ORAU Team **NIOSH Dose Reconstruction Project** Technical Basis Document for the Fernald Environmental Management Project (FEMP) – Occupational Internal Dose	Document Number: ORAUT-TKBS-0017-5 Effective Date: 05/28/2004 Revision No.: 00 Controlled Copy No.: _____ Page 1 of 42
Subject Expert: Bryce L. Rich Document Owner Approval: Signature on File Date: 05/24/2004 Terry A. Kuykendall, TBD Team Leader Approval: Signature on File Date: 05/24/2004 Judson L. Kenoyer, Task 3 Manager Concurrence: Signature on File Date: 05/27/2004 Richard E. Toohey, Project Director Approval: Signature on File Date: 05/28/2004 James W. Neton, OCAS Health Science Administrator	Supersedes: None

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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
BZ	breathing zone
cm^3	cubic centimeter
CEDE	cumulative effective dose equivalent
DAC	Derived Air Concentration
DL	decision level
DOE	U.S. Department of Energy
dpm	disintegrations per minute
ERDA	Energy Research and Development Agency
FCP	Fernald Closure Project
FEMP	Fernald Environmental Management Project
FERMCO	Fernald Environmental Restoration Management Corporation
FMPC	Feed Material Production Center
GA	general air sample
hr	hour
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICRP	International Commission on Radiological Protection
IVEC	In Vivo Examination Center
KPA	kinetic phosphorescent analysis
LEU	low enriched uranium
m	meter
MAC	maximum allowable concentration
MCW	Mallinckrodt Chemical Works
MDL	minimum detection level
mg U	milligrams of uranium
mg	milligram
MIVRML	Mobile In Vivo Radiation Monitoring Laboratory – ORNL Y-12
MPBB	maximum permissible body burden
MPC	maximum permissible concentration
MPLB	maximum permissible lung burden
MT	metric ton
MTU	metric ton of uranium
NCG	National Lead Concentration Guide (used interchangeably with MAC)
nCi	nanocurie
NLO	National Lead of Ohio, Inc.

ORO	Oak Ridge Operations Office
pCi	picocurie
PGDP	Paducah Gaseous Diffusion Plant
POOS	plutonium out of specification
ppb	parts per billion
RU	recycled uranium
s	second
SRS	savannah river site
TBD	Technical Basis Document
TRU	Transuranic
U.S.C.	United States Code
UCL	upper confidence limit
UNH	uranyl nitrate
UNSCEAR	United Nations Scientific Committee on Energy and Atomic Radiation
WEMCO	Westinghouse Environmental Management Company of Ohio, Inc.
WL	Working Level used for Radon levels = 1.3×10^5 Mev/L
yr	year
µCi	microcurie
µg	microgram

5.0 OCCUPATIONAL INTERNAL DOSE

This section provides the technical basis for estimation of occupational internal radiation dose to workers from the U.S. Department of Energy (DOE) Fernald Environmental Management Project (FEMP; previously known as the Feed Material Production Center, or FMPC).

This Technical Basis Document (TBD) provides information and assumptions for use in reconstructing employees' occupational internal radiation doses. Specific characteristics of the monitoring procedures, identification of events or processes that were unmonitored; identification of the types and quantities of radioactive materials involved, evaluation of production processes and safety procedures, identification of the locations and activities of exposed persons, and identification of comparable exposure circumstances for which data is available on which to base assumptions.

5.1 INTRODUCTION

The discussions in this introductory section derive from information from a variety of references, primarily the DOE Ohio Field Office *Recycled Uranium Project Report* (DOE 2000), *Highly Enriched Uranium Working Group Report* (DOE 1996), and Dolan and Hill FMPC-2082 (Dolan and Hill 1988). These reports summarize the efforts of teams of process engineers and operations specialists (all with long-term plant experience) in reconstructing historical plant operations and effluent experience, and represent the best information available. The information provided in this section includes operational documentation and professional judgment applied to fill in areas where data was missing or inadequate.

Construction at FEMP began in 1951 on a 1,050-acre site near the small rural community of Fernald, Ohio. Table 5-1 lists the three contractors to DOE and its predecessor agencies [i.e., the U.S. Atomic Energy Commission (AEC) and the Energy and Research Defense Agency] that have operated FEMP.

Table 5-1. Contractors during the operational history of the Fernald Site.

Contractors	Dates	Time
National Lead Co. of Ohio (NLO), Inc.	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 (FERMCO, Fluor Fernald, Inc)*	December 1992 to present	+11 yr

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

The original health and safety program was conducted with an industrial hygiene emphasis, based upon uranium heavy metal toxicology, with awareness and utilization of technology from other AEC site programs (e.g., Oak Ridge 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 a greater emphasis on radiation protection principles.

The primary missions at FEMP 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 AEC/DOE sites. The 10 plants that constituted the Fernald facility processed a variety of natural and low enrichment uranium 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 chemicals in processes that were inherently dusty, producing 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 FEMP operations. The work environs included a continuous/chronic potential for internal exposure, as demonstrated by the comprehensive air monitoring program and the urine sampling program for uranium (documented by air sample and urine uranium data sheets). During the early years, plant workers were routinely required to wear respiratory protection because of significant radioactive dust levels approaching or exceeding FEMP airborne alpha activity guidelines referred to as maximum allowable concentrations (MAC) [or National Lead of Ohio Concentration Guide (NCG)] depending on the period (the Lead referred to the name of the contractor, not the contaminant). In addition, over the operational history of Fernald, metric tons (MT) 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 7,300 personnel worked at FEMP 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, transuranics and fission products from recycled uranium, and thorium.

Pitchblende ores from the Belgian Congo, which have 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 from the uranium extraction process were stored in two dedicated concrete silos (by agreement, they belonged to the Belgian government). Additional pitchblende residues from another AEC site (Mallinckrodt Chemical Works) 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 remains on the Fernald site in the K-65 Silos and represents a continuing internal exposure potential (for any operations in which direct contact with these residues is required) from the unusually high concentrations of ^{226}Ra and its progeny, ^{210}Pb and ^{210}Po (these three radionuclides comprise approximately 90% of the total activity in the K-65 materials). The uranium daughter ^{230}Th is also present in significant quantities (7 to 10%) in this location (see Table 5-16). Historical environmental releases from this location, with the resultant internal dose potential, occurred from the ingrown radon and thoron gases and their daughter products.

Fernald processed thorium during from 1954 to 1979 and was the national thorium materials repository for the 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, $Th(OH)_4$] containing primarily, ^{232}Th and its long-lived daughter ^{228}Th. In addition, the beta emitter ^{228}Ra was present. The original thorium inventory was contained in approximately 15,000 storage containers of various sizes. In 2002, the inventory was verified to be approximately 500,000 pounds 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. Recycled uranium (RU) was received and processed at Fernald as early as 1961. Recycled uranium is that which has been used in a reactor environment (such as fuel elements or transuranic production targets) and then processed to separate the unspent uranium from fission products, activation products, and transuranic elements. RU contains trace quantities of plutonium isotopes, ^{237}Np, and ^{99}Tc as the primary contaminants. Some recycled materials from the uranium enrichment facilities (in

the form of tower ashes) had higher than average transuranic contaminant concentrations. The levels of contaminants in these RU materials were known before receipt and blended with other uranium feed stock for processing. Personnel exposed to uranium contamination could also be exposed to the RU contaminants, which could have contributed to unmonitored internal exposure.

5.2 RADIONUCLIDES OF CONCERN

Table 5-2 lists the primary radionuclides that could have led to internal doses during the production history of FEMP:

Table 5-2. Primary radionuclides of internal exposure concern.

Uranium & Daughters		Recycled Uranium Contaminants	Thorium & Daughters	
Isotopes	**Daughters**		**Isotopes**	**Daughters**
U-234	Th-230	Pu-238	Th-232	Ra-226
U-235	Ra-226	Pu-239	Th-230*	Rn-222
U-236**	Rn-222	Pu-240		Ra-228
U-238	Pa-231	Pu-241		Th-228
	Ac-227	Am-241***		Ac-228 [a]
	Rn-222	Np-237		Pb-212 [a]
		Tc-99		Ra-228

[a] These Thorium daughters are listed, since they are used in In vivo counting to quantify the parent Th-232 They do not contribute to the internal dose above 1%

*Present primarily as a uranium decay product.

**Though listed as an expected RU component, U-236 represents <1% of the dose resulting from exposure to uranium.

***Recognized as a potential TRU contaminant in RU and detected with sophisticated analytical techniques in the plant after 1989, but not in personnel and <1% of RU contaminants

5.2.1 Processes

The FEMP production facilities consisted of ten 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.

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. Also, UF_6 was converted to UF_4. On February 14, 1966 an accidental release of approximately 1200 kg of uranium occurred during conversion operations that resulted in elevated personnel exposures (Warner 1966). The enrichment is unknown and 2% should be assumed for any claimant identified as a subject of this incident. In addition, more than 70% of the thorium at FEMP 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 and 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 PUREX[1] 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_3 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, e.g., the Hot Raffinate Building (3E) was utilized to filter insolubles from UNH and to process raffinates.

Plant 4, the Green Salt Plant, which converted UO_3 to UF_4 (green salt), began 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 uranium (MTU) solids 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 may have been processed in Plant 4.

Plant 5, the Metals Production Plant, provided a chemical conversion of UF_4 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 transuranic (TRU) elements and uranium daughters in the MgF_2 slag. The transport and mixing of feed materials and MgF_2 slag led to inhalation exposure potential that were also in the higher level categories at the Site.

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

Plant 7, the Hexafluoride Reduction Plant, was a chemical process facility operated from 1954 to 1957 that 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, 1969, 1970, and 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.

5.2.1.1 Uranium Enrichments

Late in 1964 the Fernald site provided the first production of 1.95% ^{235}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%. The quantities of enriched material above 2% was not documented, but was 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 ^{235}U enrichments ranged from 0.20% to 1.68% with an average of 0.70%.

[1] 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.

Of the total quantity of uranium received and processed at FEMP <25% was enriched above normal (60,181 MTU of the total 246,683 MTU). Approximately 95% (208 gms of the total 218 gms) of the Pu-239 which was received at the Site came in the enriched uranium receipts. (DOE 2000)

Table 5-3 lists the primary assumptions for FEMP uranium enrichments and the isotopes associated with these enrichments. The mass percentages, relative activities in pCi/µg, and the total pCi/µg values are based upon IMBA NIOSH default values.

Table 5-3. Uranium enrichments and associated isotopes.*

Category	Isotopes	Mass %	Relative activity (pCi ug^{-1})	Activity %	Total pCi ug^{-1}
Uranium–depleted	U-234	0.001	0.062	15.46	0.402
	U-235	0.20	0.004	1.07	
	U-236	0.00031	0.0002	0.05	
	U-238	99.80	0.3354	83.42	
Uranium–natural	U-234	0.0054	0.334	48.86	0.683
	U-235	0.7204	0.016	2.28	
	U-236	0	0	0	
	U-238	99.274	0.334	48.86	
Uranium -1% enriched (EU assumption used in early In vivo calculations)	U-234	0.01	0.62	63.7	0.973
	U-235	1.0	0.02	2.1	
	U-236	0	0	0	
	U-238	98.99	0.333	34.2	
Uranium–2% enriched (Recommended dose evaluation default for this TBD)	U-234	0.02	1.243	76.9	1.616
	U-235	2.0	0.043	2.68	
	U-236	0	0	0	
	U-238	97.98	0.329	20.4	

*U-236 is less than 1% activity in DU, Ntl, 1% EU, and 2% EU

In the absence of specific enrichment information, and considering the above available data related to processing experience of uranium enrichments at FEMP, the default assumption for time periods after 1964 is 2% enrichment for bioassay data in milligram quantities of uranium. Prior to 1964 natural uranium should be assumed.

5.2.1.2 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 identified in Table 5-4 was handled in MT 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.

Table 5-4. Uranium chemical forms at the Fernald Site.

Uranium Compound	Chemical Formula
Uranium hexafluoride (gas)	UF_6
Uranyl fluoride	UO_2F_2
Uranyl nitrate	$UO_2(NO_3)_2$
Uranium trioxide (orange oxide)	UO_3
Uranium tetrafluoride (green salt)	UF_4
Uranium dioxide (brown oxide)	UO_2
Uranium tetraoxide	UO_4
Uranium oxide (yellow cake)	U_3O_8
High fired uranium oxides	UO_2
Uranium metal (exposure generally from an oxide)	U

5.2.1.3 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 cleanup 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 release of radioactive materials, spills and routine releases occurred. In addition to the routine releases at FEMP, 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 that the ventilation systems were unable to contain all of the releases.

In 2000 a FEMP team working on the DOE Ohio Field Office Recycled Uranium Project Report (DOE 2000) 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 FEMP 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 that were neither of clearly high nor clearly low radioactive material release potential.

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

Table 5-5. Fernald plant processes, materials, and dust release potential.

Plant	Process	Compound*	Airborne Dust Potential (qualitative)
5	Metal reduction & casting	UF_4, U, U_3O_8, Residues	High
8	Feed Preparation Furnaces	U_3O_8, U, Residues	High
4	Banks 7-9, Packing Stations	UO_3, UO_2, UF_4	High
2/3	Digestion, Extraction Denitration	Prepared Feed, U_3O_8, UO_2, UO_3	Medium
9	Reduction, Casting	UF_4, U, U_3O_8, Residues	Medium
Pilot	Hex Reduction, Metal Operations	UF_4, U, U_3O_8, Residues	Medium
1	Milling	U, U_3O_8, Residues	Low

* MgF_2 (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 MgF_2 typically was associated with nonuranium radionuclides (i.e., uranium daughter products and RU contaminants).

5.2.1.4 Chemical Toxicity

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, natural, and low-enriched uranium. 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 of uranium per gram of kidney tissue (based upon animal studies and some human data) was established by the International Council on Radiation Protection and Measurements (ICRP), and has been used since that time as the reference limit. Concentrations in excess of 3 µg of uranium per gram 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 µg of uranium per gram of kidney tissue, which averages to a kidney burden of 0.337 mg (Rich et al. 1988). Table 5-6 lists a summary of the FEMP chemical toxicity limits with their associated radiological limits.

Table 5-6. Chemical toxicity and associated limits.*

Solubility – Enrichment	mg intake = approx. 0.337 mg kidney – toxicity limit	OSHA Limit		µCi cm^{-3} limit based on rad. exp.
		mg m^{-3} (toxicity)	Equiv. rad. (µCi cm^{-3})	
Sol. – depleted	Type F – 7 mg	0.05	1.8E-11	6.0E-10
Sol. – natural	Type M – 37 mg	0.05	3.5E-11	3.0E-10
Sol. – 2% enriched	*Type S – 696 mg*	*0.05*	*8.1E-11*	*2.0E-11*
Insol. – depleted		0.25	9.2E-11	6.0E-10
Insol. – natural		0.25	1.8E-10	3.0E-10
Insol. – 2% enriched		*0.25*	*1.8E-10*	*3.0E-10*

* *Italicized entries* indicate limits that are controlled by radiological concerns.

The early basis for conducting routine urine analysis was to assure that uranium exposure controls were adequate to prevent chemical toxicity. Additionally 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, derived from urine sample results, were not calculated until the mid to late 1980s.

Though the primary exposure control was based on toxicity concerns, some radiological exposures were reported in addition to a few work restrictions based upon radiological dose limits. The radiological dose determinations were based upon In vivo lung counting. *In vivo* lung counts were routine from 1968 to 1989 using a Mobile In Vivo Radiation Monitoring Laboratory (MIVRML) from ORNL Y-12. From 1989 to 2001 the lung counts took place in the Fernald In Vivo Examination Center

(IVEC). The results were reported in milligrams of ^{235}U and total milligrams of uranium (mg U) as well as in percent of the maximum permissible lung burden (MPLB) or maximum permissible body burden (MPBB). The *in vivo* counting results were recorded in individual dosimetry files and summarized in formal FEMP reports. The *in vivo* lung counts were preferentially used at FEMP to determine the MPLB percentages.

5.2.2 Recycled Uranium (RU) Processes

The first uranium feedstock for the Fernald plant in 1952 and 1953 was yellow cake (U_3O_8) and pitchblende from the Belgian Congo. On February 13, 1961, the first RU was introduced to Fernald in a shipment of UF_6 that was to be converted to UF_4. During the next 3 years, RU was introduced to all of the processes with the plutonium concentrations remaining below the established specifications of 10 ppb of uranium on a mass basis (Bihl 2003). It was calculated that 218 grams of Pu^{239}, 19,048 grams of Np^{237}, and 328,740 grams of Tc^{99} was received at FEMP in 246,683 MTU of RU during the history of the plant. (DOE 2000). In 1976 the decision was made to process tower ash and other residues from the Paducah Gaseous Diffusion Plant (PGDP), which introduced RU contaminants with Plutonium-Out-of-Specification (POOS), i.e. above the 10 ppb established specification. This material was both 1) sent through the solvent extraction process and 2) blended to an oxide. The PGDP shipment accounted for a major portion of the TRU inventory at the Site. (Bassett et al. 1989). The solvent extraction process concentrated TRU in the raffinate, while the blended oxide resulted in storage of uranium with a maximum concentration of Pu of 43 ppb U. The total historical average FEMP concentrations of RU contaminants in the 246,683 MTU (RU) is approx. 1 ppb Pu^{239}, 104 ppb NP^{237}, and 1344 ppb Tc^{99} of RU.

The RU was uranium that had been recovered from irradiated production reactor fuel and plutonium production fuels. The RU was separated in the chemical processing plants at Hanford and Savannah River Site, but was known to contain traces of TRU elements and fission product impurities. Most of the RU delivered to Fernald originated from the Hanford Site, PGDP, and the Savannah River Site. The primary contaminants were ^{238}Pu, ^{239}Pu, ^{241}Am, ^{237}Np, and a fission product, ^{99}Tc. The ^{239}Pu, ^{237}Np, and ^{99}Tc were the radionuclides of greatest concentrations and were tracked and documented for control purposes.

The levels of contaminants were recorded in parts per billion on a uranium mass basis. Limits were set for Hanford product at 10 ppb total plutonium (Bihl 2003). No limits were specified for ^{237}Np or ^{99}Tc. The established 10 ppb plutonium limit resulted in radiological impurities which did not exceed 0.1% of the alpha activity from uranium. Table 5-7, which lists the actual activity percentages, shows that the activity varied with enrichment.

Table 5-7. Comparison of percent total alpha activity with 10 and 100 ppb plutonium-239.

Uranium Enrichment	Uranium Enrichment Activity (pCi μg^{-1})	Pu-239 (pCi μg^{-1}) at:		% Pu-239 of Total U Alpha Activity at:	
		10 ppb	100 ppb	10 ppb	100 ppb
Depleted	0.402	6.175E-04	6.175E-03	0.154	1.54
Natural	0.683	6.175E-04	6.175E-03	0.09	0.90
2% enriched	1.616	6.175E-04	6.175E-03	0.04	0.38

POOS feed stock was sent to Fernald for reprocessing with the prior knowledge that the material was above the routine contamination limits. The intent was to blend these materials with the inventories at the plant and still maintain the plant-wide limits that had been determined to provide an adequate level of safety, i.e., staying below the average site wide plutonium specification of 10 ppb. Workers handling the POOS materials directly (Plant 1 and other locations) were protected with airline respiratory equipment, particularly for the 1976 shipment of tower ashes from the PGDP.

In addition, several of the uranium purification processes resulted in concentrating the TRU contaminants in waste streams. Formal investigations that traced the TRU contaminants through the various plant processes were conducted; one such study defined contaminant affinity for the metal production furnace mold materials in 1975 (some TRU studies are referenced in DOE 2000). These analyses demonstrated that the extraction and metal conversion processes concentrated plutonium, neptunium, and uranium daughter product contaminants in the raffinate waste of the liquid extraction columns and in the MgF_2 from the thermite metal conversion furnaces. MgF_2 slag from the thermite metal reduction process was also processed through the chemical extraction plant. These concentration points or areas were identified by analytical methods in early years. The discussions in this TBD section make use of studies of the data from those early reports (DOE 2000).

The Fernald Health and Safety staff was aware that TRU and fission product contaminants were present in the recycled uranium, although they believed it to be <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.

A review of the Oak Ridge Y-12 radiation protection program and internal dosimetry technology (Cofield 1959a; Scott 1964; Steckel and West 1966; West 1979) indicates that the internal dose technology, techniques, procedures, and philosophy similar to Y-12's were used at Fernald. The fact that the Oak Ridge Y-12 MIVRML provided routine service tp the Fernald *in vivo* internal dosimetry program is an indicator of the close working relationship the Fernald staff had with the Y-12 program. However, this technology during the years until 1986 did not provide adequate detection for TRU or Thorium. Typical results of the MIVRML were uranium lung burdens, although a few thorium analyses results were reported at specific times. Though TRU analyses were attempted, the limitations of the MIVRML, which included limited detection sensitivities for TRU isotopes and infrequent counts, presented capabilities that lacked the ability to detect the anticipated levels at FEMP or in fact could not detect levels that met regulatory limits for TRU isotopes.

A more formal program of internal dosimetry was introduced in 1986. An immediate emphasis was placed on evaluation of the transuranic materials and fission products. From April 1986 to 1989 the site collected 675 plutonium bioassay samples from 441 workers. Only 10 individuals exhibited quantities of plutonium in the urine above 0.02 dpm/sample (this contractual MDL was determined to be unrealistically low and adjusted to the ANSI 13.30 recommended MDL of 0.13 dpm/sample . Further investigation (follow up samples) revealed that 3 of the 10 were below the specified detection limit of 0.02 dpm per 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 conclusion was that the seven of ten were technically at the urine MDL, although so close that the results were questionable false positive. Regardless, the directions were to record the results in the individual dosimetry records and perform a dose estimate calculation. The dose calculation results were not found that would serve the purposes of this report, but could appear in individual claimant files (Bassett 1989). The FEMP radiation protection practices were adjusted to account for those trace isotopes within the DOE radiation protection standards in effect at the time (e.g., DOE 1987).

Before February 1989, no smears or air sampling filters were analyzed specifically for plutonium, neptunium, or thorium isotopes (Basset 1989), although these radionuclides would have been detected by gross alpha counting. In 1989, several sets of air and surface smear samples from Plants 4 and 8 were analyzed for total uranium, $^{239}Pu/^{240}Pu$, ^{238}Pu, ^{237}Np, ^{228}Th, ^{230}Th, and ^{232}Th. Table 5-8 lists the results in derived air concentrations (DAC) from these analyses. DAC ratios are used not for dose reconstruction purposes, but only to indicate the measured relative levels of TRU contaminants in the plant in the 1989 time period, which in turn lends credence to the default assumptions in Table 5-8 for accounting for the unmeasured TRU in the plants.

Table 5-8. Derived air concentrations from the 1989 RU contaminant analyses.

Isotopes	Derived Air Concentrations
Pu-238, 239, 240	0.1-1% of Type S
Np-237	<0.03% to 0.1% of Type S
Th-228, 232	Ranged up to 3% of the Type S
Th-230	6 to 10% of Type S

Note: These initial results indicated that for those samples where the activity fraction of each radionuclide could be compared (generally those with a total alpha activity exceeding E-12 µCi/mL) uranium was the controlling activity in air samples. In addition the results were reported in percentage of the Derived Air Concentration limits in effect at the time, which were also in the range of E-12 µCi/cc.

During the next several years a major sampling and analytical program examined hundreds of process samples, air samples from the process areas, and biological samples for plutonium, neptunium, thorium, technetium, and uranium. Early in 1989 an *in vivo* counting facility was constructed on the Fernald site to replace the mobile *in vivo* counting facility from Y-12.

The process data were used by several FEMP teams to reconstruct the historical RU process experience, starting in about 1985 or 1986; the most recent report was issued May 15, 2000 (DOE 2000). The movement of recycled contaminants and calculated levels were exhaustively reconstructed from records of recycled uranium inventories, RU contaminant analyses in each plant from analytical data from the 1970s and 1980s, and process knowledge of professional staff members, who had been at the plant throughout most of its history (DOE 2000).

Process subgroups were established to guide the reconstruction as indicated in Table 5-9, which lists a summary of the RU contaminants based on the different process/chemistry that the material encountered in the indicated process subgroups. For internal dose reconstruction, this table may be used to develop activity ratios in the absence of specific bioassay data.

Table 5-9. Recycled uranium summary values by process subgroups.*

Subgroup #	Description	Pu-239 ppb U	Np-237 ppb U	Tc-99 ppb U
1A	Miscellaneous	16.035	1328.11	2399.22
1B	Miscellaneous – Minor Offsite	0.889	109.07	0.55
2	UF_6 Source UF_4 (GDP Tails)	0.502	54.90	201.61
3	UF_6 source metal & scrap	0.007	2.54	9.12
4	Normal U product, residues & scrap	0.091	67.09	26.55
5	Enriched UF_6 source products/Res.	1.259	81.39	2109.61
6A	UO_3 Purex source (A508)(Unblended)	2.884	388.97	8552.23
6B	LEUproductsA508UO_3/UF_4 (low cross)	2.321	332.94	8934.58
6C	LEUproductsA508UO_3/UF_4(highcross)	23.969	1045.29	2789.56
6D	A500 Coded Enriched Residues	4.556	143.75	1085.45
6E	SR UNH	16.527	--	--
6F	SR UO_3 – not shipped to FEMP	2.805	--	--
7A	A508 based derbies	9.305	311.97	1721.00

Subgroup #	Description	Pu-239 ppb U	Np-237 ppb U	Tc-99 ppb U
7B	A508 based ingots & Metal	1.165	263.48	447.81
8	Enriched MgF_2	96.618	1881.53	1651.23
9	Incinerator ash& scrap residues-GDPs	47.616	3164.53	263.32
10A	Tower ash & decon. Residues	412.177	10503.53	2618.36
10B	UO_3 from tower ash	20.772	498.17	2405.28
11	Waste residues	84.817	3999.32	4110.05

* Though the results in the table are all reported in ppb U, this measure is meaningless in subgroups in which there is very little uranium, such as subgroup 8, in which the MgF_2 did accumulate some isotopes, but was low in uranium by design.

Because the data from the many process subgroups did not conform well to normal or lognormal distribution patterns, the presentation of the data was not straightforward. In cases of data skew, the use of a "*bootstrap"* analysis is employed, which is a relatively recent form of analysis that employs a sampling approach to approximate representative values for the data set (DOE 2000). Table 5-10 lists the average contaminant concentrations on a site wide basis over the nearly 38-yr RU history (1961-1999) of FEMP. Note: Processing of a single project of RU started at FEMP in February 1961 and was followed in July 1962 with start-up of WINLO process. Processing continued until 1989. Receipts are listed through 1999.

Table 5-10. Plant RU history with 38-year average RU contaminant radionuclides.

Uranium Enrichments	MTU Receipts	Percent Uranium Receipts	Bootstrap Mean Calculations					
			Total Pu-239 (gm)	Pu-239 (ppb)	Total Np-237 (gm)	Np-237 (ppb)	Total Tc-99 (gm)	Tc-99 (ppb)
Enriched	60,180.7	24.4	207.9	3.5	19,047.5	316	328,740	5,382
Normal	89,649.2	36.3	4.1	<0.1	3,025.9	34	1,197.4	13
Depleted	96,853.2	39.3	5.7	<0.1	3,668.7	38	2,060.5	21
Total/ave	246,683.1		217.7	0.9	25,742.1	104	331,998.1	1,346

Because there were no TRU workplace or bioassay contaminant analyses before about 1986, another approach for internal dose reconstruction must be taken on the following bases:

- Only uranium urinalysis was performed routinely from the 1950s to 1986. There was no , direct measurement of TRU contaminants during this period, and the dose determined from only uranium urine results obtained during this period may underestimate a worker's internal dose.

- Direct *in vivo* lung monitoring began in 1968 with the MIVRML and continued in 1989 with the Fernald IVEC counting facility until 2001. No RU contaminant constituents were reported in the analyses before 1989. Although there would have been some ability to detect some TRU materials in the range of 0.1 to 10's of nanocuries depending on the radionuclide, the *in vivo* counts were not performed with a consistency or frequency to be of significant value in TRU dose reconstruction.

- Before DOE Order 5480.11 (effective in 1989), bioassay data at Fernald was not routinely used to estimate intakes and internal organ doses; rather lung counting was used to estimate percent MPLB and infer annual dose by multiplying by 15 rem yr^{-1} per MPLB. Measurements of uranium in urine were compared to limits based upon preventing toxic effects from heavy metals. Workers were removed from the higher exposure jobs when they approached the uranium urinalysis limit or placed on restrictions based on the % MPLB results from *in vivo* counting analyses, or both. However, the in vivo counting frequency seldom exceeded once per year – even for high exposure potential work groups.

- The air monitoring program was used to establish work controls, such as respiratory protection requirements for workers, and was not routinely used to establish internal intake or exposure estimates.

- Annual exposure reports listed uranium lung burdens in percent of MPLB only and did not address the systemic radiological burdens.

- Because the few studies performed indicated historical variability of TRU contaminant levels among locations and processing campaigns, it is not possible to develop an estimate of TRU exposure to individuals based upon their work place history, thus a reasonable default maximum is recommended for all of the processes.

- According to the Fernald Closure Project (FCP) Internal Dosimetry TBD (Tomes 2001) most of the POOS uranium on the site is less than 80 ppb plutonium. However, an examination of the detailed statistical data sheets for the DOE (2000) RU report (summarized in Tables 5-9 and 5-10) indicates that maximum values ranged up to 100 ppb in some subgroups as listed in Table 5-9. One subgroup had a maximum value of 412 ppb. This maximum level was associated with the tower ash from PGDP, which was handled with knowledge and additional protective precautions.

- Though the long-term average RU contaminant levels in the plant are below 10 ppb U for plutonium, there are places and materials in the plant that could have provided RU contaminants above these average values. Based upon the preceding facts and conditions, the most technically defensible and claimant favorable approach to assure that missed internal dose from unmonitored and/or undetected TRU activities (that were present throughout all of the Fernald plants from 1961) were accounted for is to determine the uranium intake and add a ratio of TRU to that intake. Therefore, it would be reasonable and claimant-favorable to add 100 ppb U for ^{239}Pu, 3500 ppb U for ^{237}Np, and 9,000 ppb U for ^{99}Tc to the calculated uranium gram value intakes calculated by the dose reconstruction staff from uranium after 1961. Table 5-11 lists conversion factors for this approach:

Table 5-11. PPB conversion factors.

RU Contaminant	ppb U × (value) = pCi gm^{-1}	ppb U × (value) = Bq gm^{-1}	ppb U × (value) = dpm gm^{-1}
Pu-239	62.89	2.327	139.6
Np-237	0.714	0.0264	1.59
Tc-99	17.15	0.6346	38.07

- The chemical forms of the RU contaminants are not known, although it is apparent from the chemical processes to which the materials were subjected during uranium processing, a variety of forms would be expected. Hence the dose reconstructor should assume the most claimant favorable solubility type for the target organ.

- In the case of evaluating internal intake from long-term calculated environmental air activity concentrations, the approach of adding a fraction of TRU contaminants to the uranium effluent values as a default is recommended. This is based upon the fact that there is no data for TRU effluent other than those calculated from analyses of feedstock contamination levels, a few sets of plant sample analyses, and calculations, based upon process knowledge. Although there were materials in various plant locations that could have been dispersed to the environs at higher levels, by far the bulk of releases to the environs were from stacks and

representative of the average plant contaminants. For consistency, the claimant-favorable RU contaminant levels for plutonium, neptunium, and technetium in Table 5-12 could apply. For example, Table 5-12 is a format guide that presents the calculated percentage increase in count rate of total uranium count on air samples:

Table 5-12. Percent of total uranium activity added to air monitoring samples.

Uranium Enrichment	Uranium ($pCi\ \mu g^{-1}$)	Plutonium 100 ppb		Neptunium 3500 ppb		Technetium 9,000 ppb*	
		($pCi\ \mu g^{-1}$)	% U act.	($pCi\ \mu g^{-1}$)	% U act.	($pCi\ \mu g^{-1}$)	% U act.
Depleted	0.402	6.175E-3	1.54	2.45E-3	0.61	0.152	37.8
Natural	0.683	↓	0.90	↓	0.36	↓	22.3
2% enriched	1.616	↓	0.38	↓	0.15	↓	9.4

*Technetium-99 is a beta emitter and at least 5 orders of magnitude less hazardous by activity. If beta counts were taken on the air sample, it is possible in some unusual circumstances that the count could be predominantly due to the Tc-99.

With an alpha count, which was the preferred analysis method for counting air samples, and at the 100 ppb Pu contaminant level, the alpha count on the filter would normally be less than 1% due to plutonium or neptunium contamination (i.e., the increase would be indistinguishable in the normal uncertainty of the counting statistics). As a default, the calculated air concentrations should be increased by the percentages in Table 5-12. For the 2% enrichment default, the total plutonium activity would be increased by just 0.4%, the neptunium activity by just 0.15%, and the technetium activity by 9.4%.

5.2.3 Thorium Processes

Much of the thorium production data has been lost, and the plant and bioassay monitoring data recovered to date has been sparse. A comprehensive effort to reconstruct the effluent of uranium and thorium from the Fernald plants in 1988 discovered that a large number of records and files were destroyed in the early 1970s during declassification efforts (Dolan and Hill 1988). Reviews of AEC records in Oak Ridge and Atlanta failed to uncover additional details.

Thorium processes had been shut down and most of the thorium processing equipment had been removed prior to the effluent data reconstruction, which made the reconstruction more difficult. The estimates in this section are based on process flow descriptions and available production yields. Where production data was not available, estimates were based on product volume and yield information. These estimates were researched from the various files of FMPC, Oak Ridge, AEC, and FMPC customers. This information was used to develop process flow sheets and locations of possible emission sources that were identified from the components of each process.

Part of the reconstruction process involved interviews of long-time current and retired employees about thorium production. The interviews included the following questions:

- With which processes are you familiar?
- How long did you work in that area?
- What was your job function?
- What type of emission monitoring/control equipment existed in that process?
- What was the production rate?
- What problems were experienced in this operation?
- Where would you say were the greatest sources of emissions?
- How many and what types of scrubbers existed in this system?
- Did all the dry processes vent through a dust collector?

Information from these interviews was used to refine processing times, plant locations, processes, and working conditions.

Though the processes and products involved with uranium production were not precisely the same as those in the processing and production of thorium products, comparison of the volume and mass of the thorium materials with those of uranium is instructive and is used in this report as bases of some default recommendations. The data reconstruction indicates that thorium processing was limited to three plants over short periods in the 38-year production history of FEMP.

In 1972 the site was appointed as the national repository for thorium. For the purposes of dose reconstruction, effective equilibrium is a logical, claimant-favorable assumption because thorium was present from the earliest times and was stored at FEMP after the industry need for thorium products was past. However, for purposes of perspective Figure 5-1 presents thorium equilibrium growth curves.

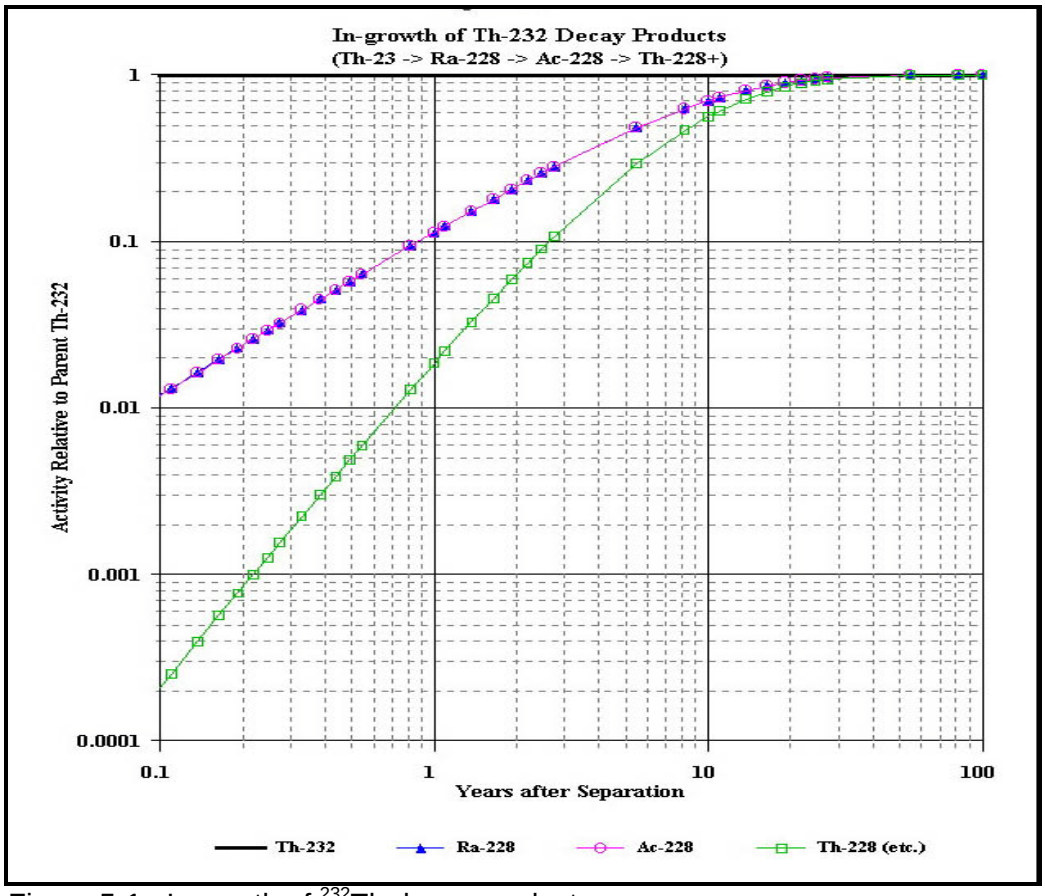

Figure 5-1. Ingrowth of ^{232}Th decay products.

Table 5-13 provides a tabular presentation of the thorium production estimates in MT by year, compared to uranium production in the same plants. The production values in Table 5-13 represent only the uranium and thorium production in the specific areas as identified in the studies. A total of 980,048 MTU was processed site wide, compared to 2,855 MT of thorium (or approximately 0.3% of total uranium mass). In the three plants that processed thorium, the largest quantity (71%) was processed in the Pilot Plant. However, even in this plant, thorium represented only 25% of the total mass of uranium processed. Table 5-14 lists a summary of the thorium plant processes with chemical forms and solubility types.

Table 5-13. Thorium production estimates (MT) by year, with uranium production.

Fiscal Year	Pilot Plant		Plant 9		Plant 8
	Thorium (gel, oxalate, extraction)	Uranium UF_4	Thorium (metal, briquette)	Uranium (metal, briquette, Zirnlo)	Thorium (oxalate, hydroxide)
1953	0	0	0	0	0
1954	0	0	212	0	0
1955	0	0	244	0	0
1956	0	0	0	0	0
1957	0	0	0	0	0
1958	0	0	0	1,735	0
1959	0	0	0	2,965	0
1960	0	0	0	3,289	0
1961	0	0	0	5,603	0
1962	0	1,342	0	6,310	0
1963	0	1,197	0	8,818	0
1964	98	1,468	0	12,698	0
1965	98	658	0	12,850	0
1966	251	478	0	3,165	59
1967	158	13	0	3,095	0
1968	243	0	0	824	0
1969	287	0	0	874	149
1970	249	0	0	603	100
1971	74	0	0	443	61
1972	122	0	0	599	0
1973	49	0	0	452	0
1974	50	0	0	1,031	0
1975	2	0	0	1,189	0
1976	0	0	0	304	0
1977	117	0	0	401	0
1978	117	0	0	558	0
1979	117	0	0	671	0
1980	0	0	0	460	0
1981	0	0	0	827	0
1982	0	0	0	1,050	0
1983	0	0	0	1,523	0
1984	0	0	0	1,766	0
1985	0	622	0	1,287	0
1986	0	462	0	1,984	0
1987	0	542	0	2,787	0
1988	0	1,642	0	4,443	0
Total	2,030	8,424	456	84,604	369

Table 5-14. Thorium plant processes with chemical forms and solubility types.

Plant and Process	Production Years	Chemical Form
Plant 9–metal briquetting	1954-55	Th, ThO_2
Pilot Plant–extraction	1964-80	$ThNO_3$, ThO_2
Gel	1977-79	$Th(OH)_4$
TNT crystals	1966	$ThNO_3$
Metal	1969-71	Th, ThO_2
Oxalate	1971-76	$ThNO_3$
Plant 8–hydroxide	1966	$Th(OH)_4$
Oxalate	1969-71	$ThNO_3$

The ICRP has assigned oxides and hydroxides of thorium to inhalation type S. All other compounds of thorium are type M. The claimant-favorable assumption would be either type M or S, based on the organ of interest, because all of the compounds in Table 5-14 were handled and could have resulted in intakes. The default isotopes would be ^{228}Th and ^{232}Th in equilibrium, since the degree of equilibrium is impossible to estimate due to the variable times of separation of the isotopes from the feed stock (see Figure 5-1). Table 5-15 lists the estimated thorium emissions from FEMP plant facilities in comparison with uranium emissions.

Table 5-15. Estimated thorium emissions (kg) in comparison with uranium.

Year	Total Uranium Emissions	Total Thorium Emissions
1951	123.0	0
1952	499.0	0
1953	2084.6	0
1954	15,345.0	1,028
1955	33,751.4	1,176
1956	15,376.0	0
1957	10,832.0	0
1958	8,844.5	0
1959	8,979.3	0
1960	8,941.5	0
1961	7,202.8	0
1962	6,682.1	0
1963	6,078.0	0
1964	5,295.9	344
1965	7,266.9	344
1966	2,340.9	462
1967	3,572.2	344
1968	5,787.2	344
1969	4,605.7	1,434
1970	1,812.0	1,198
1971	1,062.1	492
1972	1,574.4	141
1973	1,718.6	50
1974	2,677.8	100
1975	3,111.5	3
1976	3,515.0	0
1977	952.3	408
1978	230.6	408
1979	162.6	408
1980	275.2	0
1981	627.0	0
1982	342.5	0
1983	325.0	0
1984	962.9	0
1985	218.7	0
1986	43.8	0
1987	246.9	0
1988	97.5	0
Total	173,574.8	8,684

Realizing that the thorium data are not measurements, but are the best values that the TBD technical staff (Dolan 1988) could reconstruct on the basis of available records, recollections of professional engineers, and best estimates on the basis of process knowledge, this information represents the best available. From these data estimates it is clear that thorium represents less than about 5% of the total emissions from the plant processes and that processing occurred during fewer years. In addition, these emissions give some qualitative indication of the estimated availability of the materials to the airborne pathway. Because the contamination controls for thorium processing (ventilation, etc.) were the same or equivalent to those for the uranium processes, certain assumptions in relation to contaminants in the work place apply to both processes.

The same air sampling procedures were followed for thorium processing as for uranium processing. Some records have been recovered that indicate that basic air activity levels were recorded in fractional MAC (70 dpm m^{-3} prior to 1970 and100 dpm m^{-3} thereafter) for thorium processing. The thorium air sampling results are similar to the uranium air sample results. However, from the limited data examined, the measured MAC levels during the thorium campaigns do not appear to approach the higher MAC levels measured during the processing of uranium. The practices of wearing respiratory protection preventatively for operations known to produce dusty conditions were administered for thorium operations as they were for the uranium operations.

A fundamental difficulty of dose reconstruction for thorium processing is that either 1) *in vitro* bioassays for thorium were not performed or 2) data is not available until after 1986. An additional consideration is that air sampling data was not used to calculate intake and dose until after 1986. Air monitoring was used only to control exposures to levels below the MAC. A number of internal memoranda identify those areas with concentrations at or above the MAC of 100 dpm m^{-3} (4.5×10^{-11} µCi cm^{-3}) as areas requiring respiratory protection. However, recorded examples of exposure to multiple MAC levels without respirators indicate these violations of policy were not uncommon. In addition, the urine sampling was performed for uranium only. The only discovered record of thorium exposure has been *in vivo* lung count data sheets in a few claimant records and a single claimant record which indicates thorium urine results, counted for beta and at essentially no detectable results, from before 1986. Thorium processing was completed in 1979, with exposure from that time being limited to repackaging and shipping operations.

After 1986 thorium air sampling was used to estimate internal exposure using continuous lapel air samples as breathing zone (BZ) evaluations. From that time until the present air monitoring is used to conservatively estimate internal intake even when the worker wore respiratory protection.

The data from the report (Dolan 1988) indicates that just the Pilot Plant, Plant 8, and Plant 9 processed thorium. A single air sampling data sheet was found that indicated a thorium equipment repair operation in Plant 4 during which there were air activity concentrations above MAC. Therefore, the three plants mentioned should be considered the primary processing sites, although there is some evidence that a few isolated thorium operations occurred in other locations. Based on evaluation of the available information, dose reconstructors should assume thorium exposure for any employee whose records establish work, and therefore exposure potential, primarily in the Pilot Plant from 1964 to 1979, in Plant 9 in 1954 or 1955, or in Plant 8 in 1966, 1969, 1970, or 1971.

This reconstruction of the operational history of the large volume thorium processing is the best available at the time of this report. Th^{230} is also found in the plant as a uranium decay product. Most of the thorium from this source is removed from the ores during solvent extraction and is found in quantity in the raffinates. See Table 5-16 for content of the Silos 1 & 2, where Th^{230} make up 7 to 10% of the total activity. Some thorium as a contaminant follows the uranium streams through the plant in trace quantities but will constitute <1% of the thorium default assumptions below.

In vivo counting was performed on the workers in the more likely exposed groups at least once each year. There is some evidence of urine analyses for thorium in claimant files as early as 1955, but to date no information has been found regarding how to interpret it. Although urinalysis can offer some information regarding thorium intake, it is not the preferred bioassay technique, since the material is is predominantly insoluble. Fecal sampling and *in vivo* analyses are the preferred default. This is a difficult default to derive with any degree of technical basis because:

1- There was primarily gross alpha and some gross beta air monitoring during thorium operations for the purpose of controlling worker exposures to below MAC levels. A few *in vitro* analyses for thorium were discovered primarily in claimant file records; only a few *in vivo* analyses were found. The thorium results are questionable because of the lack of information for readily interpreting them (e.g., there is no information regarding the *in vitro* separation method or counting procedure/equipment, nor is there information regarding the assumptions made to derive the *in vivo* results).

2- It is known that respiratory protection (both preventative and following MAC-level air sample results) was provided and would have resulted in at least a factor of 10 protection when used properly. However it is also known that workers were exposed to >MAC levels without

respiratory protection. Considering this information, standard respiratory protection factors cannot be assumed.

3- Limited operation times and smaller volumes and mass (which also would presuppose a more effective ventilation confinement) reduced the exposure potential, all of which would result in an assumption for limited periods of higher-level contamination.

4- The MAC of 100 dpm m^{-3} (4.5×10^{-11} µCi cm^{-3}) is 20 to 100 times larger than the current derived air concentrations for ^{232}Th.

Based upon the above information and assumptions, the recommended claimant-favorable default exposure approach to assign thorium intakes is to assume:

- An intake for an exposure period of 100 hours per year at an assumed exposure of 10 MAC is judged adequate to account for the higher levels of exposure indicated by air sampling, since few samples above 10 MAC were reported and these primarily represented short term maximized sampling (based upon descriptions on the sample sheets). Also typical and more extensive uranium air sample data demonstrate that 10 MAC is a reasonable assumption of the higher level of exposure.

- No respiratory protection factor, although not wearing respirators when air concentrations were above MAC represented procedural violations. This violation was known to have occurred and was not unusual.

- An intake for an exposure period of 500 hours per year at an average air activity of 0.1 MAC during normal operations

Using these assumptions, the claimant-favorable assumption would be:

$$500 \text{ hr} \times 0.1 \text{ MAC} + 100 \text{ hr} \times 10 \text{ MAC} = 1050 \text{ MAC-hr exposure} \quad (5\text{-}1)$$

This exposure results in an intake of about 60 nCi per year. Therefore, in the absence of monitoring data a claimant-favorable default intake is 30 nCi/y (82 pCi/d) each of Th-232 and Th-228 (the alpha emitting isotopes detected on the air samples) plus a 60 nCi/y (164 pCi/d) intake of Ra-228. The Ac-228 beta emitter adds about 0.1% to the effective dose and therefore can be ignored in the dose calculations. Although an equal intake of Ra-224 might have occurred, because of its alpha emissions, it is adequately accounted for in the thorium intake assumption, which is based on detection of all alpha emitters on the air samples. This default intake rate applies to the most exposed craft personnel (e.g., chemical operators, process maintenance personnel, safety personnel, and first line supervisors/foremen) at the locations and during the periods noted below. For workers whose location cannot be determined the claimant favorable assumption is that they were exposed to thorium and daughter products when they were employed during the listed periods in Table 5-16. Exposures to casual workers who worked in the immediate vicinity of the plants should be evaluated in the Environmental Occupational Dose section.

Thorium chest counts may indicate that lower exposures occurred. When available, the chest count data should be used to constrain an employee's intake. In other words if there are chest count results for an employee, the smaller of the default thorium intake or the chest count determined intake should be assigned for full dose reconstructions.

Table 5-16 Default thorium exposure.

Period	Area	Worker categories	Intakes (pCi/d)
1954-1955	Pilot Plant	Chemical operators, process maintenance personnel, etc.	Th-232 82 Th-228 82 Ra-228 164
1964-1979	Plant 8		
1966, 1969-1971	Plant 9		

5.2.4 K-65 Silo Processes

Information on the K-65 silo processes is derived from air sample and external radiation dose data sheets from the 1953 period, Krach 1998, Tomes 2001 and RAC 1995. The raffinates from the Plant 2/3 processing or uranium ores were relatively high level wastes (in the μCi/gm range) and required special storage. The waste especially from processing African pitchblende was particularly high in ^{226}Ra (and daughter products) content. Large concrete tanks, called K-65 silos, were constructed from August 1951 to July 1952 for the "interim" storage of the pitchblende waste, since it technically belonged to the African Metals Corporation. The chemical extraction plant 2/3 was complete and started operation in December 1953 and disposed of the raffinates directly to the storage silos 1 and 2. However, Mallinckrodt Chemical Works (MCW) facility in St. Louis had processed African pitchblende ores prior to completion of the Fernald extraction process facilities and the K-65 storage silos, and did not have sufficient storage space. Therefore, MCW shipped their pitchblende raffinate wastes in lined 55-gallon drums to Fernald for interim storage. Shipments from MCW began in 1951 and by the end of July 1952, when the silos were complete, approximately 13,000 55-gallon drums of K-65 (approximately half the capacity of one silo) were stored on the ore storage pad around Plant 1, the Sampling Plant. The drummed waste was transferred to silos 1 and 2 from July 1952 through September 1958. The total of the radium-containing residues in Silos 1 & 2, resulting from the processing of uranium ores is approximately 10,000 MT.

Silo 3 was used for the raffinate storage from "cold metal oxide" extraction separations and contains approximately 138,000 cubic feet of raffinate. The feedstock for these processes was uranium concentrates from a variety of uranium mills in the United States and abroad. Though this material contains radioactive material, most of the ^{226}Ra had been removed at the mills and the remaining levels were not a significant source of exposure in comparison with Silos 1 and 2. The materials in Silo 3 were calcined prior to storage and are a fully-oxidized, fine powder in contrast to the K-65 material in Silos 1 & 2, which are approximately 30% moisture. Silo 4 was never used as a waste storage silo and contains only a small amount of low level contaminated water.

The silo waste became the property of the United States in 1983 and has become 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 daughter products. A dirt berm was added around the silos (primarily as silo wall support but provides shielding also), as were roof sealants and a semi-permeable cap on the waste itself, all of which reduced the routine releases of radon plus daughters 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 of July 1953 to September 1958 was an operation in which external and internal

exposure to the radium and other associated contaminants occurred. In addition there were several operations to improve the confinement of the silos for the radon gases and associated daughter products, although the waste materials have not been directly disturbed since they were put into storage. Air sample data sheets from the 1953 time period have been found and provide insight into the operation and exposure levels. The only bioassay information related to internal exposures to radium or the associated contaminants are a few radion breath samples in the 1953 time period. In vivo counts were periodically performed by the Y-12 mobile counting laboratory; however these in vivo counts did not begin until 1968, a long period after completion of the handling and dumping of the 13,000 drums of waste.

Table 5–16 is a summary of the radiochemical analyses of Silos 1 and 2 core samples taken in 1993, and is provided for internal dose reconstruction purposes in visualizing the potential particulate source term (Tomes 2001). It should be noted that in Silo 1 approximately 92% of the total activity is composed of ^{226}Ra and the longer lived daughters ^{210}Pb and ^{210}Po. In Silo 2 the same isotopes comprise approximately 88% of the total activity.

Table 5–16. Isotopic composition of K-65 Silos 1 and 2.

Isotope	Silo 1		Silo 2	
	Activity (nCi/gm)	Activity Fraction	Activity (nCi/gm)	Activity Fraction
Uranium – Total	1.68	1.61 E-3	2.37	3.04 E-3
Ac-227 (β)	7.67	7.36 E-3	6.64	8.50 E-3
Pa-231 (α)			4.04	5.17 -3
Pb-210 (β)	202	1.94 E-1	190	2.43 E-1
Po-210 (α)	281	2.70 E-1	231	2.96 E-1
Ra-226 (α)	477	4.58 E-1	263	3.36 E-1
Th-228 (α)	2.28	2.19 E-3	7.36	9.42 E-3
Th-230 (α)	68.9	6.62 E-2	76.2	9.75 E-2
Th-232 (α)	1.11	1.07 E-3	0.99	1.26 E-3

The information that is available upon which to base estimates of intake of radium and its daughter products to an unknown number of workers consists of a few air sample data sheets in late 1952 to early 1953 with alpha analyses ("O" drive Data Capture\601-881\010000784\000784.pdf) and personnel dosimetry records of external dose to workers specifically identified as working on the MCW raffinate dumping operation ("O" drive Data Capture\601-881\010000867\000867.pdf). The basic facts from this data are as follows:

- Movement of the 13,000 55-gal drums of MCW pitchblende raffinates to Silos 1 and 2 during the six-year period of July 1952 to Sept 1958 (74 months) could have been a continuing effort averaging 176 drums per month (approximately 8 per day). However, one of the air sample data sheets in 1953 indicated an 80 drum per day rate (assumed to be a 3 shift-24 hour day), or 3.3 drums per hour, this seems reasonable, based upon other notes relating to the need for shoveling the contents, "knocking" on the barrels, etc. At a rate of 80 drums per day the movement could have been accomplished in 163 days.

- The description of this operation was inferred from brief descriptions contained on the air sample data sheets from 1952 and 1953. Both elevated air sample activity and the existence of elevated radiation levels were clear from the sampling location descriptions, e.g. "behind shielding", etc. and analytical results. The operational descriptions came primarily from descriptions on air sample sheets, i.e. removing/replacing lids and liners, beating the rim to dislodge contents,

digging into the drum contents with a shovel, standing on the conveyer, sweeping and washing, etc.

- From the information in the series of air sample data sheet analyses in 1952 and 1953, it is clear that an air monitoring program was in place. The results indicate both GA (5 to 30 minute duration) and BZ (1 to 5 minutes) sampling with results <0.2 MAC, many >1 MAC, and a few in the 50-60 MAC range - and one at about 180 MAC. On a couple of data sheets there was a note that no respirators were worn. It is presumed that the note was made to record an unusual event, since standing air monitoring policy was for the purpose of evaluating the need for respiratory protection.

- There were air samples taken for the determination of radon levels during the barrel dumping operation (as inferred from a single "Air Radon Sample" data sheet on 10-29-53), the results of which were 1 and 2.3 x 10^{-10} Ci/L Rn.-or 1 to 2.3 WL.

- No records were found of any bioassay results for radium or daughter products during this time period – with the exception of a series of radon breath samples. No In vivo counts were made on personnel during this period.

- The information on the data sheets indicates that in spite of the fact that the contents of the drums were wet, the operations resulted in significant airborne contamination.

- A group of external dose data sheets were found during the information search that recorded radiation doses for 22 workers specifically identified as working on the K-65 dumping operation. Conclusions derived from these data sheets can be summarized as follows:

    - Dosimeters were processed weekly during 10 one-week time periods from 9/8/52 through 11/31/52.

    - During this period the 22 assigned individuals worked for periods ranging from 1 to 6 weeks (average 4.7 weeks) each and received an average of 275 mrem/week.

    - Of those 13 workers who received the highest doses (ranging from158 to 500 mrem average/week) 3 worked 5 weeks and 10 worked 6 weeks with an overall average of 312 mrem per week.

    - The highest individual dose in any week was 1200 mrem. Though radiation dose rate survey records are not available, it is apparent that working levels would be in the 40 to 60 mrem/hr., i.e. 1200 mrem/20 to 30 hours per week in the highest radiation fields = 40 to 60 mrem/hr. Actual radiation fields could have been higher.

    - Work schedules on 1/16/52 recorded 3 shifts of 4 workers per shift, indicating that during the most intense transfer period work proceeded 24 hours per day.

    - Assuming that those workers receiving the highest weekly doses were those workers who would also be the most likely to be in the highest particulate air activity, the external dose limits would restrict exposure times to approximately 3 months per year, i.e. (4000 mrem/312 mrem wk^{-1}) $(4.3\ wks/month)^{-1}$ = 3 months. This assumes that doses above 4 rem would not be administratively planned to avoid exceeding the 5 rem/year limit. In any event external dose limits would restrict personnel exposure to the recorded maximum air activity to approximately 3 months per year.

- From the information derived in the external dose data sheets and the air monitoring sample sheets, it appears that the transfer could have been limited to a period of 10 weeks per year with no individual working more than a period of 6 weeks in the year in order to control external dose within the regulatory limits. This could have resulted in 80 drums per day x 10 weeks x 5 days per week = approximately 4000 barrels per year, which would have been double the rate necessary to transfer the 13000 barrels in the 74 month time actually utilized by the Site.

For internal dose reconstruction of workers in the Silos 1 & 2 areas in the absence of specific in vitro and/or in vivo data, a maximization approach, based upon air sample results, is proposed:

- assume a worker was assigned to all of the dumping operations during 6 weeks of each of the years in the 6 year period;

- maximum dumping rate, and hence maximum exposure rate, 80 drums/day; and

- 100 MAC (4.5 E-9 uCi/ml alpha analysis) exposure levels with no respiratory protection at 9.6 E+06 cm^3 per day breathing rate. The calculation is:

  4.5 E-9 µCi/cc x 9.6E+06 cc/day x 30 days = 1.3 µCi intake per year for the period of July 1953 to September 1958. The contaminants are assumed to be composed of Table 5-16 Isotopes in their indicated activity fractions. For purposes of dose reconstruction, it should be noted that the MAC air concentrations were determined by gross alpha counts, requiring the addition of the appropriate percentage of the beta emitting isotopes.

From a single radon sample data sheet on which the analyses of two samples were recorded on 10/29/53, the higher of the two samples indicated a result of 230 pCi/L radon gas, which verifies the logical assumption that radon gas was released as the drum lids were removed. In addition to the default particulate intake (determined as previously stated), a conservative/bounding analysis of possible radon plus daughter product exposures can be derived:

- Assume 230 pCi/L (2.3 WL) with 100% daughter product equilibrium for 1304 hrs. (163 day x 8 hr/day)/ 74 months of the dumping operations = 17.6 average hours/month exposure. Then 2.3 WL 17.6/167(the fraction of a full working month) x 12 months = 2.9 WLM exposure per year.

This assumed bounding exposure to radon plus daughter products would be in addition to any assigned exposure that may be derived from the Section 4, Occupational Environmental Dose calculations.

It is evident that these estimates are based upon assumptions that are cumulatively conservative, claimant favorable, and 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. Calculations of internal intakes resulting from exposures to the raffinate dusts generated during dumping operations should be used only for claimants for whom a work history on this specific project can be established. An examination of external penetrating radiation dose for workers who were known to have worked with and handled these drums of raffinate wastes show significant (several100 mrem per week) penetrating dose accumulation. Therefore a criteria to determine and/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.

As previously stated, the contents of the silos have not been disturbed during storage to any large degree. However, it has been calculated that during the 1953 to 1978 period 5,000 to 6,000 Ci/year of ^{222}Rn were released from the silos (RAC 1995). Considering the expected large differences in release rates due to barometric pressure changes, the release rates would average up to 15 to 20 Ci/day after addition to the Silos were complete. Again, no monitoring or operational descriptions of the maintenance and/or silo upgrades are available, which would involve extensive work near the silos at the time of this writing. If it is known that a specific claimant was a part of a crew which spent significant time near the silos during the upgrade operations, intake of Rn^{222} plus daughters will best be calculated as defined in section 4, Occupational Environmental Dose from the data in Table 4-9a.

Radon and thoron plus daughter product sampling was conducted inside each plant building at the Fernald Site primarily in the late 1980s (Weaver 1987). Documentation at that time indicated levels in terms of percentage of 0.4 WL (ICRP 32 limit) generally in the 0.5 to 7% range, with a few locations that ranged in the 10 to 30% levels. This would indicate minimal occupational internal exposure above normal expected radon/thoron backgrounds from the Silos source. However, a default of 5% of 0.4 WL could be added to plant exposures. This equals an exposure of 0.24 WLM per year.

5.3 RADIOLOGICAL CONTROLS PROGRAM

The initial health and safety organization (the Industrial Hygiene and Radiation Division) at FEMP was organized and directed by an occupational medical physician and staffed primarily with industrial hygienists. The Fernald staff worked with the Oak Ridge National Laboratories Y-12 staff, because of similar radiation safety issues associated with their uranium processing, although Y-12 typically had a more highly enriched uranium source term. FEMP also used Y-12's Mobile In Vivo Radiation Monitoring Facility (MIVRML).for over 20 years.

5.3.1 Radiological Controls 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 FEMP 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 FEMP 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 two-day work break to allow elimination of uranium cleared rapidly via the GI tract (this material causes relatively little dose)
- *In vivo* analysis once a month for high exposure-potential workers 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 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.

5.3.2 Air Monitoring Program

The large quantity of data in the archives shows that FEMP maintained an aggressive air monitoring program from the beginning of operations. Both high and [primarily] low-volume general area (GA) and breathing zone (BZ) air samples were collected (most for 3 to 30 min) 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 National Lead of Ohio MAC guidelines or the National Lead Concentration Guide (NCG) of 100 dpm m^{-3} (70 dpm m^{-3} was used as the MAC/NCG until the 1970s). The 100 dpm m^{-3} equates to about 4.5×10^{-11} µCi cm^{-3}.

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.

Since 1989 BZ samples were (and continue to be) converted to DAC-hours in the case of thorium exposures. On occasion, these air sample analyses are converted to intake, dose, or both. The calculated doses observed in the records were low, i.e., 10s of mrem CEDE. The air sample results are used with the thorium isotope (230 or 232) and a conversion to % DAC (2E-02 Bq m^{-3} for Th^{232} and 9E-02 Bq m^{-3} for Th^{230}), and then converted directly to dose in CEDE.

For reference, Table 5-17 lists the abbreviations and codes on the sample and dose calculation datasheets. Table 5-18 cross-references FEMP air-monitoring methods with the applicable MDLs.

Based on the preceding information on FEMP air monitoring, the following default explanations and assumptions may be applied for the purpose of dose reconstruction.

Table 5-17. Air monitoring analysis sheet codes.

Code	Description
MAC	NLO maximum allowable concentration – 100 dpm m^{-3} and 70 dpm m^{-3} prior to the 1970 time period
NCG	NLO concentration guide – used interchangeably with MAC
R	Sample collection rate in m^3 min^{-1}
T	Sample time in minutes
Q	Sample volume in m^3 $Q = R \times T$
GA	Sample collected in a general area
BZ	Sample collected as close to the breathing zone 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 non-radiological samples

Table 5-18. Cross-reference of FEMP air-monitoring methods with the applicable MDLs.

Type of Analysis	Method	Period	Frequency	MDL*
Air sampling	Breathing zone sampling	1952–1993	Variable and frequent, depending upon work	0.2–0.02 MAC depending on sample volume
Air sampling	Breathing zone lapel samplers	1993–present	Job specific: when Th-230 or Th-232 exposure potential exists. BZ Sampling for Uranium is also performed for work controls	1–2 DAC-hrs with a Minimum Detectable Intake** of 0.001 - 0.002 corrected DAC-hrs. for thorium

* From 1952 to 1993 BZ samples were taken 5 to 30 min at 20 L min^{-1} (0.1 to 0.6 m^3,of air) while today the lapel sampler is worn continuously at about 4 L min^{-1} for 8 hours/day (approximately 2 m^3 of air collected) and then counted for longer times with more sensitive laboratory counters. ** Detectable Intake is lower due to the application of a respiratory protection factor.

From 1953 to 1986, the air monitoring program was conducted as a primary control element. However, the measured air concentrations 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 air sample-based controls. The MDL for routine air sampling was in the 0.2 to 0.02 MAC range, and when multiplied by an 8-hr workday, MDL levels in MAC-hr of approximately 1.5 oer day 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-min sample (0.06 m^3) in dusty areas, which equates to the 1.6 MAC-hr MDL, and up to a 30-min sample (0.6 m^3) 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.

Current (1993 to present) air monitoring is performed with the intent of calculating thorium intake and dose if necessary for low-dose cases in addition to maintaining site control of personnel exposures. The air sample data is 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 is directly converted to intake and organ dose, if the results are positive.

5.3.3 Bioassay Program

A urinalysis program was administered at FEMP starting in 1953 or possibly before. The NLO industrial physician and industrial hygienists performed and documented a number of studies to establish the uptake or MPBB for workers exposed to uranium. The initial study was based solely on heavy-metal toxicological limits for kidney damage. Table 5-19 lists the derived limits, which remained essentially unchanged throughout the history of the site.

Table 5-19. Routine uranium *in vitro* bioassay capabilities at the FEMP.

Type of analysis	Method	Period	Frequency	MDL
Urine - uranium	Fluorophotometry	1952–1993	Weekly to annual – job specific	14 µg L^{-1}*
	Chemchek KPA	1993 to 9/2002	Bimonthly	0.17 µg L^{-1} (total U)
	ICP-MS	Sept. 2002 to Present	As requested	0.15 µg L^{-1} (total U)
Fecal**	Fluorophotometry	Various	No routine schedule	Unknown – assume environmental levels of 2 µg per sample
	Contract lab	Occasional	No routine schedule	0.1 pCi per sample

* Y-12 listed a sensitivity of 1.6 µg L^{-1} in 1973 using the fluorometric process for 0.7% U-235. Fernald frequently listed less than 0.003 mg L^{-1} in the bioassay data reports. Several blank samples on intercomparison studies also list results as 0.003 mg L^{-1}. A value of 0.008 mg L^{-1} has also been quoted in the records as the MDL. However, a formal response on January 21, 1993, (Blalock 1993) to a deficiency in the

ability to detect 100 mrem CEDE with the existing 0.014 mg L^{-1} MDA is accepted as the most reliable representation for historical MDAs for this analytical procedure.
** 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.

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 least] bimonthly.

5.3.4 Environmental Levels and Fecal Sampling Program

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 Health and Safety Laboratory – National Lead of Ohio (HASL-NLO) Study (as recorded on analytical data sheets of October 4, 1968). However, fecal sampling has never been a part of the routine bioassay program at FEMP, except in special conditions.

As documented in the most recent FCP Technical Basis for Internal Dosimetry at the Fernald Closure Project, SD 2008 (Tomes 2001), UNSCEAR and ICRP publications estimate natural intake of uranium in the general U.S. population in the range of 0.7 to 2 µg a day with an expected excretion of 0.05 to 0.5 µg a day in urine and 1.4 to 1.8 ug per day in the feces:

> *"Current bioassay methods are sufficiently sensitive to detect dietary intakes of uranium in urine. Therefore, an environmental Decision Level (DL) was determined and used in place of the MDA for establishing bioassay sampling frequency. The DL was determined from the measured results of samples analyzed from newly hired employees (a control group) who had no known occupational exposure to uranium. The DL was set at a 99.99 percent confidence interval. This means a sample whose concentration measures above this level has a 10^{-4} probability of being the result of dietary intake. This would be a false positive result for occupational exposure. Therefore, a value above the DL is considered indicative of occupational exposure and requires additional sampling and follow-up. The DL for uranium was determined to be 0.8 µg U/l [of urine]" (Tomes 2001).*

The same procedure was followed at other DOE sites to determine the occupational indicator. The DL value for urine at the Idaho National Engineering and Environmental Laboratory, for example, has been established at 0.16 µg L^{-1} (Rielly 2001).

It has been recognized at Fernald, particularly since 1986, that fecal sampling can provide useful information, particularly in cases of exposure to less soluble compounds (types M & S). However, it is also recognized that the natural environmental levels vary considerably and for that reason are less reliable for routine use. FEMP did not conduct routine fecal sampling; however, since the mid 1990s, when fecal samples were collected, they were analyzed under contract with a specified maximum MDA of 0.1 pCi per sample. If lower sample MDAs are found in records associated with contracted fecal uranium analyses, these lower MDAs should be used. Uranium results that are within a factor of 10 of the DL should be adjusted for full dose reconstructions.

5.3.5 Analytical Program

The urinanalyses procedure 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. As indicated in Table 5-19, the MDA (MDL) was not formally established in the early periods (as evidenced by a records search). In addition, the large fluctuation in uranium in the diet of nonoccupationally exposed personnel provided implied limits (although apparently not well understood in the early periods). The values listed in the bioassay data sheets generally range from 0.003 mg L^{-1} to 0.008 mg L^{-1} in urine. The effective MDA is now understood to have been near 0.014 mg L^{-1}; therefore, values below this can be accepted as positive but without statistical value as indicators of occupational exposure.

In early 1993 other DOE laboratories were changing to kinetic phosphorescent analysis (KPA), with detection capabilities as low as 0.02 µg L^{-1}, which is far below the natural environmental background in most people. The MDA of 0.17 µg L^{-1} was established consistent with other laboratories. The DL of 0.8 µg L^{-1} at FEMP since 1993 represents the best estimate of the non-occupational excretion of uranium, although values below the DL are reported in the dose history records. Specifically, when Fernald changed to the KPA, 0.8 µg/L became the reporting level until 1997, after which all calculated results were reported as derived. Some database printouts for samples after 1997 will include < signs for results that are less than 0.17 ug/L, e.g., <0.06 µg U/L. These results should be interpreted as less than 0.17 ug/L. This represents a database formatting issue rather than a lowering of detection limits.

The method currently being used at Fernald for urinalysis is inductively coupled plasma mass spectrometry (ICP-MS) which has an *a priori* MDA of 0.15 µg L^{-1}.

For reference, Table 5-20 lists the FEMP uranium urinalysis sample type codes. Table 5-21 lists the FEMP sample type coding system and personnel assignment codes.

Table 5-20. Uranium urinalysis sample type codes.

Types of Samples – General	
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
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 prior to 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

Types of Samples – General	
Code	**Description**
BDL	Below Detection Level
NA	Not Applicable/Available

Table 5-21. FEMP sample type coding system and personnel assignment codes.

Sample Type Coding System – Specific	
1	During 1st hour of shift
2	During 2nd hour of shift
3	During 3rd hour of shift
4	During 4th hour of shift
5	During 5th hour of shift
6	During 6th hour of shift
7	During 7th hour of shift
8	During 8th hour of shift
9	After completing work
Sample Type Coding System – Further Definition	
5A	Off the job, overnight composite sample
5B	Off the job, overnight individual specimen
5C	Sample to test for possible correlation with abnormal clinical lab findings
5D	24-hour individual sample from confined patients
5E	24-hour composite sample from confined patients
5F	24-hour individual sample from unconfined patients
5G	24-hour composite sample from unconfined patients
5H	On-the-job individual sample collected in the work area
Personnel Assignment Codes	
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	Plant 6 and 9 Machining
270	Pilot Plant
280	Plant 8
430	Boiler Plant
436	Mechanical Department
452	Cafeteria
455	Security Department
462	Stores
463	Transportation Department
465	Garage
466	Service – Porters & Laundry

5.3.6 *In Vitro* Procedures for Other Radionuclides

As previously stated, the fundamental and primary bioassay for the first 35 years (1951 – 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 the TBD efforts did not reveal a dose record with non-uranium urinalysis results. There are records of special studies, but no documented intent to analyze for radionuclides other than uranium. The primary contract laboratory for FEMP *in vitro* analyses was United States Testing Company in Richland, Washington. Table 5-22 lists MDAs for various analyses.

Table 5-22. MDAs for non-uranium radionuclides.

Type of Analysis	Method	Period	Sample Frequency	MDA*
Urine – plutonium	Chemical extraction/ gross count	Prior to 1988	No routine schedule – unknown	~0.1 pCi/sample
	Extraction/alpha spec. count	1990s to present	Special study samples only	0.1 pCi/sample
Fecal – plutonium	Extraction/alpha spec. count	1990s to present	Special study samples only	0.1 pCi/sample
Urine – thorium	Extraction/alpha spec. count	1990s to present	Not performed	0.1 pCi/sample
Fecal – thorium	Extraction/alpha spec. count	1990s to present	Only in cases of significant exposure	0.1 pCi/sample

* 0.1 pCi/sample was and is the contractual MDA for all analyses of this type, since the mid 1990s .

As early as 1958, the Fernald site reported internal dose experience to the AEC in an annual report. Table 5-23 summarizes the data from 1958 to 1966. The data provide instructive indication of recorded annual urinary experience in summary form.

Table 5-23. Urinary uranium averages summary.

Urine Average ($\mu g\ L^{-1}$)*	Total Number of Exposed Workers in Each Category								
	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	-	-
41-45	9	5	15	6	1	1	-	-	-
46-50	7	4	6	3	1	-	-	1	-
51-55	5	3	7	1	-	-	-	-	-
56-60	2	1	3	1	-	-	-	-	-
61-70	3	4	4	-	-	-	-	-	-
71-90	3	5	-	-	-	-	-	-	-
91-110	3	3	-	-	-	-	-	-	-
111-155	45	2	-	-	-	-	-	-	-
Total	94	136	229	145	37	37	33	7	4

* Various annual reports reported the units in mg L^{-1}, which is an obvious typographical error. The permissible urine concentration, averaged throughout the year, was 0.05 mg L^{-1}.

5.3.7 *In Vivo* Analysis

Lung counting became available to FEMP in 1968 in the form of the Mobile In Vivo Radiation Monitoring Laboratory. 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 the AEC listed the number of workers

who exceeded 50% of the MPLB and the calculated annual doses to the lung in rem. The uranium MPLB used was 0.0175 µCi, assuming 1% enrichment. Table 5-24 presents the typical reporting of internal dose as determined by direct lung counting.

Industrial Hygiene & Radiation Department Internal Deposition Action Levels procedures from about 1970 indicate actions related to the determination of percent maximum permissible lung burden 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 Th-234 progeny assumed to be in equilibrium with the U-238. Thorium-232 and Th-228 activities were determined based on equilibrium assumptions and detection of their progeny, most likely Ac-228 for Th-232, but Pb-212 may have been used for assessment of both Th isotopes. Thorium-230 is not readily detectable by *in vivo* measurements. There appeared to be no attempt to detect TRU contaminants with the MIVRML. In fact, the only determination made with the mobile van was a quantification of the uranium lung burden in micrograms of uranium, with the assumption of 1% enrichment and of occasional thorium lung burdens as indicated by some claim records.

Table 5-24. Typical reporting of internal dose determined by direct lung count.

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

The results from the MIVRML were calibrated in µCi of ^{235}U and reported in mg of uranium in the lung, which was translated to MPLB based on the assumed enrichment (generally 1%). The percent body burden was then multiplied by 15 rem to obtain the assigned annual dose in rem.

The 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 upon their job exposure potentials, as shown in Table 5-25:

Table 5-25. Typical MIVRML counting schedule at Fernald in the 1970s.

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 classifications were scheduled to be counted, and each worker would be counted at least once during a 2-yr period.
Mechanical Department laborer	
Laundry group	
Industrial truck operator	
Locomotive operator	
Switchman	
Graphite shop machinist	
Machine tool operator	
Degreaser	
Crane operator	
Stamper	

Labor Category Description	*In vivo* Counting Schedule
Plant 6 laborer	
Furnace operator heater	
Mill man	
Decontaminator	
Transportation laborer	
Cafeteria	Salaried personnel and workers in these classifications were not routinely counted because of low chronic exposure and low potential for unobserved acute exposures.
Water treatment group	
Power plant group	
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	

5.3.8 *In Vivo* MDA

The In Vivo Examination Center (IVEC) operated at the FCP from 1989 to 2001, a subject with a 2.5 cm chest wall thickness had the MDAs listed in Table 5-26 at the 95% confidence interval for a 3,600 sec 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 5-26 lists the MDAs for FEMP *in vivo* analyses.

Table 5-26. Uranium *in vivo* MDAs for the lung.

Radionuclide	Facility	MDA (nCi)	Depleted U MDA (mg)	Natural U MDA (mg)	2% enriched MDA (mg)
U-235	MIVRML 1968 to 1989				*100µg(20%)
U-238					*6.5 mg(1%)
Th-232		6 mg thorium			
U-238(Th-234)	FCP IVEC 1989 to 2001	2.5	7.4	7.5	7.6
U-235		0.18	36.0	11.3	4.2
Pu-239		190			
Am-241		0.25			
Th-232 (Ac-228)**		1.2			
Th-232(Pb-212)		0.3			
Ra-226		≥ 3.0			

* The recorded MDA reported in Scott et. al. (1969, p. 169). Values reported in a claimant record were below 10% MPLB, which implies an MDA of less than 1 nCi total 1% (assumed) U-235.

** The degree of equilibrium is seldom known in a plant setting because these daughter products constitute a complex decay chain and the first controlling daughter has such a long half life (5.75 yr). As a consequence, *in vivo* measurements are used only as a screening technique.

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Selected examples of FEMP dosimetry data records in claim files.

GLOSSARY

Class

The respiratory tract classification scheme in ICRP 30 for inhaled material according to its rate of clearance from the pulmonary region of the lung.. Materials are classified as D (days), W (weeks), or Y (years), according to how fast they clear the lungs; the half-times are class D in less than 10 days; class W in 10 to 100 days; class Y in more than 100 days. Recent recommendations in ICRP Report 66 modified the lung model and now instead of class D, W, and Y with lung absorption Types F (fast), M (moderate), and S (slow).

Becquerel

A unit of radioactivity equal to one disintegration per second

curie

A special unit of activity. One curie exactly equals 3.7×10^{10} nuclear transitions per second.

Depleted uranium

Uranium nuclide that has undergone a process to remove the ^{235}U isotope, resulting in a relative increase in the percentage of the ^{238}U isotope

Dose equivalent (H)

The product of the absorbed dose (D), the quality factor (Q), and any other modifying factors. The dose unit is rem.

Dose

A specific amount of energy from ionizing radiation absorbed per unit of mass.

Dosimetry

The science of assessing absorbed dose, dose equivalent, effective dose equivalent, etc., from external or internal sources of radiation.

Enriched uranium

Uranium which has been processed to contain a higher abundance of the isotope ^{235}U.

exposure

As used for external dosimtry, exposure refers to a measure expressed in roentgens (R) of the ionization produced by photon radiation (i.e., gamma rays) in air.

In vitro

In glass. Outside the living body and in an artificial environment. Typically used for bioassay of a contaminant in excreta, such as in fecal or urine samples.

In vivo

In the living; In the living body of a plant or animal. Bioassay counting analysis of radionuclides in the human body.

Isotope

Nuclides having the same number of protons in the nuclei (same atomic number), but having a differing number of neutrons (different mass number).

maximum permissible lung burden (MPLB)
During the 1970s the occupational limit for radionuclides in the lung was expressed in terms of a quantity of the isotope that could be present at any given time to deliver 15 rem per year to the lung at the end of a 50-y period of chronic exposure.

Maximum permissible body burden (MPBB)
Same as MPLB except that any internal organ – or combination of a number of organs could be the subject of the determination.

Millirem
A unit of radiation dose equal to one-thousandth of a rem (see rem).

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

Natural uranium
Uranium occurring in the natural state that has not been through a ^{235}U enrichment process.

Rad
A unit of absorbed dose equal to 100 ergs/gm of any material.

Radiation
Energy transferred through air or some other media in the form of particles or waves (see ionizing radiation)

Radionuclide
A radioactive species of an atom characterized by the constitution of it nucleus specified by the number of protons, neutrons, atomic number, and mass number.

rem
A unit of dose equivalent, equal to the product of the rad absorbed dose and the quality factor.

Type
Refers to the rate of material absorption from the lung to the blood and includes types F (fast), M (moderate), and S (slow).

Transuranic (TRU) materials
Radioisotopes of nuclides having an atomic number greater than 92.