sample results at Fernald that sometimes appear in claimant records indicated that they were not well enough defined to be of value.
- [3] Brackett, Elizabeth. ORAUT Team. Principal Internal Dosimetrist. August 2013. Use of the solubility class (absorption type) of BZ results from claimant records, DAC hours "with respirator" column, and then selection of the "max" solubility option for dose evaluation of BZ samples ensures doses that are favorable to the claimant.
- [4] Potter, Eugene. ORAUT Team. Principal Health Physicist. August 2013. A review of HIS-20 indicated that there were "custom" air monitoring nuclides identified as BL-13, BL-65, CELL 8, KS-65, and RT-210, and these might be in a claimant's records. A methodology for calculating an EDAC was developed.
- [5] Potter, Eugene. ORAUT Team. Principal Health Physicist. January 2015. A comparison and statistical analysis performed by Eugene Potter indicated that thorium BZ results from 1993 to 2006 in the HIS-20 database did not exceed the upper-bound limit of 0.1 DAC times 2,000 hr/yr.
- [6] Bryce, Rich L. ORAU Team. Principal Health Physicist. The <sup>233</sup>U and trace quantities of <sup>232</sup>U came from thorium from Hanford from 1977 to 1979. The thorium was "recycled thorium" with a contamination level of approximately 130 g <sup>233</sup>U per 10.8 MTTh. The total <sup>233</sup>U plant inventory spread through the 351 MTTh was approximately 4.2 kg. Trace levels of <sup>232</sup>U could have been present.
- [7] Bryce, Rich L. ORAU Team. Principal Health Physicist. The quantities stored in the process buildings of 15 MTTh awaiting immediate processing are not known from records or interviews. It is reasonable (based upon a knowledge of processing practices) to move materials to a ready point and the amount assumed is considered adequately conservative. The primary storage locations were outside on pads or in adjoining storage facilities.
- [8] Bryce, Rich L. ORAU Team. Principal Health Physicist.
  Thorium feed materials were received from a variety of separations facilities stockpiles. A 1-year delay since separation was judged to be a reasonable and conservative average.
- [9] Arno, Matthew. ORAU Team. Dose Reconstructor. June 18, 2007. Lognormal distributions typically provide the best fit to the available data and are a distribution suitable for input into IREP.

- [10] Arno, Matthew. ORAU Team. Dose Reconstructor. June 18, 2007. The error in individual bioassay results has a normal distribution because the dominant source of uncertainty is the counting statistics. Although the underlying group statistics have a lognormal distribution, each result was treated as if it had a normal distribution to match what was done for analysis of an individual's bioassay data and because the lognormal distribution of the data is addressed by analyzing both the 50th and 84th percentiles of the data.
- [11] LaBone, Thomas. ORAU Team. Deputy PID. December 18, 2007. The assumption of equilibrium between <sup>212</sup>Pb and <sup>228</sup>Th requires an assumption that none of the noble gas <sup>220</sup>Rn escapes the lung. Radon-220 has a half-life of only 55.6 s, which makes this a reasonable assumption.
- [12] Morris, Robert. ORAU Team. Principal Health Physicist. November 14, 2007. Inspection of in vivo monitoring records confirms that many of the workers counted during the first visit had "thorium worker" or "former thorium worker" noted on their individual MIVRML log sheets.
- [13] LaBone, Thomas. ORAU Team. Deputy PID. May 19, 2014. Bias factors were developed based on the Fernald post-SEC thorium methodology.
- [14] Arno, Matthew. ORAU Team. Dose Reconstructor. June 18, 2007. Lognormal distributions typically provide the best fit to the available data and are a distribution suitable for input into IREP.
- [15] Morris, Robert. ORAU Team. Principal Health Physicist. November 14, 2007. The Fernald SEC petition includes 1989. The thorium operations in 1989 were unchanged from those in 1988 and it is judged reasonable and efficient to extend the 1980 through 1988 period to include 1989.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 58 of 188
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Document No. ORAUT-TKBS-0017-5 Revision No. 01 Effective Date: 07/22/2016 Page 65 of 188

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	Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 68 of 188
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Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 71 of 188
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## GLOSSARY

## absorption type

Categories for materials according to their rate of absorption from the respiratory tract to the blood, which replaced the earlier inhalation clearance classes. Defined by the International Commission on Radiological Protection, the absorption types are F: deposited materials that are readily absorbed into blood from the respiratory tract (fast solubilization), M: deposited materials that have intermediate rates of absorption into blood from the respiratory tract (moderate rate of solubilization), and S: deposited materials that are relatively insoluble in the respiratory tract (slow solubilization). Also called solubility type.

## acute exposure

Radiation exposure to the body delivered in a short period. See chronic exposure.

### becquerel (Bq)

A unit of radioactivity equal to one disintegration per second.

## chronic exposure

Radiation dose to the body delivered in small amounts over a long period (e.g., days or years). See *acute exposure*.

### class

See inhalation class and absorption type.

### curie (Ci)

Traditional unit of radioactivity equal to 37 billion  $(3.7 \times 10^{10})$  becquerels, which is approximately equal to the activity of 1 gram of pure <sup>226</sup>Ra.

## decision level (DL)

Minimum level at which a particular device can detect and quantify exposure or radiation. Also called lower limit of detection and detection limit or level.

## depleted uranium (DU)

Uranium with a percentage of <sup>235</sup>U lower than the 0.7% found in natural uranium.

#### dose

In general, the specific amount of energy from ionizing radiation that is absorbed per unit of mass. Effective and equivalent doses are in units of rem or sievert; other types of dose are in units of rad, rep, or grays.

### dose equivalent

In units of rem or sievert, product of absorbed dose in tissue multiplied by a weighting factor and sometimes by other modifying factors to account for the potential for a biological effect from the absorbed dose. See *dose*.

### dosimetry

Measurement and calculation of internal and external radiation doses.

### enriched uranium (EU)

Uranium in which processing has increased the proportion of <sup>235</sup>U to <sup>238</sup>U to above the natural level of 0.7% by mass. Reactor-grade uranium is usually about 3.5% <sup>235</sup>U; weapons-grade uranium contains greater than 90% <sup>235</sup>U.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 72 of 188
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#### exposure

(1) In general, the act of being exposed to ionizing radiation. See acute exposure and chronic exposure. (2) Measure of the ionization produced by X- and gamma-ray photons in air in units of roentgens.

### inhalation class

Former respiratory tract inhalation classification scheme developed by the International Council on Radiological Protection for inhaled material according to its rate of clearance from the pulmonary region of the lung. Materials were classified as D (days, half-life less than 10 days), W (weeks, 10 to 100 days), or Y (years, more than 100 days). See *absorption type*, which superseded this concept.

### in vitro bioassay

Measurements to determine the presence of or to estimate the amount of radioactive material in the excreta or in other biological materials removed from the body.

#### in vivo bioassay

Measurements of radioactive material in the human body utilizing instrumentation that detects radiation emitted from the radioactive material in the body.

### isotope

One of two or more atoms of a particular element that have the same number of protons (atomic number) but different numbers of neutrons in their nuclei (e.g., <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U). Isotopes have very nearly the same chemical properties.

## maximum permissible lung burden (MPLB)

Historical occupational limit for radionuclides in the lung defined as the quantity of the radionuclide that could be present at any given time to deliver 15 rem per year to the lung at the end of a 50-year period of chronic exposure.

#### microcurie

A measure of radioactivity equal to one-millionth of a curie. See *curie*.

### millirem

A unit of radiation dose equal to one-thousandth of a rem. See rem.

# minimum detectable activity (MDA)

Smallest amount (activity or mass) of an analyte in a sample that can be detected with a probability  $\beta$  of nondetection (Type II error) while accepting a probability  $\alpha$  of erroneously deciding that a positive (nonzero) quantity of analyte is present in an appropriate blank sample (Type I error).

### natural uranium (NU)

Uranium as found in nature, approximately 99.27%  $^{238}$ U, 0.72%  $^{235}$ U, and 0.0054%  $^{234}$ U by mass. The specific activity of this mixture is 2.6 × 10<sup>7</sup> becquerel per kilogram (0.7 microcuries per gram).

## nuclide

Stable or unstable isotope of any element. Nuclide relates to the atomic mass, which is the sum of the number of protons and neutrons in the nucleus of an atom. A radionuclide is an unstable nuclide.
Document No. ORAUT-TKBS-0017-5 Revision No. 01 [Effective Date: 07/22/2016] Page 73 01	ment No. ORAUT-TKBS-0017-5	17-5 Revision No. 01	Effective Date: 07/22/2016	Page 73 of 188
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### potential alpha energy concentration (PAEC)

Kinetic energy in units of working levels potentially released in a unit volume of air by alpha particles emitted by the short-lived radioactive progeny of <sup>222</sup>Rn (<sup>218</sup>Po, <sup>214</sup>Pb, <sup>214</sup>Bi, and <sup>214</sup>Po) and <sup>220</sup>Rn (<sup>216</sup>Po, <sup>212</sup>Pb, <sup>212</sup>Bi, <sup>212</sup>Po). See *potential alpha energy exposure* and *working level*.

### potential alpha energy exposure (PAEE)

Average potential alpha energy concentration to which a worker is exposed multiplied by the time of exposure in working months of 170 hours (units of working level months). PAEE is the potential alpha energy concentration multiplied by time. See *potential alpha energy concentration* and *working level month*.

### rad

Traditional unit for expressing absorbed radiation dose, which is the amount of energy from any type of ionizing radiation deposited in any medium. A dose of 1 rad is equivalent to the absorption of 100 ergs per gram (0.01 joules per kilogram) of absorbing tissue. The rad has been replaced by the gray in the International System of Units (100 rads = 1 gray). The word derives from radiation absorbed dose.

### radiation

Subatomic particles and electromagnetic rays (photons) with kinetic energy that interact with matter through various mechanisms that involve energy transfer.

### radioactive

Of, caused by, or exhibiting radioactivity.

### radionuclide

Radioactive nuclide. See radioactive and nuclide.

### rem

Traditional unit of radiation dose equivalent that indicates the biological damage caused by radiation equivalent to that caused by 1 rad of high-penetration X-rays multiplied by a quality factor. The sievert is the International System unit; 1 rem equals 0.01 sievert. The word derives from roentgen equivalent in man; rem is also the plural.

### solubility type

See absorption type.

### transuranic (TRU) elements

Elements with atomic numbers above 92 (uranium). Examples include plutonium and americium.

### working level (WL)

Unit of concentration in air of the short-lived decay products of <sup>222</sup>Rn (<sup>218</sup>Po, <sup>214</sup>Pb, <sup>214</sup>Bi, and <sup>214</sup>Po) and <sup>220</sup>Rn (<sup>216</sup>Po, <sup>212</sup>Pb, <sup>212</sup>Bi, <sup>212</sup>Po) defined as any combination of the short-lived radioactive progeny of radon or thoron in 1 liter of air, without regard to the degree of equilibrium, that results in the ultimate emission of 130,000 megaelectron-volts of alpha energy; 1 WL equals  $2.083 \times 10^{-5}$  joules per cubic meter. See *potential alpha energy concentration*.

### working level month (WLM)

Unit of exposure to radon progeny defined as exposure for 1 working month (170 working hours) to a potential alpha energy concentration of 1 WL; 1 WLM equals 1 WL times

170 hours, which is 0.00354 joule-hours per cubic meter. See *potential alpha energy exposure* and *working level*.

### TABLE OF CONTENTS

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### LIST OF TABLES

# TABLETITLEPAGEA-1Timeline of PGDP shipments to Fernald78A-2Totals of recycled feeds to Fernald78

### A.1 Summary

The concern that the recommended sitewide default of 400 ppb U Pu<sup>2</sup> for the 1970s and 1980s might not bound the dose from RU contaminants for all categories of workers because of the higher values in process subgroup 10A is addressed in this attachment. The 10A process categorization represents "tower ash and decontamination residues from Portsmouth and Paducah GDPs" with the high values in the activity distribution coming from PGDP in a single shipment in 1980.

In the 1970s (until 1977) the ash and residues from PGDP had elevated contaminant concentrations, but they were generally under 10 ppb U Pu and were sampled and blended directly with uranium concentrates to attain operational concentrations equivalent to those processed directly from the primary sites. (See the Table 5-6 values of process subgroup 6A at 4.6 ppb U Pu at the 95th percentile.) During this period there were approximately 10 g of plutonium received, which generally increased the levels in the Fernald process streams, but the latter were still held to levels below 10 ppb U Pu. The concentrates ran out in 1977, requiring blending with the lowest process streams available to stay under the 10-ppb U level. In June of 1980 a single shipment of approximately 24 MTU PGDP process plant ash was shipped in 16 T-hoppers and contained 25.3 g of plutonium. This essentially doubled the plutonium inventory for the entire operational history of the plant to that point.

This shipment was sampled and stored primarily to develop blending material inventories and processes to accommodate this unusual shipment. After approximately a 2-year period, there is a 1982 record of repackaging five of the T-hoppers with the highest contaminant levels into large containers in Plant 4 to facilitate process needs in Plants 8 and 2/3. Air sampling records associated with this repackaging are recorded as well as the time required, which was done primarily on the first shift of several days. It is assumed that this record was generated to document an unusual operation with specific contamination control problems. Although these were five of the 16 T-hoppers with the greatest concerns and difficulties, the times of repackaging were used to estimate the repackaging of the hoppers that contained contaminants above the 400 ppb U Pu plantwide default. It is important to note that of the 16 T-hopper containers in the 1980 shipment, two contained 66% of the total plutonium contaminants, while six contained 88% of the plutonium itself. It is also of note that only eight of the hoppers contained plutonium contaminant concentration above 400 ppb U.

Based on the recorded times of the five hoppers with the greatest problems, 150 hours was conservatively estimated for repackaging those eight hoppers with concentrations above 400 ppb U Pu. This would be the handling time in April and May of 1982. There are recorded descriptions of further processing, the next step (after another delay) being blending with sump cake in Plant 8 of appropriate content to create a calcium urinate feed for further dissolution and process in Plant 2/3, these being semi-remote operations. The final recorded processing resulted in a formal approval to process the Plant 2/3 UO<sub>3</sub> product, which was in the range of 20 to 42 ppb U Pu.

### A.2 The Issue

The remaining issue related to acceptance of 400 ppb U Pu as a sitewide default is the question relating to the NIOSH decision not to use the Fernald process stream 10A (95th percentile of 1,732 ppb U Pu), as described in the Ohio Mass Balance Report (DOE 2000a) in establishing the

<sup>&</sup>lt;sup>2</sup> For the sake of simplicity, this attachment discusses only plutonium. The same principles apply to the other primary contaminants (neptunium and technetium), which are also listed with appropriate defaults in the guidance to the dose reconstructors.

values of the defaults. This process stream represents the wastes received from the GDPs (but primarily and specifically PGDP) starting in about 1975 or 1976, but realistically starting in 1980. Because the contaminant distribution within the materials of this process stream were handled episodically (as opposed to continuously) and of short duration in nature and blended with low-level feed materials to attain approved limits, the materials in this process stream were not used to establish the defaults.

The primary remaining question relates to the length of time that can reasonably be established as the maximum exposure time for any given individual worker to the contaminant ratios in the 10A process stream. Though the materials with the high levels of contaminants above the 400 ppb U Pu default were handled for a short period of time in 1982, were recognized as materials above permissible levels, and preventative measures were used to protect the workers from exposure, the presence of the materials of this process stream is a concern to FMPC Working Group reviewers as a potential exposure above those established.

### A.3 General Background

Quoting from Attachment E.2, "Selected Non-Routine Campaigns," of the Ohio Mass Balance Report (DOE 2000a, p. 900):

Over a period of several years, a large inventory of uranium-bearing residues was accumulated at the PGDP. Most of the residues were near-normal in isotopic assay and were highly diverse in physical and chemical characteristics. Although much of the residue inventory was generated by Paducah site operations, a significant quantity of the  $UF_4$ -type residues was generated at the FEMP and was shipped to Paducah for possible use as feed material in their operations. Very little of the  $UF_4$  shipped from the FEMP was used at Paducah because their operation was suspended shortly after the transfer was completed.

By early 1975, the Paducah residue inventory contained approx. 400 MTU. After assessing the operational and environmental impacts of processing this inventory at Fernald, DOE-ORO directed that the inventory be shipped to the FEMP for recovering the uranium in conjunction with the ore concentrate campaign that was in progress in Plant 2/3. The introduction of this inventory as a blend with ore concentrates began in 1976. Paducah continued shipping various types of residues after 1976. These materials were placed into FEMP's inventory of recoverable residues for eventual enriched uranium process campaigns in Plant 2/3.

### A.4 Timeline

The brief timeline and mass summary Tables A-1 and A-2 indicate two important points relevant to this discussion:

- 1. The concentration of contaminants relative to uranium was generally below 10 ppb U Pu before the single shipment of 16 T-hoppers of feed plant waste in 1980.
- 2. The total mass of 22.5 MTU in this shipment was only 0.3% of the total MTU received during this period.

Table A-1. Timeline of PGDP shipments to Fernal	able A-1.	1. Timeline of PGE	OP shipments	to Fernal
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Date	Discussion	References
1966–1974	UO <sub>3</sub> , U <sub>3</sub> O <sub>8</sub> incinerator ash materials – Pu at 6 ppb	Spenceley 1985; DOE 2000a,
	U Pu	p. 889
1975–1976	291 MTU scrap and fluoride residues – 2.6 g Pu at	Spenceley 1985; Spenceley 1985;
	9 ppb U	NLO 1957–1984; Author unknown
	Start of major transfer of PGDP scraps and waste	undated b
	materials to Fernald	
1979, 1981–1983	1,814 MTU of UO <sub>3</sub> , UNH –	Spenceley 1985
	10 g Pu at 5.6 ppb U Pu	
1980	22.5 MT Feed Plant ash –	Spenceley 1985; DOE 2000a;
	25.3 g Pu at 1,124 ppb U Pu. This shipment	NLO 1957–1984; Author unknown
	consisted of 16 T-hopper containers, 2 of which	undated b; Robinson 1989; Author
	contained 66% of the Pu. The highest container	unknown undated a; WMCO
	contained 39% of the Pu.	1986c; Walker 1986

Table A-2.	Totals of recy	vcled feeds to	Fernald (S	Spenceley	/ 1985).
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Materials	MTU/%	Pu-239 g	Pu-239 ppb U
UO <sub>3</sub>	6,112/82%	17.4	2.9
UNH	699/9%	4.7	6.7
Incinerator/tower ash	311/4%	0.2	0.7
UO <sub>2</sub>	39/0.5%	0.01	Not significant
Feed plant ash	22.5/0.3%	25.3	1,124
1975–1976 miscellaneous scrap	291/4%	2.6	9

### A.5 Discussion Points

- PGDP wastes had accumulated after shutdown of processing facilities in the early 1970s. The approximately 400 MTU were directed to Fernald for recovery after considering the practicality, radiological impact, and ability to blend the materials to acceptable levels for processing. During the period from the early 1970s through 1979 the concentrations from PGDP were generally below 10 ppb, though higher than the feed materials from the primary RU source facilities (Hanford and SRS specifically) by perhaps a factor of 2 or greater (DOE 2000a, p. 1,099; see process 6A values in Attachment F.1; 95th percentile value is 4.6 ppb U Pu). This material was easily blended directly with ore concentrates to bring the contaminant concentrations relative to uranium into the levels that were being processed straight from the PUREX processes at Hanford and SRS. However the supply of concentrates ran out in about 1977, which made it necessary to blend with lower levels of feed materials (but not NU).
- 2. However, this did introduce an added total quantity of plutonium to Fernald from the "secondary" sources (GDPs) that had not been through the decontaminating mechanisms of the diffusion cascades. That is, the materials that were sent during this time were scraps and residues and were in ranged from 6 to 10 ppb Pu rather than the parts per ton range.
- The 1980 shipment of primarily feed plant ash was unusually high in contaminant concentrations, adding over 25 g of plutonium to the plant inventory (essentially doubling the inventory for the entire plant history), and presenting an increased challenge to effectively blend the materials to acceptable levels (Spenceley 1985, pp. 7–17, 19, 20, 34; Mengel 1982, p. 4).

4. This shipment was not processed immediately: "These materials were placed into FEMP's inventory of recoverable residues for eventual EU process campaigns in Plant 2/3." The shipments were received at Plant 1 for immediate sampling and then storage until needed.

The following from Robinson (1989, p. 50) is instructive:

D.C. Bonfer answer – Paducah Feed Plant Ash was prepackaged from hoppers into drums in Plant 4. Drummed material was later blended with sump cake in the rotary kiln operation in Plant 8 producing seven lots of calcium urinate, which was used as feed for the refinery. Most of this material was converted to  $UO_3$ ; a quantity remains as UNH in the refinery.  $UO_3$ , which was contaminated with the plutonium, remains on inventory at NLO. The remainder of the  $UO_3$  was converted to  $UF_4$  and blended with three parts of  $UF_4$  produced from Hanford  $UO_3$  in the metal reduction operations.

- 5. Interviews with four senior former managers (ORAUT 2008b, 2011b, 2011c, 2011d) during the period in question were productive in providing operational insights and perceptions, but there was not a direct recollection of the specific time required for the processing the 16 T-hoppers to the point that the contaminants would be below the default level of 400 ppb U Pu.
- 6. The T-hoppers were stored for approximately 2 years (until 1982) when five of the highest level T-hoppers were taken to Plant 4 for repackaging into containers compatible with the other plant facilities (Mengel 1982). Repackaging took longer than expected due to some of the material having become solid in at least some of the containers. Instead of processing at a rate of two to three hoppers per shift (ORAUT 2011d), two of the containers required two shifts each (with an estimated 5 hours per shift due to airline respiratory protection and associated protective clothing requirements, for a total of 10 hours; Hinnefeld 1988), one container required three shifts (15 hours), one required five shifts (25 hours), and the remaining container required eight shifts (40 hours). This time requirement was complicated and the activities delayed by the need to correct procedures that were spreading contamination (Mengel 1982, p. 4, Table I). The first shift on April 28, 1982, was the only time removing material from the hopper to a tray on the floor was indicated.
- 7. This repackaging effort took place mainly on the first shift, presumably to minimize routine operations impact. Conclusions from these records place an upper bound on the time of potential exposure to the highest contamination in process 10A materials in the repackaging operation. All of the POOS from PGDP came in 16 T-hoppers. (39% of the activity was in the highest container, 67% in the highest two containers, 88% in the highest seven containers). The hopper with the highest contamination levels required five shifts or approximately 25 hours on five different shifts.
- 8. The Bonfer quotation in item 4 above indicates that the material in the 1980 shipment was taken to Plant 8 "sometime later" after repackaging and blended with "sump cake" in the rotary kiln. From the same quotation it is apparent that the blended calcium urinate (no longer process 10A contamination levels; that is, below the 400 ppb U Pu default level) was dissolved in Plant 2/3 resulting in liquid UNH. Again the materials were stored until appropriate processing to UO<sub>3</sub> was available (but again the materials had lower levels of contaminants below the recommended 400 ppb U default).
- 9. The next set of records are from about 1985 to 1987 when the material had been dissolved and run through Plant 2/3 extraction with UO<sub>3</sub> product that had plutonium levels of 26 to

42 ppb (Jelinek and Thomas 1985). In addition, a training program specifically for the POOS processing in Plants 4 and 5 discussed the processing of 168 MTU (UO<sub>3</sub>), including blending of the 42 ppb U Pu UO3 on a "one-to-one" basis with "in spec. materials" (Author unknown undated a). The training course also discussed of the fact that FMPC had raised the "target" limit to 20 ppb, but DOE headquarters had reduced the limit to 10 ppb again. However, the training was for the procedure of processing the 42 ppb U. This operation is not of interest in answering the question of whether the 400 ppb U is adequately bounding.

10. Conclusions about time of potential exposure to the materials after repackaging are based on the following discussion. After the repackaging in Plant 4, the material was again stored for a convenient time and circumstances for further processing and blending in Plant 8. It is possible that one or a few of the drums of material that were not free flowing could have been milled in Plant 1 before sending to Plant 8. This possibility is supported in DOE (2000a, p. 556), which indicates that one of the data points in the 10A process stream is from a Plant 1 dust filter. Both of these operations involved handling the large drums on automatic lifts and in enclosed systems. Time of exposure would be less than the repackaging times. The Plant 8 blended material was no longer above the 400 ppb U in question.

Therefore, the exposure times to the highest contaminant levels would be 20 shifts multiplied by 5 hours per shift or 100 hours for any single worker that was present for each of the 20 shifts required for repackaging five hoppers, or 150 hours to process the eight hoppers that exceeded the 400 ppb U. This assumes that each of the eight hoppers above 400 ppb required the same time as the problematic five recorded. This is extremely conservative due to:

- Only 3 of the hoppers were above 1,000 ppb, and only 8 of the 16 were above 400 ppb.
- The time of continuous exposure is significantly overestimated and particularly due to the use of airline respiratory protection and the equivalent protective clothing.
- The other 11 hoppers probably required much less time, as indicated in the recording of the five highest and more problematic.

This repackaging time would have been spent in 1982, assuming that the same worker was present during the repackaging of each of the 16 hoppers. Therefore, only 8% of the annual operating time in that year could have been spent in contact with this repackaging operation, and then only 50% of the containers had contaminants above 400 ppb U. After the repackaging the workers would be assigned to work with materials that were probably an order of magnitude or 2 lower than the 400 ppb U Pu.

11. The mass of the 1980 shipment, although containing 25.3 g of plutonium, was only 22.5 MTU. As time went on and after 4 to 5 years, much of this contamination was spread through the Fernald processes, raising many of them above previous levels. However, handling of the material did not require a consistent effort, as has been discussed, because it was in storage for most of this period. The mass did not account for more than a fraction of a percent of the material in process.

### A.6 Conclusion

It is the conclusion of this investigation that the levels of 1,732 ppb U Pu (at the 95th percentile for process group 10A) is adequately covered by the 400 ppb U Pu plantwide default. The 400 ppb Pu default was calculated by conservatively considering the maximum levels in any of those process groups that represent continuous operations. The highest levels in the ash and residues from PGDP were contained in less than 0.3% of the wastes from PGDP and required a maximum of a few hundred hours of handling before downblending. This represents a short exposure time with documented safety airline respiratory protection. The remaining exposure time in 1982 for any worker would have been with materials below the recommended defaults. There is no reason to believe that any worker could have an exposure for which the recommended default would not be adequately conservative.

### A.7 Discussion of References

### Ohio Mass Balance Report (DOE 2000a)

Transfer of PGDP wastes began in 1976 and continued. "These materials were placed into FEMP's inventory of recoverable residues for eventual EU process campaigns in Plant 2/3." Some of the wastes were blended and some were not (p. 900). See p. 901 for blending ratios, feed evaluations, and classification of scrap residues. Table E.2-1 is a listing of PGDP scrap classification, amounts, descriptions, pretreatments, etc. Table E.2-2 gives the Fernald permissible levels. The wastes were blended with ore concentrates until 1977, when the supply ran out. Between 1978 and 1986 77.5 MTU of residues were recovered in Plant 2/3 (Table E.2-4). The scraps from K-25 and Portsmouth GDP are discussed on p. 908.

### "Joint Task Force on Recycle Material Processing" (Spenceley 1985)

See pp. 4, 5, 7 to 20, and 34 particularly for a comparative, time-based summary of the PGDP wastes as received. 1980 was the high point. Table 1 on p. 34 gives the listing of the 16 T-hoppers with the 25 g of plutonium that came in 1980.

### "Air Sampling at Plant 4 Repackaging of Paducah Reprocessed Feed Plant Ash" (Mengel 1982)

This is a four-page summary of the air sample levels during repackaging the wastes from five T-hoppers into large drums during 1982 (2 years after receipt). The effort took about 20 days on the day shift for a total of around 100 hours. The implication is that the effort was interrupted for other routine processes. A "mistake" of "breaking up" materials outside the containers on a single shift on April 28, 1982, was corrected. Personnel wore airline masks, etc. This is a clear indication of the fact that the processing was not done all at once after 1980, but occurred as they identified materials satisfactory for blending.

### Evaluation of Plutonium Bioassay Data (Robinson 1989)

A variety of files dealing with:

- Evaluation of bioassay analytical data, p. 2, including detection limits, p. 9;
- Responses to 1985 Task Force Question set, p. 20, see Table 1, p. 25, and Bonfer response on p. 50, note Table I, p. 51;
- POOS blending processes, see pp. 61, 63, and 86;
- Precautions during blending, see p. 68; authorization to process >20 ppb U Pu in 1985;

### ATTACHMENT A

### RATIONALE FOR 400 PPB U PU FOR 10A PROCESS STREAM (continued)

- Summaries of plutonium levels by year;
- Neton recommendations, p. 91; and
- Others.

### "POOS History and Risk Assessment" (Hinnefeld 1988)

See report starting on page 121 for another analytical description of the repackaging operation in Plant 4, including Table 1 on page 123. Again, this is documentation of the results of this particular operation of repackaging 5 of the 16 T-hoppers.

## DOE Contact Report, Processing 168 MTU of UO<sub>3</sub> containing Recycle Contaminants (Jelinek and Thomas 1985)

This is documentation in 1985 (3 years after the repackaging of the five T hoppers; at that point it was through the Plant 2/3 and at 26 to 34 ppb U Pu. There is a hazards analysis with a call for bioassay, etc.

### Interview with Former FEMP employees (ORAUT 2008b)

A description of blending PGDP ash (p. 21). It was sampled and milled in Plant 1 then blended in Plant 4. Statement made that Plant 1 equipment was not "large scale" for blending.

### "Discussion of and Questions Relating to the Specific Processes and Time Required to Down Blend the 10A Sub Group Process" (ORAUT 2011b)

A former Fernald employee suggested that his recollection was that the POOS materials were dissolved in nitric acid in the Plant 2/3 front end and stored as UNH until suitable blending materials could be used to meet processing limitations.

### Former Fernald Employee, September 9, 2011 (ORAUT 2011c)

A worker indicated that his recollections were not exact, but that the nature of the materials (nonflowing, etc.) would require milling in Plant 1 and some processing in Plant 8 before final processing in Plant 2/3.

### Interview with Former Fernald Employee, October 4, 2011 (ORAUT 2011d)

To his recollection, the repackaging and blending operation was conducted in Plant 4 because this facility had the equipment and ability to handle the large containers. The containers were emptied into a receiver on the fourth floor, which fed in an enclosed transfer facility to the receivers on the first floor. All of the equipment was enclosed and vented to filters. The only contamination occurred during unusual leaks or spills. He indicated that while he did not recall specifically, he would estimate that under normal conditions two or three hoppers could be processed per shift.

### Processing Uranium Containing Plutonium Above Target Limit (Author unknown undated a)

Specifically mentions training for the 168 MTU (UO<sub>3</sub>) at 42 ppb U and blending 67 MT with 67 MT of "in spec" material in Plant 4 (p. 5). Also mentions 75,000 gal of POOS raffinate to be processed. Page 6 discussion of FMPC 20 ppb U target limit, then DOE set target limit at 10 ppb U, with the materials to process at FMPC at 42 ppb U.

## Collection of Reports and Data Including Discussion of Early RU Programs (Author unknown undated b)

Statement of blending 1976 PGDP wastes with the last of the ore concentrates on p. 128. Page 139 shows a 1999 spreadsheet listing of analytical results of T-hopper T-449 container (which had the

highest activity levels) with an average of approximately 7,000 ppb U (multiple sampling of each container, inhomogeneous results, two containers above 7,000 ppb U, other two below 1,000). A table of document references for enriched RU programs at FMPC is listed on pp. 193–198. Page 365 shows a report of <sup>99</sup>Tc at FEMP.

## Type C Investigation: Discovery of Out-Of-Specification Plutonium Bearing Material During Inspection and Handling of Plant 1 Pad Residues for Waste Shipment (WMCO 1988)

Drums of 1977 PGDP wastes that had been mislabeled and set aside. Levels were 17.6 ppb U.

### POOS Handling and Urinalyses Hazards Analyses (Traub 1985)

1985 analysis by Pacific Northwest National Laboratory about change in maximum permissible concentrations based on contaminants.

### Plant 4 Smear Results from October to November, 1986 (WMCO 1986c)

Pre- and post-POOS processing sample results.

### "POOS Raffinate Spill" (Walker 1986)

This reference contains a technical discussion of 40 ppb U versus 10 ppb U.

### ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES

### **TABLE OF CONTENTS**

SECT	ION <u>TITLE</u>	<u>PAGE</u>
B.1	Introduction	86
B.2	Thorium Process Inventories and Thoron Potential	88
B.3	Thorium Waste Repository Locations and Quantities	89
B.4	Factors to Consider in Determining Potential Alpha Energy Concentration	90
B.5	NIOSH Responses to SC&A Comments on Methods for Reconstructing Thoron Doses at Fernald Presented at the 12/4/14 Working Group Meeting B.5.1 Background B.5.2 Discussion B.5.3 Thoron Measurements B.5.4 Thoron Daughter WL Measurements B.5.5 Conclusion B.5.6 1954 Thoron/Thoron Daughter Results	95 95 97 99 99 100 105
		109

### LIST OF TABLES

### TABLE

### TITLE

B-1	Historical thorium mass balance	
B-2	Thorium storage inventory, 1987	
B-3	PAEC for thoron and progeny	
B-4	Building dimensions and ventilation systems	
B-5	Summary of thoron exposure estimates	
B-6	Thoron exposure	
B-7	Summary of the 1996 through 1998 thoron measurement data	
B-8	Summary of WL measurements	
B-9	Three high measurements near Plant 8 silo and bins in 1986	
B-10	Locations and numbers of samples	
B-11	Thoron and thoron daughter results	

### LIST OF FIGURES

### **FIGURES**

### TITLE

### B-1 B-2 B-3 B-4 Measurements and lognormal fits, 1996......107 WL comparisons, 1986 through 2003......108 B-5 GA center of rod storage area......111 B-6 GA north end of rod storage area......112 B-7 GA TNT and sump cake storage area.....112 B-8

### PAGE

PAGE

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 85 of 188

Γ

### ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

B-9	GA wet area, main floor1	113
B-10	GA Wet area, top deck1	113

### B.1 Introduction

From the beginning of FMPC operations in late 1953 until the "completion of the disposition of the containerized waste inventory" (DOE 2007), there have been uranium and thorium processes (summarized in Table B-1) with feed materials and the associated waste materials that have resulted in elevated levels of 3.823-day half-life<sup>222</sup>Rn (radon) and 56-second half-life<sup>220</sup>Rn (thoron) along with their particulate progeny. Radon is the first progeny of <sup>226</sup>Ra, which was present in significant quantities in the raffinates from processing high uranium-bearing pitchblende ores. The elevated radon exposure levels at Fernald from this source are addressed in ORAUT-TKBS-0017-4 (ORAUT 2015b). This attachment addresses the potential exposures from elevated levels of thoron during processing and storage of thorium at Fernald during the post-SEC periods.

Primary					
buildings	Period	Daily	Annual	Total	Operations and materials
Plant 9	1954–1956	0.5–0.6	100–200	380	ThF <sub>4</sub> to metal and machining of cores
					for reactors. Wastes stored in Plants 1
					and 9 storage areas.
Plant 4	1954–1956	0.5–0.6	150–300	461	Thorium oxide conversion to ThF <sub>4</sub> for
					use in Plant 9.
Plant 6	1959–1963	0.02	10–20	80	Converted furnace for Plant 9 1954 to
					1956 Th recovery from scraps and
					waste.
Plant 1	1954–1979	<1	Unknown	Unknown	Receive, weigh, sample, and store
					source and special materials.
Pilot Plant	1954–1956	0.01–0.75	8–381	2558	68% of FMPC Th processed in Pilot
	and				Plant. Metal production, dense thoria,
	1964–1979				thorium nitrate to thoria gel, thorium
					hydroxide production, processes
					developed for scale up to production
					levels.
Plant 8	1965	0.025	92	303	ThF <sub>4</sub> to thorium hydroxide, thorium
	and				sump cake calcined, thorium residue
	1968–1972				processing to thorium hydroxide
Plant 2/3	1968	0.12	45	45	Short-term denitration experiment and
					uranium recovery from thorium
					hydroxide from plant 8.
Total	Not	Not	Not	3,827°	Not applicable
processed	applicable	applicable	applicable		
FMPC	1954–2006	Not	Not	65	Discarded type Super S thorium
waste pits	(closure)	applicable	applicable		material as waste

Table B-1. Historical thorium mass balance (MTTh).<sup>a,b</sup>

a. Sources: Dolan and Hill (1988); DOE (1987b); Mead, Savage, and Fugate (ca. 1986); Clark et al. (1989); Grumski (1987); Bogar (1987); Starkey (1964).

b. For perspective, a metric ton of pure thorium metal at the theoretical density occupies a relatively small volume. For example a 10- to 11-gal can (about 15 in. diameter and 15-in. tall) would weigh approximately 1,000 lb or 0.45 MT. Three of these relatively small cans could hold in excess of a metric ton of thorium.

c. Not including the unknown amount from Plant 1.

As illustrated in the decay diagram of <sup>232</sup>Th (Figure B-1), thoron is the second progeny of <sup>228</sup>Th and, in a couple of weeks after processing for thorium purification, can be considered to be in full equilibrium with the parent <sup>228</sup>Th. Thorium-228 is a third progeny of the long-lived <sup>232</sup>Th and generally in 40% to 65% equilibrium for materials processed at FMPC (Figure B-2). The degree of equilibrium is dependent on both (1) the decay of <sup>228</sup>Th without replenishment from the 5.7-year half-life <sup>228</sup>Ra after

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 87 of 188
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removal of the thorium progeny and (2) the time it takes the <sup>228</sup>Ra to build into equilibrium with <sup>232</sup>Th (Figure B-2). For the post-SEC period (after 1979), 65% equilibrium is assumed as the default. Thorium-228 is also a first progeny of <sup>232</sup>U, which could have been present in small quantities from recycled thorium materials from Hanford in the 1977 to 1979 process, but not in sufficient levels from this source to result in a significant change in the defaults defined in this attachment [6].



Figure B-1. <sup>232</sup>Th decay diagram, including the <sup>220</sup>Rn (thoron) decay chain.

Thoron was present and a portion released during the processing and storage of metric tons of a variety of forms of thorium. Although this radioactive gas and its short-lived progeny are present in variable levels as a natural background, elevated levels with those of radon have been identified and studied and associated with increased respiratory cancer potential (i.e., uranium miner studies and others listed).

The primary focus of Fernald radiological safety programs for thorium was to define the thorium air concentrations in air in the work place coupled with in vivo lung counts for thorium, the results of which provided the means of controlling worker exposures to levels below the then-current permissible levels (Tomes 2001). In addition the metric ton quantities of thorium in process (with the measured particulate releases), or in storage, provided sources of elevated thoron gas release with its subsequent progeny. The records indicate that there was an awareness of the potential for exposure to thoron and its progeny through documentation of reported "thoron" measurements and holding of the routine BZ and GA samples for up to 96 hours to allow the short-lived radon and thoron progeny to decay (Weaver 1987; Starkey 1964). This provided a more meaningful measure of the long-lived thorium concentrations (Starkey 1964). Because air activity sample analyses specifically for thoron progeny are insufficient to directly document the routine concentrations in the work places at all times, an approach for bounding the possible workplace concentrations of thoron and progeny is presented.



ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

Figure B-2. Decay and buildup of <sup>228</sup>Th and progeny after processing.

This attachment presents the methods and considerations of bounding the work place exposures to thoron and primarily the short-lived progeny, based on theoretical analyses and conservative assumptions.

### B.2 Thorium Process Inventories and Thoron Potential

During the 35-year operating history of the Fernald site, approximately 70% of the time (24 years) involved the production of tens to hundreds of MTTh parts and products on an annual basis for a total of over 3,800 MTTh (Table B-1). The production of that quantity of products required processing of approximately 4,200 MTTh, and leaving over 750 MTTh of scrap and waste materials (see Tables B-1 and B-5). On the surface this appears to be a large amount, although the primary material at FMPC was uranium. Several historical texts refer to amounts of thorium being "minor" in nature because the mass of thorium was generally less than 1% of the mass of RU receipts alone. Still, the tens to hundreds of metric tons of thorium in most of the FMPC facilities at any given time during the operational history was a recognized hazard and a source of thoron gas (<sup>220</sup>Rn), a very short-lived progeny of <sup>228</sup>Th (thoron has a half-life of 56 seconds). Table B-1 outlines the rough thorium mass balance and estimated inventories during the indicated periods.

As shown above, the processing averages during the 24 years of thorium operations were in the range of 15 to 20 MTTh/mo or <1 MTTh/d, while process feedstocks (quantities in the process facility preparatory to processing) could have been in the range of 2 to 15 MT levels. That is, during processing campaigns small quantities were probably placed in the process facilities temporarily in

queue for the operations [7]. Most of the storage was held in containers in adjacent storage buildings as well as barreled wastes held on pads outside the buildings

The primary products were metal cores for the Hanford Site and Savannah River Plant reactors in 1954 to 1956 and a wide range of chemical forms and compounds in the succeeding periods as indicated in Table B-1. Process scraps, out-of-specification product, etc. resulted and became a storage or process category (or both). The processes were also varied, ranging from chemical, metallurgical, metal machining, waste storage, repackaging, and shipping.

The primary process from 1977 to 1979 was unusual in that it was primarily disposal and storage of excess reprocessed thorium materials from Hanford. It involved 6,900-gal tanks of nitrate solution, each with 10.8 MTTh for conversion to a stable and therefore storable thoria gel. These tanks were received at a rate of up to two per month for a maximum process rate of 21.6 MTTh/mo, which is less than 1 per day with an overall total of 351 MTTh. Uranium-233 was a contaminant of the thorium at approximately 130 g per tank load or approximately 12 ppm Th. The total mass of 4.2 kg of <sup>233</sup>U was not separated from the thorium and was stored with the thorium (Grumski 1987).

The total mass inventory is only instructive in relation to the thoron inventory when the equilibrium ratio of <sup>228</sup>Th to <sup>232</sup>Th is known in addition to the fact that thorium-bearing materials were seldom 100% thorium. The theoretical activity of <sup>232</sup>Th is  $1.09 \times 10^5$  pCi/g, but for consistency with other reports, including the safety analysis report for the thorium storage facility, a specific activity for <sup>232</sup>Th and it progeny of  $0.989 \times 10^5$  pCi/g of thorium materials will be used as an upper limit, which accounts for lesser densities of thorium compounds and other materials in the product.

The equilibrium age of the thorium processed at FMPC ranged from recently separated to 1 or 2 years since separation of the nonthorium progeny [8]. Figure B-2 illustrates the disequilibrium that occurs when thorium is separated from or purified from its progeny. Radon-220 is a progeny of the 1.9-yr half-life <sup>228</sup>Th, which decreases to approximately 65% of equilibrium with <sup>232</sup>Th in 1 year after separation of the radium and actinium progeny and 43% in 5 years after separation. Full isotopic equilibrium of <sup>232</sup>Th and <sup>228</sup>Th can be assumed for long-term storage materials after 35 years or so. For the functional period, 65% will be assumed, which represents a material 6 months to 2 years after separation and should be adequately conservative. Assuming 65% equilibrium would make <sup>220</sup>Rn inventory equivalent to 0.64 × 10<sup>5</sup> pCi/g of <sup>228</sup>Th compounds in the processes, and assuming full equilibrium with each of its progeny.

### **B.3** Thorium Waste Repository Locations and Quantities

FMPC was appointed the DOE thorium materials repository in 1972, and thorium processing was shut down in 1979. During the storage period up to 2,300 MTTh were stored pending final disposal off site in Nevada and Utah. The repository thorium wastes and surplus thorium materials were accumulated from Fernald and other sites in the repository era from 1972 until the "completion of remediation and the disposition of the containerized waste inventory" or final closure in 2006. See Table B-2 for a tabulated summary as of 1987of 13,329 drums of thorium plus 184 MTTh in bulk powders (or granules) or liquid. Over 689 MTTh were stored in the Pilot Plant warehouse and 747 MT in four other designated storage buildings. The first shipment from this inventory was to the EnviroCare facility in Utah.

Plant	Storage		Net weight	Th weight		
area	building	Drums	(lb)	(lb)	% Th	MTTh
Plant 1	67	5,992	426,978	333,044	78	151
Plant 5	65	5,599	2,492,505	711,210	29	323
Plant 5	65W pad	240	105,032	92,955	89	42
Plant 8	Silo and bins	Bulk material	Not applicable	Unknown	50	175
Plant 9	64	181	128,345	123,141	96	56
Pilot	68	1,317	971,398	745,785	77	338
Plant						
Pilot	Liquid tank	Nitrate solution	Not applicable	Unknown	Not applicable	351
Plant	-					
Total	Not applicable	13,329	Not applicable	Not applicable	Not applicable	1,436

Table B-2. Thorium storage inventory (MTTh), 1987.ª

a. Sources: Dolan and Hill (1988); DOE (1987b); Mead, Savage, and Fugate (ca. 1986); Clark et al. (1989); Grumski (1987); Bogar (1987); Starkey (1964).

### B.4 Factors to Consider in Determining Potential Alpha Energy Concentration

The primary dose from thoron is to the respiratory tract organs and is delivered primarily by the shortlived progeny (Table B-3). Due to the short radiological half-lives and lung clearances of the particulate progeny, there are no practical bioassay measurements to define intake (NIOSH 2013; UNSCEAR 1994; Bodansky, Robkin, and Stadler 1987; Evans and Goodman 1940; Strom and Reif 1996). Therefore, the method of choice for dose evaluation is to measure the potential alpha energy exposure (PAEE; see Glossary), which is the potential alpha energy concentration (PAEC) in air multiplied by the time the worker was exposed. The PAEC is measured in working levels (WLs), and the PAEE is measured in WLM. The bounding defaults recommended in this section are listed as WLM, which relate directly to total alpha energy exposure.

				PAEC
Isotope	Half-life	Atoms/7.5 pCi	α MeV/atom	(MeV/7.5 pCi)
Rn-220	56 s	23	14.6	335
Po-216	0.15 s	<1	Included with Rn-220	Included with Rn-220
Pb-212	10.64 hr	15,476	7.8	1.21E+5
Bi-212	60.6 min	1,469	7.8	0.12E+5
Po-212	0.3 µs	Not required	Included with Bi-212	Included with Bi-212
TI-208	3 min	73	No alpha	0
Total	Not applicable	Not applicable	Not applicable	1.33E+5

Table B-3. PAEC for thoron and progeny.

### **Air Monitoring**

From the beginning of operations in the early 1950s the radiological safety program evaluated <sup>222</sup>Rn and its progeny in work areas with specific radon and progeny measurements. The health and safety staff was aware of thoron dose potential as evidenced by a number of samples and references to specific thoron analyses, which have been recovered or mentioned in staff reports (Weaver 1987; Starkey 1964). During the period after 1989, when WEMCO assumed the contractor role at the Fernald site, extensive and recorded radon and thoron monitoring was performed across the site (Weaver 1987). Air monitoring for particulate activity was also performed throughout the production period, and enough air sampling data has been recovered for realistic conclusions and assumptions about exposure potentials.

### **Work Place Control Functions**

To better understand the workplace ventilation dynamics and air movements, Table B-4 provides a summary of the dimensions of the buildings in the Fernald complex that handled thorium in any amount. The process ventilation systems, which include an air moving system for maintaining a negative pressure within the process equipment, a baghouse particulate filter (a few liquid scrubbers were also used), and individual stacks for release of filtered air to the environment. It was reported that there were a total of 94 of these ventilation systems and stacks during the early periods, the number of which changed with changing processes and projects. A total of 114 ventilation systems is indicated in Table B-4, and the total stack flow for each building is a summary of all the individual stack flows in that building. Although there were some general building vents for assistance in temperature reduction, these were not used continuously. The air supply for the stack discharge flows came from the building working environment around the containment systems, which in turn enclosed the process equipment. The pressure differential maintained the airflow into the equipment in which the thorium was being processed. As indicated in Table B-4, the air changes per hour for the entire facility volume would average approximately three when all the individual ventilation systems were operating. The air change rate in the immediately vicinity of the individual processes would obviously be higher (Dolan and Hill 1988; DOE 1987b; Mead, Savage, and Fugate ca. 1986; Clark et al. 1989; Grumski 1987; Bogar 1987; Starkey 1964).

			Volume	Number of	Total	Total facility
Building	Dimensions (ft)	Volume (ft <sup>3</sup> )	(m <sup>3</sup> )	systems	(ft <sup>3</sup> /min)	chgs/hr
Plant 1	100 × 160 × 60	9.6E+5	2.72E+4	15	6.1E+4	3.8
Plant 2/3	60 × 380 × 67	1.53E+6	4.33E+4	6	3.2E+4	1.3
Plant 4	225 × 165 × 94	3.49E+6	9.88E+4	12	4.38E+4	0.8
Plant 5	650 × 100 × 52	3.38E+6	9.57E+4	17	1.5E+5	2.7
and Building 55	60 × 30 × 51	0.92E+5	2.61E+3	1	4.0E+3	2.6
Plant 6	500 × 200 × 39	3.9E+6	11.0E+4	6	1.35E+5	2.1
Plant 7	110 × 80 × 114	1.0E+6	2.83E+4	4	1.7E+4	1.0
Plant 8	60 × 260 × 48	7.49E+5	2.12E+4	24	8.22E+4	6.6
Plant 9	300 × 225 × 40	2.7E+6	7.65E+4	4	7.89E+4	1.8
Pilot Plant	210 × 235 × 54	2.66E+6	7.53E+4	9	4.37E+5	9.9
Building 64	50 × 320 × 22	3.52E+5	1.0E+4	8	1.6E+4	2.7
Building 65	50 x 210 x 22	2.31E+5	0.65E+4	8	1.6E+4	4.2
Average or	Not	Not	5E+4	114	Not	3.3
total	applicable	applicable	average	total	applicable	average

Table B-4. Building dimensions and ventilation systems.

### **Release Fraction of Stored Materials**

Four examples are provided to address the release fraction (RF) of both <sup>222</sup>Rn and <sup>220</sup>Rn gases and provide a basis for estimating the release fraction of radon gases to the work environs:

 To bound the release of radon gas from a source of stored barrels of known waste concentration, measurements taken in a shipping car (~6 × 104-L volume) with 48 barrels of raffinate wastes with a 226Ra concentration of 400 nCi/g were used. This is the maximum measured isotopic characterization of silo waste – determined at a much later date. A level of approximately 25 Ci of 226Ra was calculated as the inventory in the 48-drum waste shipment. The maximum measured 222Rn progeny concentration (the highest of a number of samples) in the transport carrier was 400 pCi/L in the unventilated shipping car. Using these values, the de facto 222Rn RF from the drums is calculated to be 400 pCi/L ÷ (2.5 × 1013 pCi/6 × 104 L) = 1 × 10-6. These shipping containers were "enclosed containers," but obviously not vapor tight.

This is a measurement of 222Rn with a 3.8-day half-life and the associated higher diffusion capabilities, in comparison with 220Rn, which would have shown a much lower RF.

- 2. Another example and specifically for short-lived thoron gas release is found in the maximum measured 220Rn progeny levels in Building 65 (0.7 x 104 m3) in 1996. Building 65 is one of the primary DOE repository storage facilities and the maximum of many measurement concentrations was 267 pCi/L. Assuming this measurement was the progeny of 220Rn, the radon level would be 1.3 x 104 pCi/L. The facility contained approximately 421 MTTh at the time of the measurement with the associated 27 Ci of 220Rn. Some of the containers had not been double packed, and the lower level drums were in poor repair corroded and leaking. Total release of the 220Rn inventory would result in 2.7 x 1013 pCi/0.7 x 107 L = 4 x 106 pCi/L for an RF of 1.3 x 104 ÷ 4 x 106 = 3 x 10-3. There are many unknowns in this example: ventilation of the building, fraction of the stored material containers that were breached, location of the air sample in relation to leaking drums, etc. Therefore, it is not be used as a quantitative example, but rather to indicate that the RF for some storage facility operations is be in the 1 x 10-3 range and is more likely to be comparable to the storage silo example below.
- 3. A third example can be demonstrated through measurements made at the top of the Plant 8 3,000-ft3 storage silo (an elevated cylinder, 17 ft in diameter and 23 ft high with conical sections on the bottom and top) with approximately 85 MTTh. Thorium oxide and hydroxide granular material at approximately 50% thorium had a measured density of 200 lb/ft3 in the top of the oxides, which provides a thorium density of 100 lb/ft3. An air activity concentration of 71 WL was measured at the top of the tank, which had a 15-ft high by 17-ft diameter air space in the tank above the thorium materials (1 × 105 L). The cone-shaped surface area at the top thorium material (it was deposited by a conveyor to the center of the silo) was calculated as approximately 350 ft<sup>2</sup>.
  - With a diffusion distance of approximately 2 cm (Banjanac et al. 2006; and assumed 1 in.), the effective thoron quantity available for diffusion to the headspace was calculated as:
  - $(350 \text{ ft}^2)(1/12 \text{ ft})(100 \text{ lb/ft}^3)(454 \text{ g/lb})(0.64 \times 10^5 \text{ pCi/g}) = 8.5 \times 10^{10} \text{pCi}$
  - This compares to  $(85 \text{ MTTh})(6.4 \times 10^{10} \text{ pCi/MTTh}) = 5.4 \times 10^{12} \text{ pCi}$  in the total silo or 1.6% of the <sup>220</sup>Rn available for release.
  - The maximum expected <sup>220</sup>Rn in the 1 × 10<sup>5</sup>-L headspace volume is  $(8.5 \times 10^{10} \text{ pCi}) \div$  1 × 10<sup>5</sup> L = 9 × 10<sup>5</sup> pCi/L (total <sup>220</sup>Rn in 1 in. of the surface).
  - This is compared to a measured 71 WL= (375 pCi/L  $^{220}$ Rn/WL)(71 WL = 3 x 10<sup>4</sup> pCi/L.
  - This results in an effective RF of  $(3 \times 10^4) \div (9 \times 10^5 = 3 \times 10^{-2})$  of that within 1 in. of surface.
  - Therefore, the RF considering the total <sup>220</sup>Rn (from 85 MTTh) in the silo is  $(3E+4) \div (5.4 \times 10^{12}) \div (1E \times 10^5) = 6 \times 10^{-4}$ .

From this example we see that the Plant 8 storage silo is essentially a big barrel of thorium, and the amount of 220Rn in the headspace would be on the order of  $1 \times 10-3$  to  $1 \times 10-4$  of

the total in the container. Migration of the 220Rn from the container (big or small) to the work environment would be in the  $1 \times 10-6$  range (reasonably estimated based on 222Rn migration from barrels of waste).

4. The fourth example is found in the Plant 8 elevated double-bin storage tanks. Similar to the silo example above, a lesser RF is calculated, which further verifies the conservatism in the assumptions.

Therefore, it is judged to be conservative to use an RF of  $1 \times 10-6$  for stored inventories in intact but unsealed containers (10- to 50-gal drums, etc.) and  $1 \times 10-4$  for areas immediately in the vicinity of open thorium materials such as silo storage, leaking drums in closed buildings, etc.

### **Release Fraction of Materials in Process**

For process areas the RF can be conservatively calculated based on general thorium particulate air activity in comparison with the total thorium material in process. Routine air sampling in the operating areas recorded thorium concentrations ranging from tens to a few samples in the tens of thousands of dpm/m<sup>3</sup> and mostly in the very early period of 1954 to 1955 when processes were being developed. It could then be assumed that a 1-MT ( $1 \times 10^6$ -g) daily process load in a  $5 \times 10^4$ -m<sup>3</sup> volume process building in which process releases had resulted in  $1 \times 10^4$  dpm/m<sup>3</sup>. However, it is not reasonable to assume that the air concentrations in the building would have been at the maximum levels throughout the entire area, and these levels should be assumed to exist in a 20-ft hemispherical air space near the process equipment (475 m<sup>3</sup>). This is based on the fact that the higher value air samples used were undoubtedly BZ samples taken during high-level maintenance (e.g., clean-out evolutions, etc.). For these reasons it is reasonable to assume that the air samples represented maximum concentrations in the built of the equipment rather than average building concentrations. Therefore, the concentration in the air, if all the process materials were completely released, would be ( $1 \times 10^6$  g) ( $0.64 \times 10^5$  pCi/g) (2.22 dpm/pCi)  $\div 475$  m<sup>3</sup> =  $0.3 \times 10^9$  dpm/m<sup>3</sup>. The RF would be ( $1 \times 10^4$ )( $0.3 \times 10^9$ ) =  $3 \times 10^{-5}$ .

Some injection operations (dumping barrels of feed material into hoppers, etc.) could be expected to release the free thoron in the headspace of the container, which would be a ratio of 2 cm to the height of the materials in the container – in the range of 0.1 to 0.01 as indicated in the discussion of release fractions from stored materials. Even then the particulate dusts would serve as an absorption and removal mechanism of the progeny and clear in a short period of time (limiting the exposure time). An RF of 0.01 was assumed. There was a variety of process equipment that was used, including liquid extraction vessels, furnaces, mixers, etc., all of which were designed for containment.

### Equilibrium of Thoron and Progeny <sup>212</sup>Pb, Bi-212, Po-212 and TI-208

Quoting from Harley and Chittaporn (2006) of the New York University School of Medicine, "The inhaled solid decay products of thoron deposited on the lung airways deliver the bronchial dose." Harley (2004) also reports:

... the  $F_{eq}$  [fraction of equilibrium] cannot exceed 0.04 and calculations of  $F_{eq}$  for indoor radon and thoron confirms the values of 0.4 and 0.02. The value of  $F_{eq}$  for thoron is supported by two large data bases, one reported by NIRS [National Institute of Radiological Science] with measurements in China and one reported by NYU [New York University] in residences and also at the former uranium processing facility at Fernald, OH.

Pillai and Paul (1999) reported equilibrium values of thoron and its progeny of 0.002 to 0.007 in a monazite processing plant in India. This analysis uses 0.02 as adequately bounding.

### Summary Production Rate and Available Thoron Inventory for Release

As indicated above the average daily production rate was <1 MTTh (ranging from 0.03 to 0.8) with  $6 \times 10^{-2}$  Ci thoron. Temporary storage in the production facilities during processing campaigns could be 5 to 15 MTTh with the associated 0.3 to 0.9 Ci thoron. DOE long-term storage at the Fernald site was 100 to 450 MTTh in any of the storage facilities with the associated 6 to 27 Ci thoron.

### **Occupancy Time**

The nature of storage facilities results in a minimal occupancy time for required functions. This analysis assumes 500 hr/yr (or 3 mo/yr) for routine storage conditions accompanied by required repackaging. For long-term storage of high-integrity storage containers, 1 mo/yr is assumed. The time of occupancy during production periods will be assumed as 1750 hr/yr, because an average 1-MTTh continuous production rate is assumed. At this 1-MTTh daily rate, the recorded annual quantities processed would be completed in just a fraction of a year.

### **Demonstrated Local Diffusion Factor**

It has been demonstrated that for thoron progeny, the concentrations are approximately a factor of 10 higher near the source of the release than the concentration at a distance of 3 ft (Stranden 1980). Because this is probably dependent on ventilation, this demonstrated reduction factor to the BZ of workers is ignored.

### **Respiratory Protection**

It was established practice to wear respiratory protection during the processing of large volumes or masses of hazardous materials. The wearing of respirators would remove essentially all of the progeny through filtration and electrostatic attraction in the filter. However, in the interest of favorability to the claimant due to a few recorded cases of procedure violation (there were sample sheets and other logs that made note of personnel not wearing masks), it is assumed that no respirators were worn. The notes themselves indicate an unusual condition. Therefore, this is an admitted overly conservative assumption based on both (1) the established protection policies and practices and (2) demonstrated difference in calculated intakes of thorium based on air monitoring results in comparison with in vivo measured lung burdens.

The following summarizes the assumptions for thoron exposures:

- 1. Materials in process averaged less than 1 MT/d and less than 400 MT/yr.
- 2. Materials in process facilities in temporary storage ranged from 2 to 15 MT, and 15 is assumed.
- 3. Long-term DOE storage ranged from 100 to 450 MT in any given storage location, so 450 is assumed.
- Specific activity of thoron in thorium materials at Fernald is 0.64 × 10<sup>5</sup> pCi/g (e.g., <sup>228</sup>Th/<sup>232</sup>Th = 0.65).
- Facility volumes were large (averaging 5 × 10<sup>4</sup> m<sup>3</sup>). In the interest of conservatism, it is assumed that the mixing or exposure volume was near the process equipment and represented approximately 20-ft radius, hemispherical volume in the total facility (475 m<sup>3</sup>).

- 6. RF in temporary or long-term storage is  $1 \times 10^{-6}$ .
- 7. RF in temporary open storage or with compromised containers is  $1 \times 10^{-3}$  to  $1 \times 10^{-4}$ .
- 8. RF in general process areas is 1 × 10<sup>-5</sup>; 1 × 10<sup>-4</sup> was assumed for Plant 9 during process development period in 1954 to 1955.
- 9. Fraction of equilibrium (*F*<sub>eq</sub>) for thoron is 0.02. In a plant configuration with large buildings and engineered ventilation, the recommended value of 0.02 is judged to be conservative. In this case, it would require a concentration of <sup>220</sup>Rn/<sup>216</sup>Po at 375 pCi/L to produce <sup>212</sup>Pb/<sup>212</sup>Bi concentrations at 7.5 pCi/L and therefore result in 1 WL.
- 10. Diffusion factor of thoron progeny from source release point is 1 (no reduction). Fraction of progeny near source compared to 3 ft away is 0.1, but 1 is assumed.
- 11. Occupancy time in storage facilities is 500 hr/yr and 1 mo/yr for long-term storage.
- 12. Occupancy time in process facilities is a maximum of 1,750 hours.
- 13. Protective respiratory protection is assumed not to have been used.
- 14. Thoron WL is 7.5 pCi/L with 100% equilibrium with progeny, which is  $1.3 \times 10^5$  MeV/L (Table B-3. WL = 375 pCi/L <sup>220</sup>Rn and <sup>216</sup>Po to produce measured progeny <sup>212</sup>Pb and <sup>212</sup>Bi at 7.5 pCi/L (see the discussion of  $F_{eq}$  factor of 3 and summary item 9 above.)

Estimates of possible exposures to thoron indicate that for some periods and in some locations (primarily long-term storage areas in which some extended work periods were required), exposures could be as high as 1.6 WLM/yr. The work could involve activity with poorly ventilated facilities with open containers, such as repackaging failed containers, preparing for shipment, etc. Even though the RFs are relatively small for both storage configurations and operational conditions and the short half-life of <sup>220</sup>Rn limits the diffusion range in all materials to 2 cm or less, possible quantities and exposures can still be of concern. UNSCEAR (1994) indicates that due to the relatively longer half-lives of the progeny of thoron in comparison with radon, significant dose can be experienced with relatively small WLM exposures.

Table B-5 provides a comparative summary of the WLM intake estimates during the entire operational period including the SEC periods. Table B-6 provides a summary of the WLM intake of <sup>220</sup>Rn progeny that could have occurred during processing of thorium in the post-SEC periods. The 1.6-WLM estimate for work in the storage facilities in preparing thorium materials with higher integrity containers and preparations for shipment is conservative. The work would require a very limited time and not the entire indicated 17-year storage period. In addition, the work was recognized as requiring special controls including protective clothing, respiratory protection, and ventilation.

### B.5 NIOSH Responses to SC&A Comments on Methods for Reconstructing Thoron Doses at Fernald Presented at the 12/4/14 Working Group Meeting

### B.5.1 Background

At the Fernald Working Group meeting on September 4, 2014, NIOSH presented a white paper called *Fernald Dose Reconstruction Methodology for the Post Special Exposure Cohort (SEC) Period, 1979-*

2006 (NIOSH 2014c). In addition to thorium recommendations, the paper included thoron exposure recommendations (WLM per year), based on the information from Sections B.1 to B.4 to generate Tables B-5 and B-6.

	Table B-5.	Summary	/ of thoron	exposure	estimates. <sup>a</sup>
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	Available	Reduction and	Effective facility	Rn-220/ Po-216	Pb-212/ Bi-212	
Deried and leastion	thoron	calculation	volume	activity	activity	
			(L)"			
1954–1955, Plant 9 and Plant 4	1  IVI I h/d =	RF 1E-4	5E+5	12.8	0.034	0.35
In process per day	6.4E+10	F <sub>eq</sub> 2E-2				
	pCi/d Rn-220	Focc 10.2 mo/yr				
Available for release	15  MTTh/d =	RF 1E–6	5E+5	2	5.3E–3	0.016
	1E+12 pCi/d	<i>F</i> <sub>eq</sub> 2E–2				
	Rn-220	F <sub>occ</sub> 2.9 mo/yr				
1959–1963, Plant 6	0.02 MTTh/d	RF 1E–5	5E+5	0.026	3.5E–3	Not
In process per day	= 1.3E+09	<i>F</i> <sub>eq</sub> 2E–2				signifi-
	pCi/d Rn-220	Focc 10.2 mo/yr				cant
Available for release	2 MTTh/d =	RF 1E–6	5E+5	0.26	NS	Not
	1.3E+11 pCi	<i>F</i> <sub>eq</sub> 2E–2				signifi-
	Rn-220	Focc 2.9 mo/yr				cant
1965 and 1968–1972, Plant 8	0.03 MTTh/d	RF 1E–5	5E+5	0.04	NS	Not
In process per day	= 2E+09	<i>F</i> <sub>eq</sub> 2E–2				signifi-
	pCi/d Rn-220	F <sub>occ</sub> 10.2 mo				cant
Available for release	2 MTTh/d =	RF 1E–6	5E+5	0.26	NS	Not
	1.3E+11 pCi	<i>F</i> <sub>eq</sub> 2E–2				signifi-
	Rn-220	Focc 2.9 mo/yr				cant
1954–1956 and 1964–1979, Pilot Plant	0.8 MTTh/d =	RF 1E–5	5E+5	1	0.0027	0.027
In process per day	5E+10 pCi/d	F <sub>eq</sub> 2E–2				
	Rn-220	Focc 10.2 mo/yr				
Available for release	15 MTTh/d =	RF 1E–6	5E+5	2	0.0053	0.016
	1E+12 pCi	F <sub>eq</sub> 2E–2				
	Rn-220	Focc 3 mo/yr				
1954–1989, various storage sites	300 MTTh =	RF 1E-4	1E+7	200	0.53	1.55
Available for release	2E+13 pCi	F <sub>eq</sub> 2E–2				
	Rn-220	Focc 3 mo/vr				
1972–2006 .final closure storage	300 MTTh =	RF 1E–4	1E+7	200	0.53	0.53
Available for release	2E+13 pCi	F <sub>eq</sub> 2E–2				
	Rn-220	Focc 1 mo/vr				

a.  $F_{eq}$  = fraction of equilibrium;  $F_{occ}$  = occupancy time; NS = not significant.

b. For maximizing purposes, the mixing of the thoron and progeny during the processing of thorium was assumed to be in a 20-ft hemispherical volume immediately around the release point, which in turn is assumed to be the process workstation and for the short-term storage of thorium in the process facilities. For long-term storage facilities, the volume of the storage facilities was used.

Table B-6.	Thoron	exposure.
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Period	Plant	WLM intake each year during period
1954–1956	Plant 4 and 9	0.4
1959–1963	Plant 6	No significant dose

Period	Plant	WLM intake each year during period
1965 and	Plant 8	No significant dose
1968–1972		_
1977–1979	Pilot Plant	0.03
1972–1989	Storage facilities, repackaging, etc.	1.6
1972-2006	Closure, various storage	0.5

Discussions at the December 4, 2014 Fernald Working Group meeting included a Sanford Cohen & Associates (SC&A) presentation, SC&A Review of Proposed NIOSH Methods for Reconstructing Thorium Doses at Fernald (1979–2006) (SC&A 2014). Slides 32 and 33 of the presentation list SC&A's summary and critique of NIOSH's "Major assumptions" regarding thoron dose reconstruction:

- 1. The white paper appears to contradict itself on the assumption of 300 MT thorium in the storage sites (the preceding page appears to quote 450 MT, the introduction quotes over 2,000 MT of thorium materials in addition to the Plant 8 silos and bins and Pilot Plant storage).
- 2. The release fraction should be better established. Quoted release fractions in the white paper appear to range from  $1 \times 10^{-6}$  all the way to  $1 \times 10^{-3}$ .
- 3. The equilibrium factor of 0.02 (or 2%) is not well founded. The stated reference indicates equilibrium factors could range from 2% to 10%, but also notes "more precise studies are warranted."
- 4. SC&A could not determine the rationale behind occupancy times of 3 months (up through 1989) and 1 month (during final closure).
- The specific activity of thoron was given as 6.4 × 10<sup>-4</sup> pCi/g assuming exposures occurred 6 to 12 months after separation and an equilibrium fraction of <sup>228</sup>Th/<sup>232</sup>Th of 0.65. The equilibrium fraction of materials stored in Building 65 was at least 0.95.

A related item under "Main Conclusions" on Slide 34 states:

The main parameters for estimating exposure to thoron should be better described and established to assure that thoron exposures are calculated in a scientifically defensible, claimant-favorable and/or bounding approach.

### B.5.2 <u>Discussion</u>

The information from Sections B.1 to B.4 to generate Tables B-5 and B-6 were not intended to provide precise estimates. Nor is it even possible to do so because of the general lack of site-specific thoron data by time and location. Rather, it was developed to provide a reasonably conservative estimate of the exposures. It is clear from the site documents collected that thoron was recognized as a potential problem from the start of thorium operations. There are numerous references to thoron in site documents. For example, see the measurements in Plant 9 from 1954 that are discussed in the following sections (NLO 1954). In March 1980, a standard operating procedure entitled *Radiological Controls for Repackaging Thorium* specified that <sup>220</sup>Rn working-level samples be taken at least twice per shift (WMCO 1990c, p. 6). In 1998, the Plant 9 Project Radiation Protection Plan specified that a minimum of 25% of the workers in each work group/crew (minimum of 1 worker) would be required to wear a pump and filter assembly to monitor for radon and thoron (FEMP undated, p. 112).

## <u>NOTE</u>: The comments are addressed in the following sections. Comments 2 and 4 are addressed together, after Comments 1, 3, and 5.

<u>SC&A Comment 1 (Thorium Inventory)</u>: The white paper appears to contradict itself on the assumption of 300 mT thorium in the storage sites (the preceding page appears to quote 450 mT, the introduction quotes over 2000 mT of thorium materials in addition to the plant 8 silos and bins and pilot plant storage).

NIOSH Response: SC&A's statement that the introduction quotes over 2,000 MT of thorium materials apparently refers to the total of the net weights in Table B-2. This table shows the estimates for various storage locations. The MT weight in storage locations is the source of the discrepancy between the 450 MTTh in Sections B.1 to B.4 and the 300 MTTh used in the calculations for storage areas. The data in Table B-2 come primarily from Dunaway (1990). These same data are also in Collier (1987) and Weinreich (1987). The 175 MTTh in the Plant 8 Silo and bins is from the 1985 data in NLO (1965–1988). The 351 MTTh in the Pilot Plant "Liquid Tank" represents the thorium nitrate shipped from Hanford in the late 1970s. There are various descriptions of the quantity of thorium nitrate in the site documents (e.g., Bonfer 1988; FMPC 1952-1992; Dunaway 1977). Most of the material was converted into dried thoria gel. There were 6,500 gal of thorium nitrate remaining in 1987, as noted below. Documents also vary on the quantity of thoria gel produced (see Dolan and Hill 1988; RCK 1997; Heatherton 1977; Audia 1979). The 338.3 MTTh quantity in Audia (1979) matches the quantity stored in Building 68 in 1987 (Dunaway 1990). In 1987, only 9.2 MT of thorium in thorium nitrate was stored in Tank T-2 at the Pilot Plant (NLO 1985, p. 176). Therefore, the use of 351 MTTh represents a bit of double counting of some of the thorium. The value of 450 MTTh represents an upper bound on the thorium stored in any one location, but the 300 MTTh quantity used in the calculations better represents a more typical storage quantity. Using the 450 MTTh estimate with the existing assumptions would result in a 50% increase in the thoron and thoron daughter concentrations to 300 pCi/L and 0.8 WL, respectively. As will be seen in the measurements presented below, this increase would not affect the conclusion of this appendix.

## <u>SC&A Comment 3 (Equilibrium Factor)</u>: The equilibrium factor of 0.02 (or 2%) is not well founded. The stated reference indicates equilibrium factors could range from 2%–10%, but also notes "more precise studies are warranted."

<u>NIOSH Response</u>: SC&A correctly states that the range of equilibrium factors between thoron and its daughters was reported by the site to range from 0.02 to 0.10 (Daniels 1995, p. 13). It is not clear how these values were determined. In a 2006 paper, Harley et al. reported that the indoor thoron equilibrium factor is 0.02 and cannot exceed 0.04. The large diffusion coefficient of the very short-lived <sup>216</sup>Po results in a high removal rate to surfaces. The value of the equilibrium factor for thoron was supported by two large databases, one of which included measurements made at Fernald (Harley and Chittaporn 2006).

# <u>SC&A Comment 5 (Assumption of <sup>228</sup>Th/<sup>232</sup>Th Equilibrium Fraction)</u>: The specific activity of thoron was given as $6.4 \times 10^4$ pCi/g assuming exposures occurred 6–12 months after separation and an equilibrium fraction of Th-228/Th-232 of 0.65. The equilibrium fraction of materials stored in Building 65 was at least 0.95.

<u>NIOSH Response</u>: The estimates were generalized to cover all areas over the site's history. There are individual examples that do not fit the conditions assumed in that discussion. This comment points out one of them. However, using the 323 MTTh stored in Building 65 with a <sup>228</sup>Th-to-<sup>232</sup>Th equilibrium fraction of 0.95 would result in only a 50% increase in the thoron and thoron daughter

concentrations to 300 pCi/L and 0.8 WL, respectively. As with the 450 MTTh example in the response to Comment 1, this increase would not affect the conclusion within this appendix.

## <u>Comment 2 (Release Fraction)</u>: The release fraction should be better established. Quoted release fractions in the white paper appear to range from 10<sup>-6</sup> all the way to 10<sup>-3</sup>.

## <u>Comment 4 (Occupancy Time)</u>: SC&A could not determine the rationale behind occupancy times of 3 months (up through 1989) and 1 month (during final closure).

<u>NIOSH Response</u>: Rather than focus on SC&A's immediate concerns about release fractions and occupancy times, this response seeks to demonstrate that the estimates provided are consistent with actual measurements that NIOSH has analyzed. It is acknowledged beforehand that this comparison will not be totally satisfying. Because the measurements are either in terms of thoron concentration in pCi/L or thoron daughters by WL, the comparison involves comparing dose rates, not total doses. However, if the dose rates are shown to be as low as, or lower than, the estimates, the discussion will be moved forward. Then, perhaps some agreement can be reached on occupancy factors for select populations. In the discussion below, thoron measurements and thoron daughter measurements will be addressed separately.

There are some issues with the available data. Most of the data is from 1986 and later. These data have the disadvantage of not covering the earlier periods. The period before 1979 is already included in the SEC, which means that the respiratory track cancers are already being compensated for much of the site's history. On the other hand, many of the later measurements were made with improved technology, such as track-etch detectors and WL monitors, which likely collected more and better data than would have been possible in the early years. However, even some fairly recent WL measurements were done with short-term (on the order of minutes) air sample measurements that were recounted to determine the contribution from thoron daughters. This probably resulted in greater variability in the measurement values. Many of the later-period measurements were taken in locations where thoron levels were known to be a potential problem which likely resulted in the mean of the measurements being higher than the true site mean.

### B.5.3 Thoron Measurements

Thoron calculations based on air samples collected in 1954 for Plant 9 are in NLO (1954). The concentrations were recalculated from the data on sample worksheets. They are discussed in Section B.5.6. The thoron concentrations were  $21.8 \pm 17.2$  pCi/L before the ventilation was turned off. The 12.8 pCi/L estimate in Sections B.1 to B.4 lies within one standard deviation of these results. The underlying thoron daughter measurements will be discussed below.

Thoron measurements for 1996 to 1998 have been recovered. The measurements were obtained with track-etch dosimeters and were reported in pCi/L. The dosimeters were calibrated by placing two types of dosimeters in a chamber and exposing them to known concentrations of radon and thoron. Both dosimeter types prevented the entry of the daughter products. One type ("filter") allowed both radon and thoron gas to enter the dosimeter and expose the CR-39 (or equivalent) plastic. The other type ("membrane") retarded the infiltration so that most of the thoron would decay before exposing the CR-39, resulting in a measurement of radon only. The difference in the two readings (in tracks per unit area) was interpreted as the thoron exposure in pCi/L (Pearson and Spangler 1991).

The track-etch dosimeters were deployed for at least one quarter (3 months). Generally, over 40 locations were measured simultaneously, including both high- and low-exposure-potential locations.

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 100 of 188
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Over 300 quarters of data were accumulated in the three-year program. For this appendix, the annual data were fit to lognormal distributions. All three data-weighting methods produced similar results. Table B-7 summarizes the data. The GM and 95th percentile are shown for the "uniform" (unweighted least squares) lognormal fit.

Period	Locations	Geometric mean	95th percentile	Reference	Page(s)	
1996	68	0.435	7.92	Renk 1988	65-66	
1997	42	0.470	1.66	Renk 1988	38	
1998 <sup>a</sup>	54	0.524	1.61	Renk 1988	38	
1000	54	0.524	1.01	1300	00	

Table B-7. Summary of the 1996 through 1998 thoron measurement data (pCi/L).

a. Annual value was not available in reference; average for 3 quarters was calculated.

The data in Table B-7 may be compared to the thoron concentration estimate of 200 pCi/L for "1954-1989 Various Storage Sites" and "1972-2006 Final Closure Storage" (Table B-5). This comparison is plotted in Figure B-3, which shows the GMs and 95th percentiles for all three fitting methods reported by the Lognormal Fitting Utility Program (Version 2006-10-12.0; Battelle 2007). It is apparent from the comparison that the Sections B.1 to B.4 estimate of 200 pCi/L is at least a factor of 25 higher than the 95th percentile of the 1996 to 1998 measurements.

There are two individual measurements cited in the references that are worth discussing. The first is a measurement of 267 pCi/L for Building 65, which was included in the 1996 distribution above. It is a high outlier, as can be seen in Figure B-4. The measurement was for December 1995 through March 1996. There were no measurements for the other three calendar guarters in this location. The second measurement for discussion is also from Building 65. It was located in the 1995 ALARA design review for the Building 65 Thorium Overpacking Project (Allen 1995a, p. 31). This reference cites a thoron concentration of 112.75 pCi/L based on "thoron cup data" (another term for track-etch dosimeters). This was apparently a value determined with only natural ventilation, since it was used in the design review to calculate the effect of forced ventilation through HEPA filters (Allen 1995a, p. 32). At the time, 5,600 drums of thorium were stored in Building 65 (Allen 1995a, p. 10) with very limited access and with radiological controls imposed. The design parameters in various documents assumed that only 20 workers would be involved in the overpacking of Building 65 (e.g., Toler 1995, p. 187). The above-cited measurements were the only two values greater than 100 pCi/L found in the site documents. From this discussion, it is apparent that Building 65 was a separate distribution. Although the 267 pCi/L was included in the 1996 analysis, it really should not be considered as a part of a site-wide distribution of thoron results.

Additionally, the 267 pCi/L measurement from Building 65 was used, along with the inventory of stored thorium to generate a release fraction of  $3 \times 10^3$ . This appears to be an error, as the 267 pCi/L was assumed to be a progeny measurement, and used an equilibrium factor of 0.02 to revise the radon concentration to  $1.3 \times 10^4$  pCi/L. Renk (1988, p. 67) indicates that this result was a thoron track etch result, meaning it was a measurement of thoron gas, not progeny. Consequently, the release fraction should be 50 times lower than  $3 \times 10^{-3}$ , or more in the  $3 \times 10^{5}$  range.

### B.5.4 Thoron Daughter WL Measurements

It is useful to illustrate how the WL estimate in Sections B.1 to B.4 was calculated from the thoron estimate. The parameters are:

- 200 pCi/L, calculated as a conservative value in Sections B.1 to B.4 for "1954-1989 Various Storage Sites" and "1972-2006 Final Closure Storage" (Table B-5). This covers the periods/locations where measurements are available.
- 7.5 pCi/L/WL, for thoron with daughters in full equilibrium.
- 0.02 to 0.04 (2% to 4%), equilibrium factor. As Comment 3 points out, the equilibrium factor range listed in site documents is 0.02 to 0.10 (e.g., Daniels 1995, p. 13). However, later research (which included Fernald data) indicates that the indoor equilibrium factor is 0.02 and cannot exceed 0.04 (Harley and Chittaporn 2006, p. 4).

Therefore, for 200 pCi/L thoron, the WL for thoron daughters with an equilibrium factor of 0.02 is:

$$WL_{daughters} = \frac{200 \text{pCi/L}}{7.5 \frac{\text{pCi}}{\text{L}} / WL} \times 0.02 = 0.53 \text{ WL}$$
(B-1)

Or, for an equilibrium factor of 0.04:

$$WL_{daughters} = \frac{200 \text{pCi/L}}{7.5 \frac{\text{pCi}}{\text{L}} / \text{WL}} \times 0.04 = 1.07 \text{WL}$$
(B-2)

Note that a value of 0.53 WL was used in Sections B.1 to B.4 for "1954-1989 Various Storage Sites" and "1972-2006 Final Closure Storage" (Table B-5). The 1.07-WL value is also shown on the WL plot in this appendix (Figure B-5).

The WL measurements that were obtained are as limited as the thoron measurements. As noted above, some WL measurements were done by recounting air samples to determine the contribution from thoron daughters. As such, some measurements represent the WL averaged over only several minutes. When several measurements of this type were obtained from one location or facility, they are the most useful; but, unfortunately, this was not always the case.

As discussed above, thoron daughter measurements in Plant 9 in 1954 were obtained (NLO 1954). The results, which required some manipulation, are in Attachment 1 of this appendix. Briefly, the measurements were taken at six locations to determine the effect of turning off the ventilation. A series of five measurements were taken at five of the locations. The samples were counted twice. The difference in concentrations between the first and second counts was due to the <sup>212</sup>Pb, <sup>212</sup>Bi and <sup>212</sup>Po activity. This concentration can be converted to units of pCi/L and then can be divided by 7.5 pCi/L per WL to determine the number of WLs. The first 11 measurements were taken before the ventilation was disrupted. These measurements averaged 0.058 ±0.05 WL (±1 standard deviation). It is not clear whether any additional measures were taken to maximize the effect of the reduced ventilation, such as closing windows and doors that would normally be open. Given this uncertainty, the results appear to be consistent with the Sections B.1 to B.4 value of 0.034 WL. Because of their unique nature, these measurements are not included in the analyses of the 1986-and-later results below.

Due to the lack of data, both individual measurements and distributions of measurements were included in the 1986-and-later analysis. For Buildings 64 and 65, WL measurements were obtained in

1996 by using WL monitors. The detectors recorded hourly measurements for 7 to 8 days. These measurements are different from most of the discovered data in two ways. First, they more closely fit a normal rather than a lognormal distribution. Second, there was the potential for time dependence to interfere with the analysis. This was evident in the 1996 Building 65 measurements where it appeared that the WL monitor was started and the building ventilation was subsequently decreased. The first 22 measurements collected were the 22 lowest measurements, and they increased in an exponential fashion. These measurements were eliminated from the fit, which greatly improved it. The remaining points fit a normal distribution well, with an R-squared value of 0.978.

The summary of the WL measurements appears in Table B-8.

Table B-8. Summary of WL measurements.

	Number		Mean, GM, or	95th		
Time and/or	of		measurement	percentile		
location	results	Distribution	(WL)	(WL)	Reference	Page(s)
1986 Plant 8	13	Lognormal	0.0650	30.2	WMCO 1996	17, 63
silo & bins						
1986 various	22	Lognormal	0.0217	0.133	Weaver 1987	4
locations						
1987 various	55	Lognormal	0.0117	0.180	Weaver 1987	4, 5
locations						
1990-1991	85	Normal	0.108	Not	FEMP 1996–1997	68
Building 71				applicable		
(before <sup>a</sup> )						
1991 Plant 9	182	Lognormal	0.023	Not	Renk 1998	118
				applicable		
1992 Building	1	Not	0.002	Not	WMCO 1992b	3
3045		applicable		applicable		
1992 Building	1	Not	0.016	Not	FMPC 1992	3
15 C-29A Cave		applicable		applicable		
1992 Building	1	Not	0.064	Not	FMPC 1992	3
15 N-4		applicable		applicable		
1993 Plant 1,	1	Not	0.394	Not	FERMCO 1993a	3
Ore Silos BZ		applicable		applicable		
1993 Plant 1,	1	Not	0.239	Not	FERMCO 1993a	8
Ore Silos BZ		applicable		applicable		
1993 Plant 1,	1	Not	3.1	Not	FERMCO 1993a	10
Ore Silos BZ		applicable		applicable		
1995 Building	1	Not	7	Not	Toler 1996a	13
65 w/o		applicable		applicable		
ventilation						
1995 Building	1	Not	0.02	Not	Toler 1996a	13
65 w ventilation		applicable		applicable		
1996 Building	168	Normal	0.1030	0.1631	Renk 1998	22–26
64 C.P. (1-wk)						
1996 Building	168	Normal	0.0599	0.0987	Renk 1998	45–49
64 N. (1-wk)						
1996 Building	191	Normal	2.20	2.82	Renk 1998	51–56
65 (8-day)						
1996 Building	18	Normal	0.007	0.011	FEMP 1996–1997	68
71 (after*)						
2002–2003	34	Lognormal	0.0131	0.953	FEMP 1993-2004	175–240
Pilot Plant		Ŭ				

a. "Before" and "after" refer to sealing of the floor in Building 71.

The data in Table 2 were compared to both the 0.02 equilibrium calculation in Sections B.1 to B.4 and the 0.04 equilibrium calculation shown above. The results appear in Figure B-5. Most of the individual measurements, means or GMs, and 95th percentiles are less than the 0.53-WL estimate in Sections B.1 to B.4. The five exceptions are discussed individually below.

### Exception 1: 1986 Plant 8 Silo and Bins (95th percentile = 30.2 WL)

Three of 13 measurements taken before the remediation of the Plant 8 Silos and Bins (aka Project 1) were greater than 0.53 WL (two were below detection). The three points were all significantly greater

than the remaining measurements, resulting in a GSD of 41.8 and a high 95th percentile. However, all three measurements were taken in areas not normally occupied, as shown in Table B-9.

8 Silo and bins in 1986.	
Location	WL Measurement
Top of silo	17
Top of silo, upwind side	2.3
Top of silo, downwind side	71

Table B-9. Three high measurements near Plant 8 silo and bins in 1986.

The remaining measurements, excluding the ones shown as "none detected," ranged from 0.0091 to 0.44 WL, well within the 0.53 WL estimated in Sections B.1 to B.4.

**Exception 2: 1993 Plant 1, Ore Silos Breathing Zone (BZ) (Individual Measurement = 3.1 WL)** The Plant 1 Ore Silos Project involved the dismantling of 14 silos which had formerly contained cold raffinates from uranium concentrate processing (FERMCO 1995, p. 88). One of three samples from a BZ sampler recovered for the Plant 1 Ore Silos Project in 1993 was above the 0.53 WL estimate in Sections B.1 to B.4. Normally a BZ sample indicates that it was hands-on work with appropriate protections provided to the few employees involved. In this case, no monitored employee name was on the sample sheet which is unusual for a BZ sample. The location description was "Plant 1 ore Silos Project (Silo #14) 3' inside from top." It is likely that this was a BZ sampler being used as an area sampler and that this was not an occupied area.

### Exception 3: 1995 Bldg 65, Without Ventilation (Individual Measurement = 7 WL)

Three WL values were reported were for Building 65 in 1995 for three corresponding ventilation conditions. Building 65 was the facility that contained 5,600 drums of thorium materials, some of which had deteriorated. The measurement quoted above was apparently the maximum of several measurements. The Health Physics Plan for the Thorium Overpacking Project states, "Baseline WLM measurements in Building 65 indicate that airborne radon/thoron progeny may approach 7 WLs" (Toler 1996b, p. 12). With 16,000 cubic feet per minute ventilation, the value quoted was less than 0.02 WL. With three of the HEPA ventilation units running, the value guoted was 0.10 WL. The latter two values are less than the 0.53 WL used in Sections B.1 to B.4. In any case, the area was restricted and protections were in place for the relatively-small workforce that entered the facility for overpacking. Respiratory protection was required for building entry (Hazlewood 1995, p. 154). This would have been effective for the particulate daughters, although not for the thoron itself. Another 1995 reference quotes WL values that vary anywhere from 2 WLs to 10 WLs but that are normally in the 3 to 6 WLs range. The numbers cited were for radon and thoron, and the reference notes that the WL monitors normally indicate 95% to 100% thoron (Allen 1995b, p. 155). Because the range from 2 to 10 WL is consistent with the 7-WL measurement, it is assumed that these values also apply to static or no-ventilation conditions, and that the values were much lower when workers were present.

## Exception 4: 1996 Bldg 65, Eight Days of Hourly Measurements by a WL Monitor (mean = 2.2 WL)

This series of measurements is related to the 1995 Building 65 measurements cited above. As previously discussed, the measurements increased with time for the first 22 measurements (about one day). This seems to indicate some decrease in the ventilation. After equilibrium is achieved, the measurements are normally distributed about a mean of 2.2 WL. The same comments apply to these measurements as the 1995 Building 65 measurements; namely, the area was restricted, and protections were in place for the relatively-small workforce that entered the facility for overpacking. The fact that the levels increased to the range discussed in the previous paragraph probably indicates

that this measurement applies to static or no-ventilation conditions. Again, it is assumed that the values were much lower when workers were present.

### Exception 5: 2002–2003 Pilot Plant (95th Percentile = 0.95 WL)

This series of measurements consisted of both high-volume and low-volume air samples. Almost all of the samples were collected for only five minutes. The measurements are in a document that is marked as RI/FS samples in support of the Pilot Plant demolition (FEMP 1993-2004, pp. 175, 177, 179, 181, 183, 185, 187, 189, 191, 193, 195, 197, 199, 201, 203, 205, 207, 209, 211, 213, 215, 217, 219, 221, 223, 225, 227, 229, 230, 232, 234, 236, 238, and 240). However, the 2002 to 2003 radon/thoron samples are noted as being either weekly or monthly routine samples. The data did not fit either a normal or a lognormal distribution well. The high- and low-volume samples were also plotted and fitted separately. The low-volume samples provided a better lognormal fit, but the highvolume samples did not. The main issue with the high-volume samples was a set of three low outliers in the data, with a magnitude on the order of  $1 \times 10^5$  WL. These three points were an order of magnitude lower than any of the other WL results reported. The best visual fit to a lognormal was obtained by eliminating these data, as well as two negative results and one illegible result. Instead, the somewhat arbitrary decision was made to treat these data as less than detection and include them in the fit. The resulting estimate of the 95th percentile exceeded the Sections B.1 to B.4 value of 0.53 WL, but not the 1.07 WL value for a 0.04% equilibrium factor. None of the underlying data exceeded the Sections B.1 to B.4 value.

### B.5.5 <u>Conclusion</u>

With only one exception, the thoron measurements in 1996 through 1998 were far below the 200 pCi/L estimated in Sections B.1 to B.4. The lone exception was for Building 65 for December 1995 through March 1996. No additional details about this measurement were discovered. However, during the ALARA Design Review, a measurement of half that magnitude was considered to be typical for Building 65 under natural ventilation conditions (Allen 1995a, p. 31). The value used in the ALARA Design Review did not exceed 200 pCi/L.

In general, the thoron daughter measurements were below 0.53 WL. The exceptions were discussed in detail above and can be summarized as: being in Building 65, in areas not normally occupied (top of Plant 8 silos), or one of three BZ samples from the Plant 1 ore silos project which may have been an area sample rather than a BZ sample of an individual.

From the above, one can conclude that, compared to the obtained measurements, the thoron and thoron daughter concentration estimates in Sections B.1 to B.4 used to generate Tables B-5 and B-6 are reasonably conservative.

ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)



Figure B-3. Thoron comparison, 1996 to 1998.

ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)



Figure B-4. Measurements and lognormal fits, 1996.

ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)



Figure B-5. WL comparisons, 1986 to 2003. 1990 to 1991 measurements were made before sealing of the floor in Building 71; 1996 measurements were made after sealing.
#### ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

#### B.5.6 <u>1954 Thoron/Thoron Daughter Results</u>

Twenty-six (26) Plant 9 thoron/thoron daughter results for 1954 were located in NLO (1954). Samples were collected at six locations, as shown in Table B-10.

Location	Number of samples
GA wet area-main floor	5
GA wet area-top deck	5
GA TNT & sump cake storage area	5
GA north end of rod storage area	5
GA center of rod storage area	5
GA south end of rod storage area	1

	Table B-10.	Locations	and numbers	of	samples.
--	-------------	-----------	-------------	----	----------

The samples were taken with the ventilation turned off to determine the thoron build-up. The time that the ventilation was turned off was noted as March 8, 1954 at 10:37 a.m. (NLO 1954, p. 13). The heating unit apparently remained on, recirculating room air. The samples were collected on March 8, 1954 between 9:25 a.m. and 13:25 a.m.

The samples were counted either on March 8 or March 9 ("first count"). They were recounted ("second count") on March 15. Since the first counts were taken between 3.75 and 19.58 hours after the sample was collected, the <sup>220</sup>Rn and <sup>216</sup>Po would have decayed, leaving only <sup>212</sup>Pb and its daughters, including the alpha-emitters <sup>212</sup>Bi and <sup>212</sup>Po. Considering the branching ratios, the average alpha energy per emission is 7.8 MeV. This leads to the definition of a thoron WL as 7.5 pCi/L of thoron-daughter alpha activity. See Table B-3.

The calculations for reproducing the net counts per minute shown on the sample sheets are straightforward. This quantity has to be divided by the counting efficiency and sample volume, which are shown on the sample sheet, as well as a factor of 0.70, to produce the concentration at the time of counting. The factor of 0.70 is not shown on the sample sheet. This factor was likely a correction for sample self-absorption of the alpha particles due to dust loading of the filter [see Boback (undated, pp. 39-40), although an absorption factor of 0.5 was used in that example]. It is clear that this was not a correction for decay from sample collection to counting since it was applied to both the first and second counts as well as other counts in NLO (1954) for long-lived activity. Most, but not all, of the samples had an additional correction applied to the first count which varied from 0.60 to 0.79. This correction was assumed to be a decay correction. Since the correction could not be reproduced and was not consistently applied, this analysis uses a recalculated decay correction based on the elapsed time between the sample collection and the count times and the 11-hour half-life of <sup>212</sup>Pb. The second counts occurred between 167 and 169 hours after sample collection, or at least 15 <sup>212</sup>Pb half-lives after sample collection. Therefore, all of the thoron daughter activity had decayed, leaving only longlived isotopes, such as uranium and thorium. The difference between the first and second counts is due to the activity of <sup>212</sup>Pb and its daughters. The decay correction was applied to the difference between the activity in the first count and the long-lived activity in the second count. This subtraction did not make a significant difference, which is probably why it was ignored in the site's analysis. The concentrations determined by the above method were converted to pCi/L and then divided by 7.5 pCi/L to determine the daughter concentration in WLs.

The results and their references are shown in Table B-11.

#### ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

	Date and time		Recalculated		Reported	Calculated	Page(s)
Sample	of sample	Location	concentration (dpm/m <sup>3</sup> )	WL	thoron	thoron	(NLO 1954)
002	3/8/1954 9:25	GA wet area-main floor	2,399	0.144	3.69	54.0	14, 69
003	3/8/1954 9:25	GA wet area-top deck	1,988	0.119	3.06	44.8	14, 69
004	3/8/1954 9:26	GA TNT & sump cake storage area	256	0.015	0.4	5.8	14, 69
005	3/8/1954 9:26	GA north end of rod storage area	647	0.039	1.05	14.6	14, 69
006	3/8/1954 9:26	GA center of rod storage area	386	0.023	0.63	8.7	14, 69
007	3/8/1954 9:27	GA south end of rod storage area	683	0.041	1.11	15.4	14, 69
008	3/8/1954 10:02	GA wet area-main floor	1,902	0.114	3.2	42.8	14, 69
009	3/8/1954 10:02	GA wet area-top deck	1,027	0.062	1.73	23.1	14, 69
010	3/8/1954 10:02	GA TNT & sump cake storage area	300	0.018	0.5	6.7	14, 69
011	3/8/1954 10:03	GA north end of rod storage area	499	0.030	0.79	11.2	14, 69
012	3/8/1954 10:03	GA center of rod storage area	557	0.033	0.94	12.6	14, 69
013	3/8/1954 10:40	GA wet area-main floor	1,930	0.116	4.32	43.5	12, 13
014	3/8/1954 10:40	GA wet area	1,153	0.069	2.53	26.0	12, 13
015	3/8/1954 10:40	GA TNT & sump cake storage area	827	0.050	1.81	18.6	12, 13
016	3/8/1954 10:39	GA north end of rod storage area	1,931	0.116	4.23	43.5	12, 13
017	3/8/1954 10:39	GA center of rod storage area	1,781	0.107	3.91	40.1	12, 13
018	3/8/1954 11:15	GA wet area-main floor	2,798	0.168	6.36	63.0	12, 13
019	3/8/1954 11:15	GA wet area	2,239	0.134	5.08	50.4	12, 13
020	3/8/1954 11:15	GA TNT & sump cake storage area	2,239	0.134	5.08	50.4	12, 13
021	3/8/1954 11:15	GA north end of rod storage area	2,034	0.122	4.62	45.8	12, 13
022	3/8/1954 11:15	GA center of rod storage area	2,800	0.168	6.36	63.1	12, 13
023	3/8/1954 13:25	GA wet area-main floor	10,123	0.608	20.78	228.0	16
024	3/8/1954 13:25	GA wet area–2nd deck	6,060	0.364	12.43	136.5	16
025	3/8/1954 13:25	GA TNT & sump cake storage area	1,617	0.097	3.44	36.4	16
026	3/8/1954 13:25	GA north end of rod storage area	2,258	0.136	4.74	50.9	16
027	3/8/1954 13:25	GA center of rod storage area	3,948	0.237	8.92	88.9	16

Table B-11. Thoron and thoron daughter results (pCi/L).

#### ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

The results were plotted by location for the five locations with multiple samples. The plots are in Figures B-3 through B-10. The concentrations (in terms of WL) generally increased, as expected. The samples were collected from 9:30 a.m. to about 1:30 p.m. In one location (GA TNT and sump cake storage area), the concentration decreased between 11:30 a.m. and 1:30 p.m. Also shown on the plots is the 0.034 WL estimate from Sections B.1 to B.4. The first 11 sample times were before the ventilation was disrupted. These measurements averaged 0.058  $\pm$ 0.05 WL ( $\pm$ 1 standard deviation). If only the first measurement in each location is considered, the measurements averaged 0.064  $\pm$ 0.05 WL. It is not clear whether any additional measures were taken to maximize the effect of the reduced ventilation, such as closing windows and doors that would normally be open. It is also not clear whether the sample times were the beginning, middle, or end of the sample collection. For the recalculations, it was assumed that they were the end of the sample collection. Given this uncertainty, the results appear to be consistent with the Sections B.1 to B.4 value of 0.034 WL.

The sample sheets also reported concentrations of thoron gas in Ci/L. Because there were issues with the calculation of the concentrations of the daughters, the thoron concentrations based on them also had issues. Although the site's calculated thoron concentrations were consistent with Sections B.1 to B.4, the equilibrium factors being assumed to determine the thoron concentrations were much higher than are realistic based on later research (see the response to Comment 3 (Equilibrium Factor) in this appendix). The thoron concentrations were recalculated from the recalculated WLs and the 0.02 equilibrium factor used in Sections B.1 to B.4. The thoron concentrations were 21.8  $\pm$ 17.2 pCi/L before the ventilation was turned off and 23.9  $\pm$ 20.3 pCi/L for just the first six measurements. The 12.8 pCi/L estimate in Sections B.1 to B.4 lies within one standard deviation of these results.



Figure B-6. GA center of rod storage area.





Figure B-7. GA north end of rod storage area.



Figure B-8. GA TNT and sump cake storage area.



ATTACHMENT B ANALYSIS OF POTENTIAL THORON EXPOSURES (continued)

Figure B-9. GA wet area, main floor.



Figure B-10. GA wet area, top deck. Assumed to be the same location as "GA wet area" and "GA wet area – 2nd deck".

#### ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA

#### **TABLE OF CONTENTS**

#### SECTION

#### TITLE

PAGE

C.1	Purpose	
C.2	Data and Method Overview C.2.1 Bioassay Data Selection C.2.2 Method Description	
C.3	Analysis	119
C.4	Intake Modeling C.4.1 Assumptions C.4.2 Bioassay Fitting	
C.5	Assigning Intakes and Doses C.5.1 Intake Rate Summary C.5.2 Dose Assignment	
C.6	Coworker Data Figures	

#### LIST OF TABLES

TITLE

#### C-1 C-2 Summary of uranium urinary excretion rate analyses, 1952 to 2006 ...... 118 C-3 C-4 C-5 Derived uranium intake rates for type M materials, 1952 to 2006...... 121 C-6

# LIST OF FIGURES

#### C-1 Predicted values versus measured results, 01/01/1952 to 12/31/1993, 50th percentile, C-2 Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile, C-3 Predicted values versus measured results, 01/01/1952 to 12/31/1993, 84th percentile, C-4 C-5 Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile, C-6 Predicted values versus measured results, 01/01/1952 to 12/31/1993, 50th percentile,

TABLE

# FIGURES

#### TITLE

#### PAGE

PAGE

C-7	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile, type M	124
C-8	Predicted values versus measured results, 01/01/1952 to 12/31/1993, 84th percentile, type M	125
C-9	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile, type M	125
C-10	Predicted values versus measured results, 01/01/1952 to 12/31/1954, 50th percentile, type S	125
C-11	Predicted values versus measured results, 01/01/1955 to 12/31/1955, 50th percentile, type S	126
C-12	Predicted values versus measured results, 01/01/1956 to 12/31/1956, 50th percentile, type S	126
C-13	Predicted values versus measured results, 01/01/1957 to 12/31/1961, 50th percentile, type S	126
C-14	Predicted values versus measured results, 01/01/1962 to 12/31/1966, 50th percentile, type S	127
C-15	Predicted values versus measured results, 01/01/1967 to 12/31/1993, 50th percentile, type S	127
C-16	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 50th percentile, type S	127
C-17	Summarized results of the predicted values versus the excluded results, 01/01/1952 to 12/31/2006, 50th percentile, type S	128
C-18	Predicted values versus measured results, 01/01/1952 to 12/31/1956, 84th percentile, type S.	128
C-19	Predicted values versus measured results, 01/01/1957 to 12/31/1961, 84th percentile, type S.	
C-20	Predicted values versus measured results, 01/01/1962 to 12/31/1966, 84th percentile, type S	129
C-21	Predicted values versus measured results, 01/01/1967 to 12/31/1980, 84th percentile, type S	129
C-22	Predicted values versus measured results, 01/01/1981 to 12/31/1993, 84th percentile, type S	120
C-23	Predicted values versus measured results, 01/01/1994 to 12/31/2006, 84th percentile, type S	130
C-24	Summarized results of the predicted values versus the excluded results, 01/01/1952 to 12/31/2006, 84th percentile, type S	130

#### C.1 Purpose

Some employees at DOE sites were not monitored for internal ionizing radiation exposure, or the monitoring records are incomplete or unavailable. In such cases, data from monitored coworkers can be used to estimate a worker's possible exposure. The purpose of this attachment is to provide monitored coworker information for calculating and assigning occupational internal doses to employees at the Fernald site for whom there are no or insufficient bioassay monitoring records.

#### C.2 Data and Method Overview

ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005b), describes the general process that is used to analyze bioassay data for assigning doses to individuals based on coworker results. ORAUT-PLAN-0014, *Coworker Data Exposure Profile Development* (ORAUT 2004), describes the approach and processes to be used to develop reasonable exposure profiles based on available dosimetric information for workers at DOE sites.

#### C.2.1 Bioassay Data Selection

The bioassay results for this analysis were obtained by extracting data from HIS-20\_ORAU, a database of Fernald bioassay results (ORAUT 2007). One of the database tables, HIS20\_V\_BIOASSAY, contains results for urinalyses, fecal analyses, and in vivo analyses starting in 1952. The radionuclides in the table include <sup>228</sup>Ac, <sup>36</sup>Cl, <sup>137</sup>Cs, <sup>3</sup>H, <sup>210/211</sup>Pb, <sup>210</sup>Po, <sup>239</sup>Pu, <sup>226/228</sup>Ra, <sup>90</sup>Sr, <sup>228/230/232</sup>Th, <sup>234/235/238</sup>U, and total uranium. The vast majority of data in the table concerns the uranium radionuclides. Data were extracted from the HIS20\_V\_BIOASSAY table using the field criterion in Table C-1.

Field name	Field value			
TYPE_BIOASSAY	URINALYSIS			
NUC_NAME	U-TOTAL or 238U			
SAMPLE_TYPE	Not equal to 10, 5C, 70, VF, VR, VE			
ACT_UNITS_SU	μg/L			

Table C-1. Criteria applied to extract for uranium bioassay data.

Samples with code 10 and 70 were collected before employment or reemployment to establish the background for the individual. As such, these samples are not appropriate for use in a study to establish intake potential. Samples with code 5C were collected to test for possible correlation with abnormal clinical laboratory findings. VF, VR, and VE are visitor samples and likely not representative of the typical worker (ORAUT 2004). All results in this series were less than the MDA. Of the 403,159 uranium urinalysis results not excluded as discussed above, 144 had results of "N/A"; these were discarded. The remaining 403,015 sample results were evaluated using the time-weighted one person–one sample (TWOPOS) methodology (ORAUT 2014c), which yielded 86,964 OPOS results.

#### C.2.2 <u>Method Description</u>

The reporting methods for bioassay results changed as a function of time. Before 1986 and after 1997, the data appear to be uncensored. That is, the analytical result is reported regardless of magnitude. From 1986 through 1993, the minimum reported value incrementally changed from 3  $\mu$ g/L to a maximum of 14  $\mu$ g/L. Table C-2 lists the reporting levels as a function of time. Part of this change, and especially for 1991 through 1993, appears to be a refinement of the quantification of the detection capabilities of the methods and a change from reporting all results to results at or above the

Table C-2. Reporting level.					
From	То	CL (µg/L)			
Earliest	01/28/86	Uncensored			
01/29/86	12/30/88	3			
01/01/89	09/17/90	5			
09/18/90	10/31/90	8			
11/01/90	02/08/91	9			
02/09/91	03/04/91	8			
03/05/91	04/15/91	7			
04/16/91	06/26/91	8			
06/27/91	08/03/91	9			
08/04/91	12/11/91	10			
12/12/91	01/08/92	9			
01/09/92	02/12/92	10			
02/13/92	04/22/92	11			
04/23/92	08/27/92	12			
08/28/92	11/30/93	14			
12/01/93	06/02/97	0.8			
06/03/97	End	Uncensored			

DL and then to reporting only results above the MDA. This censoring of results below a given level skews the results of the calculated fits, which results in values that are biased high. Figure C-1 depicts how recalculating the results for 1986 with varying censoring levels (CLs) alters the regression on order statistics (ROS) fits.<sup>3</sup> The original censoring level (lowest line) was 3  $\mu$ g/L.

The higher results in 1991 through 1993 are believed to be a result of the higher CLs in those years. In 1992 and 1993, over 99% of the results are censored. In 1991, there appear to have been at least seven censoring levels that varied with time, which generates uncertainty about what results are censored. The results for all three of these years are inconsistent with the results for years before or after this period. During this period, Fernald ceased uranium production and began the process of site closure. Therefore, the potential for exposure was lower due to the lower production levels and the fact that site closure had not begun. Therefore, the results for 1991 through 1993 have been excluded from the intake modeling.

The excretion rates for each sample were normalized to represent 24-hour samples assuming 1,400 mL/d urine excretion, which is the daily volume that is excreted by Reference Man in International Commission on Radiological Protection (ICRP) Publication 23 (ICRP 1975). Then the intakes that would result in the observed excretion rates were inferred using IMBA as described in Section C.4.

The uranium in urine bioassay data from the HIS-20\_ORAU database were chronologically grouped into annual intervals by calendar year. The data in each year were fit to a lognormal distribution as described in Section 3.0. The analysis of the fits resulted in calculated excretion rates at the 50th- and 84th-percentile values at the midpoint date of the analysis interval as shown in Table C-3.

<sup>&</sup>lt;sup>3</sup> Note that this figure was produced using the OPOS method from Revision 01 of ORAUT-RPRT-0053, *Analysis of Stratified Coworker Datasets*, analyzed in Revision 02 of this document, whereas the current revision of this document used Revision 02 of ORAUT-RPRT-0053 (ORAUT 2014c) to calculate the actual intakes. However, the concept being depicted is the same regardless.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-1. Change in lognormal fit for various CLs.

Table C-3. Summary of uranium urinary excretion rate analyses, 1952 to 2006.

		Number of	50th	84th
	Effective	TWOPOS	percentile	percentile
Year	bioassay date	results	(µg/d)	(µg/d)
1952	07/01/1952	71	28.81	107.07
1953	07/01/1953	701	19.60	57.07
1954	07/01/1954	1,376	26.11	71.47
1955	07/01/1955	1,973	45.78	117.20
1956	07/01/1956	2,497	26.11	66.50
1957	07/01/1957	2,937	17.88	46.03
1958	07/01/1958	2,485	12.86	29.79
1959	07/01/1959	2,540	13.97	28.64
1960	07/01/1960	2,630	18.89	33.88
1961	07/01/1961	2,395	18.18	32.30
1962	07/01/1962	2,131	12.54	23.93
1963	07/01/1963	1,983	13.27	26.42
1964	07/01/1964	1,900	10.92	24.05
1965	07/01/1965	1,663	9.01	20.22
1966	07/01/1966	1,484	8.24	18.46
1967	07/01/1967	1,602	6.96	14.94
1968	07/01/1968	1,398	6.14	13.35
1969	07/01/1969	1,281	5.48	12.30

			50(1	0.44
		Number of	50th	84th
Veer	Effective	TWOP05	percentile	percentile
1070	Dioassay date	results	(µg/a)	(µg/a)
1970	07/01/1970	1,119	4.86	10.15
1971	07/01/1971	881	5.27	11.67
1972	07/01/1972	634	5.01	10.41
1973	07/01/1973	735	5.23	11.07
1974	07/01/1974	678	5.36	11.07
1975	07/01/1975	697	5.23	10.37
1976	07/01/1976	697	5.22	10.24
1977	07/01/1977	664	5.15	9.62
1978	07/01/1978	644	5.55	10.03
1979	07/01/1979	599	5.13	9.75
1980	07/01/1980	893	5.04	11.37
1981	07/01/1981	623	2.93	7.26
1982	07/01/1982	262	5.70	10.44
1983	07/01/1983	785	3.86	8.71
1984	07/01/1984	696	5.01	11.00
1985	07/01/1985	858	4.70	8.50
1986	07/01/1986	1,565	4.01	6.16
1987	07/01/1987	1,611	2.91	5.50
1988	07/01/1988	1,609	3.53	5.24
1989	07/01/1989	2,029	1.79	3.64
1990	07/01/1990	2,044	5.12	7.36
1991	07/01/1991	2,163	12.39	12.95
1992	07/01/1992	2,803	11.79	13.64
1993	07/01/1993	3,166	10.81	12.79
1994	07/01/1994	2,817	0.097	0.293
1995	07/01/1995	2,901	0.105	0.317
1996	07/01/1996	2,298	0.582	0.951
1997	07/01/1997	2,159	0.187	0.570
1998	07/01/1998	2,382	0.025	0.070
1999	07/01/1999	2,351	0.029	0.071
2000	07/01/2000	2,076	0.039	0.100
2001	07/01/2001	1,809	0.070	0.147
2002	07/01/2002	1,835	0.063	0.128
2003	07/01/2003	1,787	0.063	0.135
2004	07/01/2004	1,217	0.065	0.123
2005	07/01/2005	1.040	0.089	0.170
2006	07/01/2006	790	0.082	0.162

# C.3 Analysis

Bioassay data statistics were generated for each analysis interval using the methods in ORAUT (2014c). A lognormal distribution was assumed [9]. After log-transforming the data, the 50th- and 84th-percentile values were determined for each period using the methods in ORAUT (2014c). For years with uncensored data, values less than or equal to zero were treated as being censored at the lowest positive value in that year for TWOPOS implementation. However, values equal to the lowest positive value are still considered to be uncensored. Table C-3 shows the statistical analysis results for uranium urinary excretion parameters.

#### C.4 Intake Modeling

This section discusses intake modeling assumptions and intake fitting for three different material types of uranium compounds.

#### C.4.1 <u>Assumptions</u>

Each result in the intake calculations was assumed to have normal distribution [10]. A uniform absolute error of 1 was applied to all results to assign the same weight to each result. Because of the nature of work at Fernald, it is possible that intakes could have been either chronic or acute. However, a series of acute intakes can be approximated as a chronic intake. Therefore, intakes were assumed to be chronic and to occur through inhalation using a default breathing rate of 1.2 m<sup>3</sup>/hr and a particle size distribution of 5-µm activity median aerodynamic diameter (ICRP 1995).

#### C.4.2 Bioassay Fitting

IMBA was used to fit the bioassay results to a series of inhalation intakes. Data from 1952 through 2006 were fit as a series of chronic intakes. The intake assumptions were based on observed patterns in the bioassay data. Periods with constant chronic intake rates were chosen by selecting periods during which the bioassay results were similar. A new chronic intake period was started if the data indicated a significant and sustained change in the bioassay results. By this method, the period from 1952 through 2006 was divided into multiple chronic intake periods.

Because the uranium isotopes at Fernald have long radiological half-lives and the material is retained in the body for long periods, excretion results are not independent. For example, an intake in the 1950s could contribute to urinary excretion in the 1980s and later. To avoid potential underestimation of intakes for people who worked at Fernald for relatively short periods, each chronic intake was fit independently using only the bioassay results from the single intake period for type S solubility. For solubility types M and F, this approach was used where it was determined that earlier intake rates significantly biased later intake rates (i.e., the period from 1994 through 2006 was evaluated separately from earlier periods). This method results in a potential overestimate of intakes for exposures that extend through multiple assumed intake periods. Uranium urinalysis results were analyzed with IMBA to derive intake rates for 1952 to 2006. Section C.6 contains the plots that compare predicted uranium bioassay results based on IMBA-derived uranium intake rates with the measured urine results.

# C.5 Assigning Intakes and Doses

This section describes the derived intake rates and provides guidance for assigning doses. For each intake period below, the geometric standard deviations (GSDs) were determined by dividing the 84th-percentile intake rates by the 50th-percentile rates. For the calculation of doses to individuals from bioassay data, a GSD of 3 is used to account for biological variation and uncertainty in the models (ORAUT 2014a). The same models are used for fitting the coworker data, so the same uncertainty applies. Therefore, a minimum GSD of 3 was assigned for each of the intake periods.

#### C.5.1 Intake Rate Summary

Multiple intake periods were fit to the derived 50th- and 84th-percentile uranium excretion data. Table C-4 summarizes the 50th- and 95th-percentile uranium intake rates that correspond to an intake

_	50th percentile	50th percentile		95th percentile	
Dates	(µg/d)	(pCi/d) <sup>a</sup>	GSD	(µg/d)	(pCi/d) <sup>a</sup>
01/01/52–12/31/54	91.5	89.3	3.36	673	656
01/01/55–12/31/55	169	164.5	3.00	1,027	1,002
01/01/56–12/31/56	94.3	92.0	3.26	659	644
01/01/57–12/31/61	58.6	57.2	3.00	357	349
01/01/62–12/31/64	38.2	37.3	3.00	233	227
01/01/65–12/31/66	38.2	61.7	3.00	233	376
01/01/67–12/31/93	16.7	27.0	3.00	102	164
01/01/94–12/31/06	0.418	0.675	3.00	2.55	4.12

Table C. 4. Derived uranium intelse rates for type E materials 1052 to 2006

a. The specific activities used to compute this column were 0.976 pCi/µg (1% EU) for 1952 through 1964, and 1.616 pCi/µg (2% EU) for 1965 to the present.

of type F materials that was inferred from the excretion rates. The 95th-percentile intake rates were calculated using the following equation:

95th-percentile intake = 50th-percentile intake 
$$\times$$
 GSD<sup>1.645</sup> (C-1)

Table C-5 lists the same information for type M materials, and Table C-6 lists the information for type S materials. Periods with the same intake rate and GSD were combined for clarity. For periods after 2006 during which intakes were feasible, dose reconstructors should assume the 2006 intake rates.

	50th	50th	1	95th	95th
Dates	(µg/d)	(pCi/d) <sup>a</sup>	GSD	(µg/d)	(pCi/d) <sup>a</sup>
01/01/52–12/31/54	392	382	3.29	2,779	2,712
01/01/55–12/31/55	744	726	3.00	4,531	4,422
01/01/56–12/31/56	328	320	3.93	3,117	3,042
01/01/57–12/31/61	236	230	3.00	1,435	1,401
01/01/62-12/31/64	153	149	3.00	933	911
01/01/65–12/31/66	153	247	3.00	933	1,508
01/01/67–12/31/93	67.5	109	3.00	411	665
01/01/94–12/31/06	1.73	2.79	3.00	10.5	17.0

Table C-5 Derived uranium intake rates for type M materials, 1952 to 2006

The specific activities used to compute this column were 0.976 pCi/µg (1% EU) for 1952 through a. 1964, and 1.616 pCi/µg (2% EU) for 1965 to the present.

#### C.5.2 **Dose Assignment**

For most cases, individual doses should be calculated from the 50th-percentile intake rates. For cases where there is justification that the individual might have had larger intakes than the 50th-percentile intake rates, dose reconstructors should use the 95th-percentile intake rates applicable to the solubility class of the material from Table C-4, C-5, or C-6 as appropriate. Dose reconstructors should select the material type that is the most favorable to claimants and apply other RU components based on information in Section 5.3.2.5.

Select the lognormal distribution in IREP with the calculated dose entered as Parameter 1 and the associated GSD as Parameter 2. The GSD relates to the intake, so apply it to all annual doses that

Dates	50th percentile (µq/d)	50th percentile (pCi/d) <sup>a</sup>	GSD	95th percentile (µq/d)	95th percentile (pCi/d)ª
01/01/52-12/31/54	8,611	8,404	3.00	52,471	51,212
01/01/55–12/31/55	25,755	25,137	3.00	156,937	153,171
01/01/56–12/31/56	14,660	14,308	3.00	89,330	87,186
01/01/57–12/31/61	4,691	4,578	3.00	28,584	27,898
01/01/62–12/31/64	2,925	2,855	3.00	17,823	17,396
01/01/65–12/31/66	2,925	4,727	3.00	17,823	28,803
01/01/67–12/31/93	827.1	1,337	3.00	5,040	8,144
01/01/94–12/31/06	21.2	34.3	3.00	129	209

Table C-6. Derived uranium intake rates for type S materials, 1952 to 2006.

a. The specific activities used to compute this column were 0.976 pCi/µg (1% EU) for 1952 through 1964, and 1.616 pCi/µg (2% EU) for 1965 to the present.

are determined from the intake period. If used, assign the 95th-percentile intakes as a constant distribution.

#### C.6 Coworker Data Figures

This section shows comparisons of measured uranium urine bioassay results with predicted results that were calculated using IMBA-derived uranium intake rates. Blue dots represent the measured values that were retained for the fit. Red dots represent results that were excluded because they were outside the intake period being fit. The green lines represent the predicted values. The figures provide the fits as follows:

- Figures C-2 and C-3 show the individual fits to the 50th-percentile excretion rates for type F material.
- Figures C-4 and C-5 show the individual fits to the 84th-percentile excretion rates for type F material.
- Figures C-6 and C-7 show the individual fits to the 50th-percentile excretion rates for type M material.
- Figures C-8 and C-9 show the individual fits to the 84th-percentile excretion rates for type M material.
- Figures C-10 through C-16 show the individual fits to the 50th-percentile excretion rates for type S material. Figure C-17 summarizes the results for the period from 1952 through 2006.
- Figures C-18 through C-23 show the individual fits to the 84th-percentile excretion rates for type S material. Figure C-24 summarizes the type S results for the period from 1952 through 2006.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-2. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1993, 50th percentile, type F.



Figure C-3. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 50th percentile, type F.



Figure C-4. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1993, 84th percentile, type F.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-5. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 84th percentile, type F.



Figure C-6. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1993, 50th percentile, type M.



Figure C-7. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 50th percentile, type M.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-8. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1993, 84th percentile, type M.



Figure C-9. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 84th percentile, type M.



Figure C-10. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1954, 50th percentile, type S.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-11. Predicted values (line) versus measured results (blue dots), 01/01/1955 to 12/31/1955, 50th percentile, type S.



Figure C-12. Predicted values (line) versus measured results (blue dots), 01/01/1956 to 12/31/1956, 50th percentile, type S.



Figure C-13. Predicted values (line) versus measured results (blue dots), 01/01/1957 to 12/31/1961, 50th percentile, type S.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-14. Predicted values (line) versus measured results (blue dots), 01/01/1962 to 12/31/1966, 50th percentile, type S.



Figure C-15. Predicted values (line) versus measured results (blue dots), 01/01/1967 to 12/31/1993, 50th percentile, type S.



Figure C-16. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 50th percentile, type S.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-17. Summarized results of the predicted values (line) versus the excluded results (red dots), 01/01/1952 to 12/31/2006, 50th percentile, type S.



Figure C-18. Predicted values (line) versus measured results (blue dots), 01/01/1952 to 12/31/1956, 84th percentile, type S.



Figure C-19. Predicted values (line) versus measured results (blue dots), 01/01/1957 to 12/31/1961, 84th percentile, type S.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-20. Predicted values (line) versus measured results (blue dots), 01/01/1962 to 12/31/1966, 84th percentile, type S.



Figure C-21. Predicted values (line) versus measured results (blue dots), 01/01/1967 to 12/31/1980, 84th percentile, type S.



Figure C-22. Predicted values (line) versus measured results (blue dots), 01/01/1981 to 12/31/1993, 84th percentile, type S.

ATTACHMENT C INTERNAL DOSIMETRY URANIUM COWORKER DATA (continued)



Figure C-23. Predicted values (line) versus measured results (blue dots), 01/01/1994 to 12/31/2006, 84th percentile, type S.



Figure C-24. Summarized results of the predicted values (line) versus the excluded results (red dots), 01/01/1952 to 12/31/2006, 84th percentile, type S.

#### ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA

#### TABLE OF CONTENTS

<u>SECT</u>	TION <u>TITLE</u>	PAGE	
D.1	Manipulation of Fernald Chest Count Data	133	
D.2	Discussion of Actinium-228 and Lead-212 in Chest Counts	134	
D.3	Bias and Decision Level Plots	135	
D.4	Chest Burden Plots	135	

#### LIST OF TABLES

#### <u>TABLE</u>

<u>TITLE</u>

D-1	Summary of bias and DL for <sup>228</sup> Ac an	d <sup>212</sup> Pb for each year134
-----	---	--------------------------------------

# LIST OF FIGURES

#### **FIGURES**

#### <u>TITLE</u>

#### PAGE

<u>PAGE</u>

D-1	Normal probability plot of <sup>212</sup> Pb chest count results for 1978	133
D-2	Normal probability plot of <sup>212</sup> Pb chest count results for 1978 after adjusting for bias	133
D-3	Normal probability plot of 212Pb chest count results for 1978 after adjusting for bias	134
D-4	Plot of 228Ac versus 212Pb for 1978 where the levels of both nuclides are above the DL	135
D-5	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1978	136
D-6	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1979	137
D-7	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1980	138
D-8	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1981	139
D-9	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1982	140
D-10	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1983	141
D-11	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1984	142
D-12	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1985	143
D-13	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1986	144
D-14	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1987	145
D-15	Normal probability plots of <sup>212</sup> Pb and <sup>228</sup> Ac, 1988	146
D-16	Both radionuclides above the DL, 1978	147
D-17	Either radionuclides above the DL, 1978	148
D-18	Both radionuclides above the DL, 1979	149
D-19	Either radionuclides above the DL, 1979	150
D-20	Both radionuclides above the DL, 1980	151
D-21	Either radionuclides above the DL, 1980	152
D-22	Both radionuclides above the DL, 1981	153
D-23	Either radionuclides above the DL, 1981	154
D-24	Both radionuclides above the DL, 1982	155
D-25	Either radionuclides above the DL, 1982	156
D-26	Both radionuclides above the DL, 1983	157

# ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)

D-27	Either radionuclides above the DL, 1983	
D-28	Both radionuclides above the DL, 1984	
D-29	Either radionuclides above the DL, 1984	
D-30	Both radionuclides above the DL, 1985	
D-31	Either radionuclides above the DL, 1985	
D-32	Both radionuclides above the DL, 1986	
D-33	Either radionuclides above the DL, 1986	
D-34	Both radionuclides above the DL, 1987	
D-35	Either radionuclides above the DL, 1987	
D-36	Both radionuclides above the DL, 1988	
D-37	Either radionuclides above the DL, 1988	

#### ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)

#### D.1 Manipulation of Fernald Chest Count Data

The <sup>228</sup>Ac and <sup>212</sup>Pb results from chest counts at Fernald from 1978 to 1988 were analyzed separately by year. Based on previous experience, the distributional model for these annual data is that they are a combination of a normal analytical background distribution with a mean of zero and a lognormal exposure distribution (ORAUT 2009). The normal component of the data is estimated by fitting a line through the negative data on a normal probability plot. The intercept of the line is the estimate of the mean or median of the normal distribution, and the slope is the estimate of the standard deviation. If the median of the data is –0.019 nCi, which means that the data are assumed to be biased. For example, the median of the data is –0.019 nCi, which means that the data are assumed to be biased by that much. The data are adjusted for any bias and then refit to force the line through zero (i.e., make the median equal to zero). For example, the adjusted 1978 <sup>212</sup>Pb data are shown below in Figure D-2.



Figure D-1. Normal probability plot of <sup>212</sup>Pb chest count results for 1978. The line is a robust regression of the data that are less than zero nCi.



Figure D-2. Normal probability plot of <sup>212</sup>Pb chest count results for 1978 after adjusting for bias. The line is a robust regression of the data that are less than zero nCi.

The process decision level (DL) above which a measured chest burden is deemed to be "positive" is taken to be the 95th percentile of the (unbiased) normally distributed analytical background. For the 1978 <sup>212</sup>Pb data, the DL is 0.12 nCi as shown in Figure D-3. Measured <sup>212</sup>Pb chest burdens below this DL are deemed to not indicate the presence of <sup>212</sup>Pb.

ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-3. Normal probability plot of <sup>212</sup>Pb chest count results for 1978 after adjusting for bias. The line is a robust regression of the data that are less than 0 nCi, and the red point is the 95th percentile of the background normal distribution (i.e., the DL).

A summary of the bias and DL for <sup>228</sup>Ac and <sup>212</sup>Pb for 1978 through 1988 are given in Table D-1, and the corresponding plots are given in Section D.3. The DLs for <sup>212</sup>Pb ranged from 0.11 to 0.18 nCi with a mean of 0.13 nCi, whereas the DL for <sup>228</sup>Ac ranged from 0.08 to 0.17 nCi with a mean of 0.11 nCi. For simplicity, a single DL of 0.12 nCi has been adopted for both nuclides for all years. If a minimum detectable amount is needed for dose reconstruction purposes, a value of 2 times the DL, 0.24 nCi, should be used for both nuclides.

Veer	Pb-212 bias	Pb-212	Ac-228 bias	Ac-228	
rear	(nCl)		(nCI)		
1978	-0.019	0.120	0.008	0.153	
1979	-0.014	0.156	0.031	0.169	
1980	-0.045	0.123	0.025	0.091	
1981	-0.056	0.108	0.022	0.127	
1982	-0.058	0.121	0.010	0.117	
1983	-0.101	0.122	-0.008	0.097	
1984	-0.089	0.111	0.001	0.079	
1985	-0.086	0.117	0.017	0.102	
1986	-0.085	0.128	0.020	0.114	
1987	-0.047	0.123	0.008	0.106	
1988	0.002	0.181	0.058	0.107	

Table D-1. Summary of bias and DL for <sup>228</sup>Ac and <sup>212</sup>Pb for each year.

#### D.2 Discussion of Actinium-228 and Lead-212 in Chest Counts

In Figure D-4, chest burdens from 1978, where both <sup>228</sup>Ac and <sup>212</sup>Pb are greater than the DL, are presented (<sup>228</sup>Ac/<sup>212</sup>Pb plots for all years are presented in Section D.4). These data are considered to be above the level of analytical noise and represent actual material in the chest.<sup>4</sup> The diagonal blue line has a slope of 1 and an intercept of 0. Data falling exactly on this line have equal quantities of

<sup>&</sup>lt;sup>4</sup> If the <sup>228</sup>Ac to <sup>212</sup>Pb ratio included the results that are basically noise, there would be quite a range of ratios beyond the anticipated 1:1 that would be very difficult to interpret in a meaningful way.

#### ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)

<sup>228</sup>Ac and <sup>212</sup>Pb. All of these measurements, except one, are considered to indicate equal quantities of <sup>228</sup>Ac and <sup>212</sup>Pb in the chest (to within measurement uncertainty).



Figure D-4. Plot of <sup>228</sup>Ac versus <sup>212</sup>Pb for 1978 where the levels of both nuclides are above the DL. The blue line has a slope of 1 and an intercept of 0.

The lone outlier has an <sup>228</sup>Ac/<sup>212</sup>Pb ratio of about 18. The only plausible explanation for such a large ratio is the presence of unsupported <sup>228</sup>Ra. This suggests that the <sup>228</sup>Ac activity in excess of the <sup>212</sup>Pb activity should be interpreted as a <sup>228</sup>Ra intake and assessed separately from the <sup>232</sup>Th/<sup>228</sup>Th intake estimated from the <sup>212</sup>Pb. Acute and chronic intakes of <sup>228</sup>Ra measured with <sup>228</sup>Ac are readily evaluated with IMBA and do not require custom treatment.

# D.3 Bias and Decision Level Plots

Figures D-5 to D-15 plot bias and decision level. Each figure contains two plots for each year from 1978 to 1988. The first is a normal probability plot of the <sup>212</sup>Pb data, and the second is that for <sup>228</sup>Ac. The median of the <sup>212</sup>Pb data was estimated from a robust regression to the negative (less than zero) data. The median, given as Bias on the plots, was then subtracted from each result. The data on the plots are bias-corrected. The regression was repeated on the bias-corrected data to obtain the standard deviation and mean of the normal background distribution. These parameters were then used to calculate the DL, which is assigned the 95th percentile of the normal background data.

#### D.4 Chest Burden Plots

Figures D-16 to D-26 plot chest burden data by year. Each figure contains two plots, the first on a scale of 0 to 10, the second on a scale of 0 to 1:

- The first figure for each year contains two plots of <sup>228</sup>Ac versus the <sup>212</sup>Pb where both nuclides are above the DL.
- The second for each year contains two plots of <sup>228</sup>Ac versus the <sup>212</sup>Pb where either nuclide is above the DL.





Figure D-5. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1978.





Figure D-6. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1979.





Figure D-7. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1980.





Figure D-8. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1981.





Figure D-9. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1982.





Figure D-10. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1983.





Figure D-11. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1984.





Figure D-12. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1985.





Figure D-13. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1986.




Figure D-14. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1987.





Figure D-15. Normal probability plots of <sup>212</sup>Pb and <sup>228</sup>Ac, 1988.







ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-17. Either radionuclide above the DL, 1978.

ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)

















Figure D-21. Either radionuclide above the DL, 1980.

ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)









Figure D-23. Either radionuclide above the DL, 1981.

ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-24. Both radionuclides above the DL, 1982.

ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-25. Either radionuclide above the DL, 1982.





ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-27. Either radionuclide above the DL, 1983.



Figure D-28. Both radionuclides above the DL, 1984.









ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)



Figure D-31. Either radionuclide above the DL, 1985.



Figure D-32. Both radionuclides above the DL, 1986.



Figure D-33. Either radionuclide above the DL, 1986.





ATTACHMENT D EVALUATION OF ACTINIUM-228/LEAD-212 CHEST COUNT DATA (continued)







Figure D-36. Both radionuclides above the DL, 1988.







#### **TABLE OF CONTENTS**

#### SECTION TITLE PAGE E.1 Background......170 E.2

## LIST OF TABLES

#### TITLE

#### TABLE

Count of EDAC results in HIS-20	170
Activity fractions for the BL-13 EDAC	171
Isotopic composition of K-65 silo 1	172
Isotopic composition of K-65 silo 2	172
Activity fractions for the RT-210 EDAC	174
	Count of EDAC results in HIS-20 Activity fractions for the BL-13 EDAC Isotopic composition of K-65 silo 1 Isotopic composition of K-65 silo 2 Activity fractions for the RT-210 EDAC

#### LIST OF FIGURES

FIGUR	RES	<u>TITLE</u>	<u>PA</u>	GE
E-1	<sup>222</sup> Rn decay chain showing half-lives			74

PAGE

#### E.1 Background

The Fernald site historically used the most restrictive DAC to limit exposures to airborne thorium and its progeny. The standard procedure was to count the air samples in a low-background proportional counter. This instrument was capable of measuring total alpha activity (beta-gamma activity was ignored by the appropriate setting on a pulse-height discriminator). By 1995, it was recognized that this method, while conservative, did not provide accurate results (Allen 1995c). The solution was to develop an effective DAC (EDAC) for each specific mixture of radionuclides that would apply to specific locations and operations on site. The equation used was:

$$EDAC = \frac{1}{\sum \frac{f_i}{DAC_i}}$$
(E-1)

where

 $f_i$  = activity fraction of isotope *i* in the airborne mixture

 $DAC_i$  = DAC of isotope *i* (Toler 1999)

The HIS-20 database was implemented at Fernald in the mid-1990s. Some of the data it contains was migrated from legacy health and safety databases. The HIS-20 database is the source of the DOE-provided BZ data. DOE has also provided data from HIS-20 to NIOSH. The data in the BZ table indicate thorium BZ results first appear in 1993. The first EDAC, designated BL-65 (Building 65), was recorded in 1996. A technical basis for this EDAC had been published in 1995 (Allen 1995). The four other EDACs in HIS-20 are not formally documented in any of the documents captured so far. This attachment provides a method of calculating intakes from the EDAC data for dose reconstruction purposes.

Table E-1 summarizes the EDAC results in the Fernald HIS-20 database DOE provided to NIOSH.

	Count of	Year of	Year of
Radionuciide	results	minimum	maximum
BL-13	924	2003	2004
BL-65	5,260	1996	2003
CELL 8	2	2005	2005
KS-65	23,034	2004	2006
RT-210	489	2001	2006

Table E-1. Count of EDAC results in HIS-20.

## E.2 Affected Locations

The likely definitions for BL-13, BL-65, and KS-65 have been located. These and potential definitions for CELL-8 and RT-210 are discussed in the following sections.

## E.2.1 <u>BL-13</u>

The BL-13 EDAC is documented in Connell (2003). The calculated EDAC for a mixture of  $^{232}$ Th,  $^{230}$ Th,  $^{228}$ Th, and total uranium was 3.28 × 10<sup>-12</sup> µCi/mL. The author recommended that it be rounded

down to  $3.0 \times 10^{-12} \mu \text{Ci/mL}$  to be consistent with another project. The mean activity fractions based on alpha spectroscopy measurements appear in the Table E-2.

Table E-2. Activity fractions for the		
BL-13 EDAC.		
Isotope/element	Activity fraction	
Th-232	0.0504	
Th-230	0.248	
Th-228	0.0565	
U-total	0.645	

Dose reconstructors should multiply the reported number of BL-13 DAC-hours by  $3.0 \times 10^{-12}$  µCi/mL/DAC and the breathing rate ( $1.2 \times 10^6$  mL/hr) to get the total intake. They then should multiply the total intake by the respective activity fractions for <sup>232</sup>Th, <sup>230</sup>Th, <sup>228</sup>Th, and total uranium as follows:

- $^{232}$ Th intake (µCi) = (reported DAC-hr)(3.0 × 10<sup>-12</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.0504)
- <sup>230</sup>Th intake ( $\mu$ Ci) = (reported DAC-hr)(3.0 × 10<sup>-12</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.248)
- <sup>228</sup>Th intake ( $\mu$ Ci) = (reported DAC-hr)(3.0 × 10<sup>-12</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.0565)
- U-total intake ( $\mu$ Ci) = (reported DAC-hr)(3.0 × 10<sup>-12</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.645)

## E.2.2 <u>BL-65</u>

The BL-65 EDAC is documented in *Technical Basis for the Effective DAC for Th*<sup>232</sup> *Stored in Building 65* (Allen 1995). The technical basis shows that the activity of <sup>230</sup>Th is insignificant (less than 3%) in comparison with the other isotopes in this particular mixture (see Attachment B of Allen 1995, p. 8). In addition, its DAC is higher than the calculated EDAC. Therefore, the <sup>230</sup>Th is ignored. The other long-lived isotopes are assumed to be in equilibrium. Therefore, <sup>232</sup>Th, <sup>228</sup>Ra, <sup>228</sup>Ac, <sup>228</sup>Th, and <sup>224</sup>Ra are each assumed to represent 20% of the activity. The resulting EDAC based on total activity is  $2.22 \times 10^{-12} \,\mu\text{Ci/mL}$ . Dose reconstructors should multiply the reported number of BL-65 DAC-hours by  $2.22 \times 10^{-12} \,\mu\text{Ci/mL/DAC}$  and the breathing rate ( $1.2 \times 10^6 \,\text{mL/hr}$ ) to get the total intake. They then should multiply the total intake by 0.2 to get the intakes of <sup>232</sup>Th, <sup>228</sup>Ra, <sup>228</sup>Ac, <sup>228</sup>Th, and <sup>224</sup>Ra, as follows:

- <sup>232</sup>Th intake (μCi) = (reported DAC-hr)(2.22 × 10<sup>-12</sup> μCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr × 0.20)
- <sup>228</sup>Ra intake  $(\mu Ci) = (reported DAC-hr)(2.22 \times 10^{-12} \mu Ci/mL/DAC)(1.2 \times 10^{6} mL/hr \times 0.20)$
- <sup>228</sup>Ac intake ( $\mu$ Ci) = (reported DAC-hr)(2.22 × 10<sup>-12</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr × 0.20)
- <sup>228</sup>Th intake  $(\mu Ci) = (reported DAC-hr)(2.22 \times 10^{-12} \mu Ci/mL/DAC)(1.2 \times 10^{6} mL/hr \times 0.20)$
- $^{224}$ Ra intake (µCi) = (reported DAC-hr)(2.22 × 10<sup>-12</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr × 0.20)

## E.2.3 <u>KS-65</u>

The K-65 silos contained high concentrations of radium-bearing wastes from former production activities. The activity fractions for the two K-65 silos are listed in Tables E-3 and E-4.

The EDAC that was designated "KS-65" was not located in the documentation. Calculations were made for each of the silos using the Equation E-1, above.

The resulting EDACs for total activity were  $3.2 \times 10^{-11} \,\mu\text{Ci/mL}$  and  $2.0 \times 10^{-11} \,\mu\text{Ci/mL}$  for silos 1 and 2, respectively. Using the higher of the two would produce the most favorable intakes. Therefore, dose

reconstructors should multiply the reported number of KS-65 DAC-hours by 3.2x 10<sup>-11</sup> µCi/mL/DAC and the breathing rate  $(1.2 \times 10^6 \text{ mL/hr})$  to get the total intake. They then should multiply the total intake by the respective activity fractions in the above tables. Two calculations will be necessary

Isotope	Activity (nCi/g)	Activity fraction
U-total	1.68	1.61E–03
Ac-227	7.67	7.36E–03
Pb-210	202	1.94E–01
Po-210	281	2.70E–01
Ra-226	477	4.58E–01
Th-228	2.28	2.19E–03
Th-230	68.9	6.62E-02
Th-232	1.11	1.07E–03

Table E-3. Isotopic composition of K-65 silo 1.

Table E-4. Isotopic composition of K-65 silo 2.

Isotope	Activity (nCi/g)	Activity fraction
U-total	2.37	3.04E–03
Ac-227	6.64	8.50E–03
Pa-231	4.04	5.17E–03
Pb-210	190	2.43E–01
Po-210	231	2.96E–01
Ra-226	263	3.36E–01
Th-228	7.36	9.42E-03
Th-230	76.2	9.75E-02
Th-232	0.99	1.26E-03

because not only are the activity fractions different, but also Table E-4 contains <sup>231</sup>Pa in addition to the isotopes in Table E-3. The calculations are as follows:

#### Silo 1

- U-total intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2E × 10<sup>6</sup> mL/hr)(1.61 × 10<sup>-3</sup>) •
- <sup>227</sup>Ac intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(7.36 × 10<sup>-3</sup>) •
- <sup>210</sup>Pb intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(1.94 × 10<sup>-1</sup>) •
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(2.70 × 10<sup>-1</sup>) <sup>210</sup>Po intake ( $\mu$ Ci) = •
- (reported DAC-hr) $(3.2 \times 10^{-11} \,\mu\text{Ci/mL/DAC})(1.2 \times 10^{6} \,\text{mL/hr})(4.58 \times 10^{-1})$  $^{226}$ Ra intake (µCi) = •
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(2.19 × 10<sup>-3</sup>) <sup>228</sup>Th intake ( $\mu$ Ci) = •
  - <sup>230</sup>Th intake ( $\mu$ Ci) = (reported DAC-hr) $(3.2 \times 10^{-11} \,\mu\text{Ci/mL/DAC})(1.2 \times 10^{6} \,\text{mL/hr})(6.62 \times 10^{-2})$
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(1.07 × 10<sup>-3</sup>)  $^{232}$ Th intake (µCi) =

#### Silo 2

•

- U-total intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(3.04 × 10<sup>-3</sup>) • (reported DAC-hr)( $3.2 \times 10^{-11} \,\mu$ Ci/mL/DAC)( $1.2 \times 10^{6} \,m$ L/hr)( $8.50 \times 10^{-3}$ )  $^{227}$ Ac intake (µCi) = •
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(5.17 × 10<sup>-3</sup>) <sup>231</sup>Pa intake (µCi) = •
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(2.43 × 10<sup>-1</sup>) <sup>210</sup>Pb intake ( $\mu$ Ci) =
- (reported DAC-hr)(3.2 × 10<sup>-11</sup> µCi/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(2.96 × 10<sup>-1</sup>) <sup>210</sup>Po intake ( $\mu$ Ci) =
- $(reported DAC-hr)(3.2 \times 10^{-11} \mu Ci/mL/DAC)(1.2 \times 10^{6} mL/hr)(3.36 \times 10^{-1})$ <sup>226</sup>Ra intake (µCi) =

- <sup>228</sup>Th intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(9.42 × 10<sup>-3</sup>)
- <sup>230</sup>Th intake ( $\mu$ Ci) = (reported DAC-hr)(3.2 × 10<sup>-11</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(9.75 × 10<sup>-2</sup>)
- $^{232}$ Th intake (µCi) = (reported DAC-hr)( $3.2 \times 10^{-11}$  µCi/mL/DAC)( $1.2 \times 10^{6}$  mL/hr)( $1.26 \times 10^{-3}$ )

#### E.2.4 <u>CELL-8</u>

The Onsite Disposal Facility (OSDF) was used to dispose of wastes such as contaminated soil and building rubble that were too low-level to justify sending off site. The OSDF was filled in sections, called cells. The waste would have been characterized before being placed in the cells. Cell 8 was the last cell filled. The final cap of cell 8 was installed in 2006. There are only two results in HIS-20 with this EDAC designation, making up less than 0.01% of the results. The results are for two individuals on September 6, 2005. Both wore respirators and both results were below the minimum detectable DAC-hours shown in the HIS-20 records. Neither individual is a claimant. The method for addressing this EDAC has been deferred because it may never be needed.

#### E.2.5 <u>RT-210</u>

The remaining EDAC, RT-210, makes up only 1.7% of the results. There were two radon treatment (RT) systems. Because of the relatively small number of results, it is assumed that the RT-210 results apply to workers at the RT systems. The first system was a temporary arrangement constructed around 2001 that was used to lower radon emissions from the K-65 silos while the material was in storage. Little information has been found for this system. Construction of the new Radon Control System (RCS) for silos 1 and 2 (as well as some other facilities) was completed and operations began in 2003. The RCS was designed to draw radon gas from the headspace inside silos 1 and 2 and reduce the concentration of gas by at least 95% to protect workers during removal of the K-65 material. The silo 1 and 2 wastes were removed by slurry, solidified, and placed in casks. The casks were shipped to an off-site facility for disposal.

The date range for RT-210 is 2001 to 2006, which covers both RT systems. In 2006, the Silos 1 and 2 Remediation Facility, Transfer Tank Area (TTA), and RCS demolitions were completed.

The following description of the Fernald RCS was located in USACE (2011):

Centrifugal fans pulled radon-laden gas from the sources through the roughing filters for initial particulate daughter removal. The air stream was chilled and dried to enhance the dynamic adsorption capacity of the activated carbon. Condensed liquids from the gas stream were transferred to shielded holdup tanks until transfer and disposition in the Remediation Facility could be completed. There were four carbon beds, each containing 20,400 kg (45,000 lbs) of carbon. These beds were configured so that any two of the four beds were in use at any given time. This allowed for decay time of the alternate two beds, whereby no carbon changes were required over the life of the project. The RCS reduced radon concentration to less than 2% of the inlet concentration and the carbon bed outlet air was either recycled to the silos or exhausted through the 46-m (150-ft) tall stack. Approximately four inches of carbon steel shielding was designed and installed adjacent to the carbon beds to reduce general area dose rates ... ventilation requirements were considered for all tasks where radon concentrations greater than 0.01 WL are expected in the air in occupied spaces, such as the TTA and RCS buildings.

The <sup>222</sup>Ra decay chain is shown in Figure E-1. During the operation of the facility, the three long-lived progeny, <sup>210</sup>Pb, <sup>210</sup>Bi, and <sup>210</sup>Po, built up in the system's piping and filters. These isotopes would have been the concern for worker exposures during shutdowns and maintenance (e.g., filter changes). Information from the site indicates that the RT-210 EDAC was most likely calculated assuming equilibrium among the three isotopes (Hinnefeld 2014). The activity fractions are shown in Table E-5.



Figure E-1. <sup>222</sup>Rn decay chain showing half-lives.

RT-210 EDAC.	
Isotope	Activity fraction
Pb-210	0.333
Bi-210	0.333
Po-210	0.333

Table E-5. Activity fractions for the

The following DACs were obtained from 10 CFR Part 835, Appendix A:

- <sup>210</sup>Pb: 1 × 10<sup>-10</sup> μCi/mL,
- <sup>210</sup>Bi: 1 × 10<sup>-7</sup> μCi/mL, and
- <sup>210</sup>Po: 3 × 10<sup>-10</sup> μCi/mL.

From the DAC values and an activity fraction of 0.333 for each isotope an EDAC of  $2.25 \times 10^{-10}$  µCi/mL was calculated. Dose reconstructors should multiply the reported number of RT-210 DAC-hours by  $2.25 \times 10^{-10}$  µCi/mL/DAC and the breathing rate ( $1.2 \times 10^6$  mL/hr) to get the total intake. They then should multiply the total intake by 0.333 to get the intakes of <sup>210</sup>Pb, <sup>210</sup>Bi, and <sup>210</sup>Po, as follows:

- <sup>210</sup>Pb intake ( $\mu$ Ci) = (reported DAC-hr)(2.25 × 10<sup>-10</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.333)
- <sup>210</sup>Bi intake ( $\mu$ Ci) = (reported DAC-hr)(2.25 × 10<sup>-10</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.333)
- <sup>210</sup>Po intake ( $\mu$ Ci) = (reported DAC-hr)(2.25 × 10<sup>-10</sup>  $\mu$ Ci/mL/DAC)(1.2 × 10<sup>6</sup> mL/hr)(0.333)

#### ATTACHMENT F THORIUM IN VIVO COWORKER STUDY, 1979 to 1989

## TABLE OF CONTENTS

<u>SECT</u>	<u>TION</u> <u>TIT</u>	<u>ILE</u> <u>PAGE</u>
F.1	Purpose	
F.2	Thorium Mixtures	
F.3	Thorium Used for MIVRML Calibration	
F.4	Triple-Separation Thorium	
F.5	Data and Method Overview F.5.1 Data Selection F.5.2 Statistical Description Of Data	
F.6	Intake Modeling	

#### LIST OF TABLES

## **TABLE**

PAGE

F-1	Bias for <sup>212</sup> Pb in chest	183
F-2	In vivo lung counting statistics and <sup>232</sup> Th lung burdens	184
F-3	Derived thorium intakes, 1979 through 1989	185

#### LIST OF FIGURES

# **FIGURES**

## <u>TITLE</u>

## **PAGE**

F-1	Natural thorium decay chain showing isotopes and half-lives	. 177
F-2	Relative activity of the isotopes in the natural thorium decay chain as a function of time	
	after chemical purification	. 178
F-3	<sup>228</sup> Th activity after a single chemical purification	. 179
F-4	<sup>228</sup> Ra and <sup>224</sup> Ra ingrowth after thorium chemical separation.	. 180
F-5	<sup>228</sup> Th activity after a second chemical purification at its lowest equilibration point	
	(4.55 years) after the first purification	. 180
F-6	<sup>224</sup> Ra and <sup>228</sup> Ra activity after a second chemical purification when <sup>228</sup> Th was at its lowest	
	equilibration point (4.55 years) after the first purification	. 181
F-7	<sup>228</sup> Th activity after a third chemical purification when at its lowest equilibration point	
	(7.09 years) after the first purification	. 181
F-8	<sup>224</sup> Ra and <sup>228</sup> Ra activity after the third chemical separation	. 182
F-9	50th- and 84th-percentile <sup>232</sup> Th chest burdens, 1979 to 1988	. 184
F-10	Predicted thorium lung burden calculated using the three-separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	50th percentile. Type M	185
F-11	Predicted thorium lung burden calculated using the three-separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989.	
	84th percentile, Type M.	186
F-12	Predicted thorium lung burden calculated using the three separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	50th percentile, Type S	186
F-13	Predicted thorium lung burden calculated using the three separation model to derive	
	thorium intake rates compared with measured chest burden, 1979 to 1989,	
	84th percentile, Type S	. 187

#### F.1 Purpose

Some employees at DOE sites were not monitored for internal ionizing radiation exposure, or the records of such monitoring are incomplete or unavailable. In such cases, data from monitored coworkers can be used to estimate an individual's possible exposure. The purpose of this attachment is to provide information about monitored coworker exposures to thorium from 1979 to 1989. This information may be used for calculating and assigning occupational internal doses to employees at Fernald for whom no or insufficient monitoring records exist.

The thorium intake rates developed in this coworker study are to be used for dose reconstruction for the period from 1979 through 1989. During Fernald Working Group discussions, agreement was reached that the median of the coworker model would be used as a minimum value for intake rate regardless of the data in a worker's internal dose record (NIOSH 2012, NIOSH 2014b). This ensures that no in vivo count underestimates the intake rate due to a count being conducted shortly after chemical separation, before equilibration of <sup>228</sup>Th and <sup>212</sup>Pb can occur.

## F.2 Thorium Mixtures

## **Natural Thorium**

Natural thorium is composed of equilibrated <sup>232</sup>Th and <sup>228</sup>Th and their decay products as shown in Figure F-1. Because the specific activity of <sup>232</sup>Th is small ( $1.1 \times 10^{-7}$  Ci/g) in comparison with <sup>228</sup>Th (820 Ci/g) (Eckerman and Sjoreen 2006) essentially all of the mass in any thorium mixture is associated with <sup>232</sup>Th. In 1.0 g of natural thorium there would be nearly 1.0 g of <sup>232</sup>Th and only 13 ng of <sup>228</sup>Th (Eckerman and Sjoreen 2006).



Figure F-1. Natural thorium decay chain showing isotopes and half-lives. Alpha decays are represented by vertical arrows, beta decays by horizontal arrows (ORNL 2006).

Document No. ORAUT-TKBS-0017-5	Revision No. 01	Effective Date: 07/22/2016	Page 178 of 188
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After a single chemical purification of natural thorium, the isotopic ratio of progeny is disrupted and the degree of equilibrium changes as a function of time while ingrowth of progeny occurs, as illustrated in Figure F-2.



Figure F-2. Relative activity of the isotopes in the natural thorium decay chain as a function of time after chemical purification. All <sup>220</sup>Rn and decay products are assumed to be retained in the thorium material (ORNL 2006).

One of the bounding scenarios examined in this coworker model assumes that the <sup>232</sup>Th, <sup>228</sup>Th, and all progeny are in equilibrium. This is an extreme example that is unlikely to have occurred because natural thorium would have been processed after mining and before arrival at FMPC. However, this scenario could have been realized if the thorium had been refined two or three decades before processing at FMPC.

## F.3 Thorium Used for MIVRML Calibration

The MIVRML was calibrated using a thorium source described in a note published in *Health Physics* (Scott 1966). This was confirmed by an interview with a subject matter expert (ORAUT 2012). The calibration source had an activity ratio of <sup>232</sup>Th to <sup>228</sup>Th of 1.27. In other words, the equilibrium between <sup>228</sup>Th and <sup>232</sup>Th was 0.787. Because of the short half-life of <sup>228</sup>Th progeny, it is reasonable to assume that <sup>228</sup>Th and <sup>212</sup>Pb exist in equilibrium. Note that for this to occur, <sup>220</sup>Rn, a noble gas that is an intermediate member of the decay chain, is assumed to remain in place. Because its half-life is short (56.6 s), this is a reasonable assumption, even though not rigorously true [11].

Thorium material similar to this calibration source would have been chemically purified approximately 1 year before analysis and is deemed to be a realistic mixture, likely to be encountered during operations.

#### F.4 Triple-Separation Thorium

Another bounding scenario considered involves a sequence of thorium purifications that are timed to produce the greatest possible reduction of the ratio of <sup>228</sup>Th to <sup>232</sup>Th (ORAUT 2014b). Because of its long half-life the activity of <sup>232</sup>Th does not change with time, but the activity of <sup>228</sup>Th decreases then increases as shown in Figure F-3. The activity of both isotopes is assumed to be 1 Bq for illustration when chemical purification is completed. This is illustrated in a simpler depiction in Figure F-3.



Figure F-3. <sup>228</sup>Th activity after a single chemical purification. The minimum occurs 4.55 years after purification.

For all practical applications <sup>224</sup>Ra, with its short 3.66-day half-life, is always in secular equilibrium with <sup>228</sup>Th, but <sup>228</sup>Ra, with a 5.75-year half-life, is depleted and takes years to grow back to any significant degree. The ingrowth curves for <sup>224</sup>Ra and <sup>228</sup>Ra are shown in Figure F-4. The minimum in the <sup>224</sup>Ra curve coincides with the minimum in the <sup>228</sup>Th curve.

If the thorium source were again stripped of all progeny at 4.55 years, the <sup>228</sup>Th ingrowth curve in Figure F-5 would result, which has a minimum of 0.26 Bq at 2.54 years after the second separation (or 2.54 + 4.55 = 7.09 years after the first separation). The corresponding <sup>224</sup>Ra and <sup>228</sup>Ra curves after the second separation are shown in Figure F-6.

If this thorium source were again stripped of all progeny a third time at 7.09 years after the first chemical separation, the <sup>228</sup>Th ingrowth curve in Figure F-7 would result, which has a minimum of 0.19 Bq at 1.75 years after the third separation (or 1.75 + 2.54 + 4.55 = 8.84 years after the first separation). The corresponding <sup>224</sup>Ra and <sup>228</sup>Ra curves after the third separation are shown in Figure F-8.





Figure F-4. <sup>228</sup>Ra and <sup>224</sup>Ra ingrowth after thorium chemical separation.



Figure F-5. <sup>228</sup>Th activity after a second chemical purification at its lowest equilibration point (4.55 years) after the first purification. The activity minimum is reached 7.09 years after the first purification.








Figure F-7. <sup>228</sup>Th activity after a third chemical purification when at its lowest equilibration point (7.09 years) after the first purification. The activity minimum, 0.19 Bq, is reached 8.84 years after the first purification.





Figure F-8. <sup>224</sup>Ra and <sup>228</sup>Ra activity after the third chemical separation.

In summary, this scenario results in the equilibrium ratio between <sup>232</sup>Th and <sup>212</sup>Pb reaching a minimum value of 0.19. This chronic intake analysis assumes that all of the activity inhaled always has this most pessimistic isotopic makeup. That is, this scenario results in the highest intake rates and is therefore most favorable to claimants.

### F.5 Data and Method Overview

ORAUT-OTIB-0019, Analysis of Coworker Bioassay Data for Internal Dose Assignment (ORAUT 2005b), describes the general process that is used to analyze bioassay data for assigning doses to individuals based on coworker results. ORAUT-PLAN-0014, Coworker Data Exposure Profile Development (ORAUT 2004), describes the approach and processes to be used to develop reasonable exposure profiles based on available dosimetric information for workers at DOE sites.

The Y-12 MIVRML was used at Fernald from 1968 through 1989. Before its use at Fernald, the MIVRML system was described in a management memorandum to all employees (Heatherton undated). The memorandum explains that those workers with the highest uranium and thorium exposure potential would be counted most frequently and those with virtually no exposure potential would not be routinely counted at all. Workers identified to have potential exposures to thorium were given priority for counting during the first use of the MIVRML (ORAUT 2007) [12].

The technology and challenges associated with in vivo counting for thorium were described by West (1962, 1965). An interview with [name redacted] was conducted (ORAUT 2012) and provided additional specific details on the calibration and operation of the system while at Fernald.

# ATTACHMENT F THORIUM IN VIVO COWORKER STUDY, 1979 to 1989 (continued)

## F.5.1 Data Selection

Handwritten records of in vivo chest count data for individual Fernald workers were obtained (NLO 1968–1988a through NLO 1968–1988v). The great majority of the entries in these records represent data obtained between 1968 and 1988 using the MIVRML. Data from the records from 1979 through 1988 were transcribed into a spreadsheet and validated as described by ORAUT (2006a). A total of 2,994 records from this time interval were transcribed. This represented data on approximately 1,000 individuals. Most results were reported as "Pb-212" and "Ac-228" activities; only the <sup>212</sup>Pb data was used since it is most indicative of a thorium intake (ORAUT 2014b).

The ORAU Team PID reviewed the transcribed data. This review yielded a number of modifications and exclusions from the statistical analysis. The reasons for exclusion included:

- Results labeled "AEC/ERDA/DOE recount program,"
- Control samples,
- Those indicated to be "bad samples" or "unreliable," and
- Entries with no results (most of these appear to have been counts that were scheduled but not performed; comments included "claustrophobic," "off sick," and "count was canceled because of high background problems").

Samples indicated to be recounts or duplicates were averaged into a single result for the given date or sample.

## F.5.2 Statistical Description Of Data

The chest count data were statistically evaluated using yearly intervals according to the specifications and requirements in ORAUT (2006b) and ORAUT (2014c) using the TWOPOS methodology. In vivo chest count data statistics were generated for each analysis interval. The <sup>212</sup>Pb chest counts were determined to be biased, so a bias correction factor was subtracted from each individual chest count as given in Table F-1.

Table F-1.	Bias for <sup>212</sup> Pb	
(nCi) in chest [13].		
Year	<sup>212</sup> Pb bias	
1979	-0.014	
1980	-0.045	
1981	-0.056	
1982	-0.058	
1983	-0.101	
1984	-0.089	
1985	-0.086	
1986	-0.085	
1987	-0.047	
1988	0.002	

A lognormal distribution was assumed [14], and the 50th- and 84th-percentile values were calculated. All calculations were independently repeated for validation. The values were converted to <sup>232</sup>Th lung burdens by dividing by 0.19, as discussed above. Table F-2 presents the annualized results of the statistical analysis for <sup>232</sup>Th in nanocuries. Figure F-9 shows plots of the 50th- and 84th-percentile chest burden.

#### ATTACHMENT F THORIUM IN VIVO COWORKER STUDY, 1979 to 1989 (continued)

Table F-2.	In vivo lung	counting statistics	and <sup>232</sup> Th lung burdens.
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	Effective	# of TWOPOS	50th percentile	84th percentile
Year	bioassay date	results	(nCi)	(nCi)
1979	6/30/1979	124	0.031	0.126
1980	6/30/1980	149	0.027	0.107
1981	6/30/1981	129	0.034	0.113
1982	6/30/1982	167	0.039	0.152
1983	6/30/1983	167	0.026	0.122
1984	6/30/1984	322	0.032	0.108
1985	6/30/1985	355	0.038	0.104
1986	6/30/1986	433	0.031	0.101
1987	6/30/1987	523	0.037	0.097
1988	6/30/1988	108	0.045	0.104



Figure F-9. 50th- and 84th-percentile <sup>232</sup>Th chest burdens, 1979 to 1988.

#### F.6 Intake Modeling

Results from the statistical analyses, adjusted to represent <sup>232</sup>Th activity in picocuries as discussed above, were used to determine thorium coworker intake rates. The biokinetic model described in ORAUT-OTIB-0076 (ORAUT 2014b) was used to fit the in vivo measurement data associated with thorium isotopes to a series of chronic inhalation intakes for both types M and S.

A single chest count result for each period was assumed to have occurred at the midpoint of the period. A uniform absolute error of 1 was applied to all results, which assigned the same weight to each result. The intake assumptions were based on patterns observed in the in vivo data. A single constant chronic intake rate was chosen due to the consistent magnitude of the chest counts.

The resulting coworker thorium intake data are shown in Table F-3. GSD values are equal to the 84th-percentile intake rates divided by the 50th-percentile intake rates. To account for the error in biokinetic modeling, a minimum GSD value of 3.00 is assigned because that is the assumed GSD for an individual who was monitored. The data for 1980 through 1988 are extrapolated to include 1989 [15]. Figures F-10 through F-13 illustrate the result of coworker intake modeling for thorium.

#### ATTACHMENT F THORIUM IN VIVO COWORKER STUDY, 1979 to 1989 (continued)

Intake solubility type	50th percentile (pCi/d)	84th percentile (pCi/d)	GSD	95th percentile (pCi/d)
Туре М	35.3	117	3.00	215
Type S	5.46	17.0	3.00	33.3

Table F-3. Derived thorium intakes, 1979 through 1989.



Figure F-10. Predicted thorium lung burden (line) calculated using the threeseparation model to derive thorium intake rates compared with measured chest burden (dots), 1979 to 1989, 50th percentile, Type M.





Figure F-11. Predicted thorium lung burden (line) calculated using the threeseparation model to derive thorium intake rates compared with measured chest burden (dots), 1979 to 1989, 84th percentile, Type M.



Figure F-12. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest burden (dots), 1979 to 1989, 50th percentile, Type S.



ATTACHMENT F

 Year

 Figure F-13. Predicted thorium lung burden (line) calculated using the three separation model to derive thorium intake rates compared with measured chest

burden (dots), 1979 to 1989, 84th percentile, Type S.

### ATTACHMENT G CLASS W THORIUM-232 INHALATION INTAKE RATE BASED ON 10% OF THE DERIVED AIR CONCENTRATION

100% <sup>232</sup> Th DAC	5.00 × 10 <sup>-13</sup> µCi/cm <sup>3</sup>	
10% <sup>232</sup> Th DAC	5.00 × 10 <sup>-14</sup> µCi/cm <sup>3</sup>	(5.00 × 10 <sup>-14</sup> μCi/cm <sup>3</sup> )(2.22 × 10 <sup>+6</sup> dpm/μCi) = 1.11 × 10 <sup>-7</sup> dpm/cm <sup>3</sup>
		(1.11 $\times$ 10 <sup>-7</sup> dpm/cm <sup>3</sup> is based on 2.22 $\times$ 10 <sup>+6</sup> dpm/µCi)
Work year	2,000 hr/yr	(2,000 hr/yr) ÷ (8 hr/d) = 250 days
Breathing rate (ICRP 1995)	1.2 m <sup>3</sup> /hr	$1.2 \times 10^{6} \text{ cm}^{3}/\text{hr}$ based on $1.00 \times 10^{6} \text{ cm}^{3}/\text{m}^{3}$
OCAS-TIB-009	0.2	Intake per workday, not calendar day = 250 days
Annual inhalation intake of <sup>232</sup> Th	266.400 dpm/yr	$(1.11 \times 10^{-7} \text{ dpm/cm}^3)(1.2 \times 10^6 \text{ cm}^3/\text{hr})(2,000 \text{ hr/yr})$ = 266.4 dpm/yr
Annual ingestion intake of <sup>232</sup> Th	5.550 dpm/yr	$(1.11 \times 10^{-7} \text{ dpm/cm}^3)(1.00 \times 10^6 \text{ cm}^3/\text{m}^3)(0.2)(250 \text{ d/yr})$ = 5.55 dpm/yr