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ACRONYMS AND ABBREVIATIONS

ADU	ammonium diuranate
AEC	U.S. Atomic Energy Commission
AP	anterior-posterior
AWE	atomic weapons employer
CFR	Code of Federal Regulations
Ci	curie
cm	centimeter
d	day
DCF	dose conversion factor
DL	decision level
DOE	U.S. Department of Energy
dpm	disintegrations per minute
DU	depleted uranium
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
EU	enriched uranium
F	fast (absorption type)
g	gram
gal	gallon
GM	geometric mean
GSD	geometric standard deviation
HASL	Health and Safety Laboratory
HEU	highly enriched uranium
hr	hour
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in.	inch
IREP	Interactive RadioEpidemiological Program
keV	kiloelectron-volt, 1,000 electron-volts
kg	kilogram
L	liter
lb	pound
LEU	low-enriched uranium
LOD	limit of detection
m	meter
M	moderate absorption type
MDA	minimum detectable amount or activity
MDL	minimum detectable level
MeV	megaelectron-volt, 1 million electron-volts
mg	milligram
mL	milliliter
MOX	mixed oxide

MPC	maximum permissible concentration
mR	milliroentgen
mrem	millirem
nCi	nanocurie
NFS	Nuclear Fuel Services
NIOSH	National Institute for Occupational Safety and Health
NRC	U.S. Nuclear Regulatory Commission
ORAU	Oak Ridge Associated Universities
PA	posterior-anterior
pCi	picocurie
PFG	photofluorography
POC	probability of causation
R	roentgen
RU	recycled uranium
s	second
S	slow (absorption type)
SEC	special exposure cohort
SEFOR	South-West Experimental Fast Oxide Reactor
SRDB Ref ID	Site Research Database Reference Identification (number)
TLD	thermoluminescent dosimeter
U.S.C.	United States Code
yr	year
µCi	microcurie
µg	microgram
§	section or sections

1.0 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer [AWE] facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [EEOICPA; 42 U.S.C. § 7384l(5) and (12)]. EEOICPA, as amended, provides for employees who worked at an AWE facility during the contract period and/or during the residual period.

Under EEOICPA, employment at an AWE facility is categorized as either (1) during the DOE contract period (i.e., when the AWE was processing or producing material that emitted radiation and was used in the production of an atomic weapon), or (2) during the residual contamination period (i.e., periods that NIOSH has determined there is the potential for significant residual contamination after the period in which weapons-related production occurred). For contract period employment, all occupationally derived radiation exposures at covered facilities must be included in dose reconstructions. This includes radiation exposure related to the Naval Nuclear Propulsion Program and any radiation exposure received from the production of commercial radioactive products that were concurrently manufactured by the AWE facility during the covered period. NIOSH does not consider the following exposures to be occupationally derived (NIOSH 2007a):

- Background radiation, including radiation from naturally occurring radon present in conventional structures

- Radiation from X-rays received in the diagnosis of injuries or illnesses or for therapeutic reasons

For employment during the residual contamination period, only the radiation exposures defined in 42 U.S.C. § 7384n(c)(4) [i.e., radiation doses received from DOE-related work] must be included in dose reconstructions. Doses from medical X-rays are not reconstructed during the residual contamination period (NIOSH 2007a). It should be noted that under subparagraph A of 42 U.S.C. § 7384n(c)(4), radiation associated with the Naval Nuclear Propulsion Program is specifically excluded from the employee's radiation dose. This exclusion only applies to those AWE employees who worked during the residual contamination period. Also, under subparagraph B of 42 U.S.C. § 7384n(c)(4), radiation from a source not covered by subparagraph A that is not distinguishable through reliable documentation from radiation that is covered by subparagraph A is considered part of the employee's radiation dose. This site profile covers only exposures resulting from nuclear weapons-related work. Exposures resulting from non-weapons-related work, if applicable, will be covered elsewhere.

A decision has been made that internal dose from thorium cannot be reconstructed with sufficient accuracy for W.R. Grace employees during the period from 1958 through 1970. Therefore only internal dose from uranium and plutonium is considered in Section 3.0 of this site profile. The Special Exposure Cohort (SEC) evaluation report (NIOSH 2007b) defines a single class of employees for which NIOSH cannot estimate radiation doses with sufficient accuracy. This class includes all AWE employees who were monitored or should have been monitored for potential exposure to thorium while working at the W.R. Grace site at Erwin, Tennessee, for a number of workdays aggregating at

least 250 workdays from January 1, 1958, through December 31, 1970, or in combination with workdays within the parameters established for one or more other classes of employees in the SEC.

This site profile provides specific information on documentation of historical practices at the W.R. Grace and Company plant. If the dose reconstructor cannot determine whether the work was U.S. Atomic Energy Commission (AEC) weapons-related work or non-AEC-related work, the dose reconstructor should assume that all work was AEC weapons related to ensure favorability to claimants.

1.1 PURPOSE

This site profile provides technical basis information to be used to evaluate the total occupational radiation dose that can reasonably be associated with a worker's radiation exposure at the W.R. Grace and Company plant. This dose results from exposure to external and internal radiation sources in W.R. Grace facilities and to occupationally required diagnostic X-ray examinations. This site profile includes methods for estimating doses that could have occurred while an employee was not monitored or inadequately monitored, or that were missed due to analytical detection limits or incomplete or missing monitoring records (i.e., missed dose).

Dose reconstructors use the NIOSH Interactive RadioEpidemiological Program (IREP) and the Integrated Modules for Bioassay Analysis (IMBA) computer programs to evaluate radiation doses.

Information on measurement uncertainties is an integral component of the NIOSH approach to dose reconstruction. This site profile describes how to evaluate uncertainty in relation to W.R. Grace exposure and dosimetry records.

1.2 SCOPE

Sections 2.1 and 2.2 detail major W.R. Grace facilities and operations. W.R. Grace began processing materials in the late 1950s at its Erwin, Tennessee, site. The principal operation at the site has been to convert highly enriched uranium (HEU) and low-enriched uranium (LEU) from UF_6 or to recover uranium from scrap to a product that meets customer requirements. Thorium, depleted uranium (DU), ^{233}U, recycled uranium (RU), and plutonium have also been processed at various times to oxides or metals with subsequent processing into the form necessary for the manufacture of nuclear fuel. In addition, the recovery of HEU from uranium-aluminum alloy occurred in the storage, chemical, and ceramic buildings with sampling being conducted in the metallurgical chemistry and spectrographic laboratories (AEC 1959, p.6).

Section 3.0, for the estimation of internal exposure, describes the internal dosimetry program at W.R. Grace. Workers handled a variety of radionuclides as part of their routine tasks. The key elements in the source term are uranium, plutonium, and thorium.

During the AEC contract years from the late 1950s though 1970, the monitoring methodology for W.R. Grace employees for intakes of radionuclides was bioassay, with the primary focus being uranium urinalysis. The UO_2/PuO_2 mixed-oxide (MOX) facility is believed to have been completed in 1965, and plutonium processing began in 1966, when plutonium was analyzed by urine bioassay. Few feces samples in the 1960s were collected for uranium and plutonium. Air monitoring in workplaces and in the breathing zones of employees has been a common surveillance method. Although W.R. Grace apparently did not use the data acquired from the air monitoring program to prepare the dose of record for employees; these data have been used in this site profile in certain instances for individual exposure evaluations. *In vivo* monitoring began in 1970, with capabilities that focused on uranium monitoring, which expanded to plutonium and thorium monitoring in the 1990s. There was not a substantial amount of *in vivo* monitoring in the AEC contract years.

Section 4.0, for the estimation of external exposure, describes the external dosimetry program at W.R. Grace, and includes electron, photon, neutron, and occupational medical X-ray dose. W.R. Grace utilized Nuclear Chicago and then Landauer for its external dosimetry program. The program included whole-body, wrist, and extremity film badges for photon and beta monitoring. Neutron monitoring was not conducted at W.R. Grace during the AEC contract years. Thermoluminescent dosimeters (TLDs) were provided by Landauer in 1989.

Section 5.0, for the estimation of exposure to residual activity, discusses external and internal dose estimates from surface contamination. The exposures could have occurred from resuspension of removable contamination from AEC contract year operations and ongoing operations. Section 6.0 is the environmental section.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 7.0.

2.0 SITE DESCRIPTION, HISTORY, AND PROCESS

The Davison Chemical Company, a division of W.R. Grace, began processing radioactive materials in the late 1950s at the site of the current Nuclear Fuel Services (NFS) facility near Erwin, Tennessee (NFS 2005).

The *Report to Joint Committee on Atomic Energy Congress of the United Stated by the United States General Accounting Office* (GAO 1967, p.5-6) from September 14, 1967, states:

> *AEC authorized the Davison Chemical Division of W.R. Grace & Company (Grace) to receive and process special nuclear material under license number SNM-124. Grace received its first material as an AEC licensee by lease agreement in March 1958 and its first nuclear material as an AEC contractor in May 1959.*

In 1964, NFS was formed by the merger of W.R. Grace and the American Machine and Foundry Company. In 1965, the UO_2/PuO_2 MOX facility was completed, and in 1968, the light-water breeder reactor thorium, thoria, and ^{233}U oxide program was initiated (NFS 2005). Ownership of NFS transitioned from Getty Oil to Texaco as part of a stock buyout of Getty Oil in 1969. In 1987, NFS Services, a private Atlanta-based limited partnership purchased NFS from Texaco. Figure 2-1 shows the location of Erwin, Tennessee. Figure 2-2 shows the principal site infrastructure for the NFS facility as of 1986 (Congress 1986, p.108).

Figure 2-1. Location of Erwin, Tennessee.

Regulatory authority over operations was originally under the AEC (1954 to 1974) and transitioned to the U.S. Nuclear Regulatory Commission (NRC) (1975 to present) under Special Nuclear Material License SNM-124, as amended.

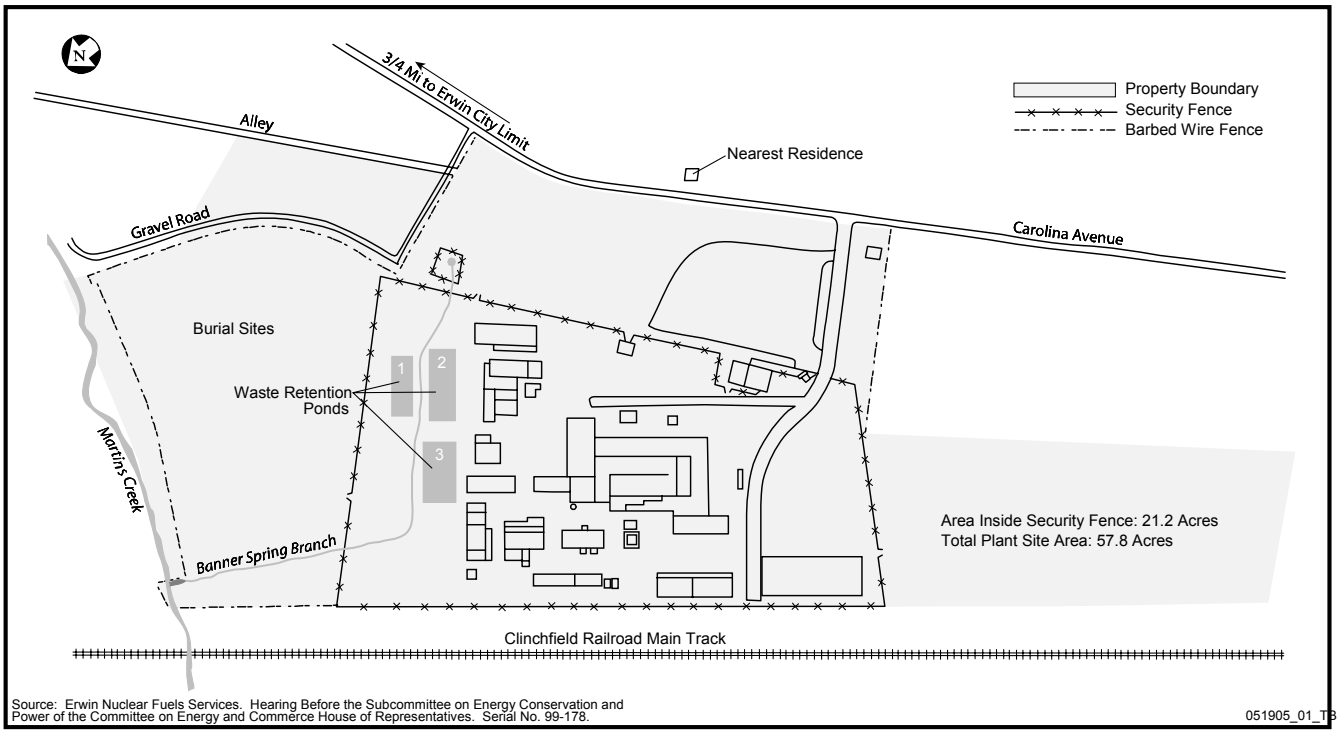

Figure 2-2. Plant site at W.R. Grace/NFS.

The principal operation at the site has been to convert HEU and LEU from UF_6 to a product that meets customer requirements. Thorium, DU, ^{233}U, and plutonium have also been processed at various times to oxides or metals with subsequent processing into the form necessary for the manufacture of nuclear fuel. Specific tasks at the facility included (1)processing ThO_2 that was mixed with ^{233}U to make the light-water breeder reactor fuel for the Shippingport Reactor and (2) fabricating plutonium and DU MOX fuel for the South-West Experimental Fast Oxide Reactor (SEFOR) in Building 234 with laboratory capabilities in Building 110; both operations ceased in 1970 (Congress 1986, p.31). Other operations were associated with the conversion operations such as scrap recovery operations (uranium and other nuclear fuel material) and cleaning and certification of empty cylinders for transport of LEU UF_6. Table 2-1 lists the approximate processing history of the site.

2.1 PROCESS DESCRIPTION

Brief summaries for the principal operations are given below, and additional details can be found in the series of Feasibility Reports in the cited references. Inherent in all of the operations is nuclear criticality safety that governs not only the operations and storage but also the movement of material within the facility. Nuclear criticality safety is maintained at the facility through the control of one or more of the conventional parameters of geometry, mass, concentration, and control of neutron interaction between subcritical units. The standard administrative policy at this and other similar facilities is to control two such parameters whenever possible. For birdcage units, the two control parameters are geometry (birdcage dimensions) and a ^{235}U mass limit that can be placed within the birdcage. Examples of birdcages that were used include a ^{233}U storage birdcage that used a shielded 55-gal drum that contained inserts with spacers; UF_6 cylinders storage birdcage that used a 2-in. × 2-in. frame made of metal that held the 5-in. UF_6 cylinder in place at its center with a total dimension of 52 in. high and 30 in. wide; and a U_3O_8 storage birdcage that used a 2- × 2-in. metal frame made of metal that held a 10.75-in. outside diameter × 12-in.-high cylinder in the center with a total dimension of 36 in. high and 30.625 in. wide (W.R. Grace 1959, p.23, Runion 1959, p.5, Glauberman 1962). A

review of the available literature shows that no criticality accidents have occurred during W.R. Grace and later NFS operations.

Table 2-1. W.R. Grace and NFS AEC weapons-related work period and locations.

Operations	Period of AEC weapons-related work[a]	Building location
Thorium (metal and oxide)	1/1/1958 through 12/31/1970	110C, 110D, 111, 130, 234B, 234C, 310
Uranium metal (HEU and DU)	1/1/1958 through 12/31/1970	110, 110E, 111, 130, 135, 234, 301
LEU UO_2	1/1/960 through 12/31/1970	301, 110E, 111, 130, 135, 301
U-233 fuel (sometimes mixed with ThO_2)	01/01/1961 through 12/31/1970	234B, 234C, 110C, 110D
Plutonium fuel[b] and MOX	1/1/1966 through 12/31/1970	234A, 234B, 234C, 110C, 110D
HEU scrap recovery	1/1/1958 through 12/31/1970[a]	130, 220, 230, 233
HEU fuel	1/1/1966 through 12/31/1970	100, 105, 120, 131, 132/133, 220, 233, 300, 302, 303, 304, 310, 330, 301
LEU scrap recovery	1/1/1960 through 12/31/1970 (AEC 1961), or (Congress 1986)	111, 220, 230, 233
LEU cylinder washing	1/1/1958 through 12/31/1970	111,130

Sources: AEC (1961), Congress (1986, pp. 107-114 and 127-143).

a. The end of 1970 is when AEC-weapons-related operations ended.

b. A review of the claimant files (to date) indicates that there are plutonium bioassay records that begin in 1967. Because the plutonium facility was completed in 1965, it is reasonable to assume that plutonium operations occurred from 1966 to 1970 (NFS 2005, p.2; Congress 1986, p.107).

2.1.1 Production of Uranium Metal and Uranium Metal Alloys Enriched up to 12% Uranium-235

The UF_6 was supplied in approved standard cylinders and received in approved packaging such as birdcages. The cylinders were check-weighed and placed into storage in special concrete cells or birdcages in one of the warehouse buildings.

2.1.1.1 Conversion of Uranium Hexafluoride to Uranium Metal

The overall process involves vaporization, reduction to UF_4, reduction to uranium metal, pickling, processing into other metal products, packaging, and shipment. The following information is from *Feasibility Report for the Production of Uranium Metal and Uranium Metal Alloys Enriched Up To 12% ^{235}U* (Housholder 1963a, p.6-8).

UF_6 was received in solid cylinders and had to be vaporized to transfer it to the UF_6-to-UF_4 reactor (the 6-to-4 unit). Vaporization was accomplished by heating the UF_6 cylinders in an electric oven. Up to six cylinders could be placed into the oven for processing.

The UF_6 was piped into the 6-to-4 unit where it was reduced with hydrogen. The solid UF_4 powder dropped into a product hopper where it was metered into safe-diameter product cans. The offgas flowed through two cyclone separators where any entrained UF_4 dust was collected in additional product cans. The offgas was then filtered to remove the last traces of UF_4 dust and was scrubbed with potassium hydroxide to remove hydrogen fluoride vapors. The gas, free of uranium and acid, was vented to the atmosphere through a flame arrester where excess hydrogen was burned off.

2.1.1.2 Reduction of Uranium Tetrafluoride to Uranium Metal

The UF_4 was weighed into a reduction batch and blended with a reducing agent such as magnesium metal. The charge was then heated under vacuum in an induction furnace to form a uranium metal derby. After cooling, the derby was broken out and separated from the slag. The slag was packaged for scrap recovery where any remaining uranium in the slag was recovered.

2.1.1.3 Pickling

The uranium derby was pickled in acid to remove adherent slag and scale. The pickle solution was sent to scrap recovery for recovery of uranium. The pickled derby was then sampled for impurities.

2.1.1.4 Other Metal Products

As necessary, the derbies were broken into smaller pieces on a large hydraulic press before shipment. On other occasions, the derbies were remelted and cast into various shapes such as slugs, rods, and plates. The uranium could also be alloyed with other metals during remelting and casting. Melting occurred in a large vacuum induction furnace. Cast pieces could be pickled as previously described.

2.1.1.5 Packaging and Shipping

All enriched uranium (EU) metal products were packaged for shipment in approved birdcages. The products could be stored on the site for a time before shipment to the customer.

2.1.2 Scrap Recovery Operations

This operation was designed to recover the scrap uranium from the manufacturing processes. The process steps were assumed to be carried out in a similar manner for scrap with either HEU or LEU (Katine 1960, p.18). The exception between handling the different enrichment levels would be in the dissolution and filtration steps. There was potential for different end processes that were adjusted based either on the customer's desired final material form (for scrap recovery from outside sources) or on how the facility was going to reinsert the recovered uranium in the fuel fabrication process.

In one particular job, dependent on whether the material was greater or less than 2% enrichment, two different batch sizes and dissolver tanks were used. For scrap material equal to less than 2% enrichment, a maximum batch of 87.95 kg uranium (220 lb) of UO_2 is dissolved in a 500-gal capacity dissolver tank. For all other enrichments, a 20-gal dissolver tank was employed and the batch would contain a maximum of 400 g of ^{235}U. This process would result in two types of solutions. There would be solutions of 2% or less enrichment at a concentration of less than 2 grams of ^{235}U per liter. There were solutions ranging from 2.8 to 9.9% enrichment at a concentration of 5 grams of ^{235}U per liter. From these two types of solutions, a third solution will be formed that will not exceed 3% enrichment, and will contain a maximum of 88 pounds of uranium (39.9 kilograms) which is the safe mass for 3% enrichment.

Of note for this process, the memorandum that was attached to a Feasibility Report for HEU scrap recovery specifically states, "the chopping and sawing of solid metal plates, billets, and rods, and the operations of grinding, screening, and blending of the resultant oxides from the calcinations step, could be possible dust sources. Air samples should be obtained at these operations and evaluated at the start of operations to ascertain the effectiveness of the existing ventilation" (Glauberman 1962, p.117). A summary of the HEU scrap recovery follows (Housholder and Runion 1962, p.122-123).

2.1.3 Highly Enriched Uranium from Scrap Recovery

2.1.3.1 Receiving and Storage

All scrap was received in approved shipping containers and birdcages and sent to storage warehouses to await processing.

2.1.3.2 Sampling

Before processing, each container was inspected to decide what head-end steps could be necessary before the scrap could be dissolved. In addition, at this time samples were taken for uranium assay to determine batch sizes.

2.1.3.3 Head-End Operations

Due to the many types of scrap the facility received, it was often necessary to pretreat the scrap before dissolution and subsequent extraction. Metal chips and turnings, for instance, were routinely received stored under oil, which had to be removed before dissolution. This was done by draining the oil off the metal in wire baskets. The metal was then washed with solvents and dried.

Solid metal plates and rods were chopped or sawed into smaller pieces to facilitate handling and weighing into batches. Combustible wastes such as filters, sponges, grinder sludge, sweepings, etc., were calcined to reduce their bulk and to remove hydrogenous and carbonaceous materials, such as water and oil. After calcining, the resultant oxides were ground and screened; material not passing the screen was recycled to the grinder. The oxides were then blended to ensure homogeneity for sampling for uranium assay, from which accountability and batch sizes could be determined.

Oxide pellets were ground and screened and could be calcined and blended to facilitate dissolution.

2.1.3.4 Final Process Steps

A review of the Feasibility Reports showed there were different final process steps that appear to have depended on the final material form needed either to be reinserted into the facility process or to be shipped back to the customer. The extracted uranium liquid (also known as O.K. Liquor) could be converted into a solid through either a boildown and crystallization process (Housholder and Runion 1962, p.124) or by precipitation and filtration followed by calcination and blending (AEC 1959, p.7-9). A summary of both final process steps follows:

- Boildown and Crystallization: The pure uranium solution could be boiled down and evaporated to produce uranyl salts, which were packaged for shipment. At times, the concentrated solution from the boildown step was packaged in bottles for shipment as a solution.

- Precipitation/Filtration and Calcination/Blending: The extracted uranium liquid was precipitated by batch process with ammonium hydroxide in an approved container with an agitator. The precipitated slurry was transferred to the filtration hood for filtration with a large Buchner funnel. The filter cake was loaded into metal trays and transferred to a hooded muffle furnace where it was calcined to U_3O_8. Once cooled, the material was transferred to a blending station for introduction into a ball mill for grinding and blending. After ball milling, the U_3O_8 was sampled, packaged, weighed, and returned to the storage area ready for shipment.

2.1.4 Production of Uranium Oxide Mixed with Thorium Oxide and Zirconium Oxide

This line of production ($^{233}UO_2/ThO_2$ and $^{233}UO_2/ZrO_2$) was in operation from approximately 1961 to 1969 for the light-water breeder reactor fuel of the Shippingport Reactor. This process was more involved than other process lines with 11 separate steps to produce the ^{233}U fuel (Housholder 1963b, p.35-37). A brief summary of the process follows.

2.1.4.1 Receiving and Storage

Uranium-233 was received as a uranyl nitrate solution in an approved shipping cask. A typical receipt shipment was approximately 7 kg of ^{233}U in storage columns awaiting processing.

2.1.4.2 Solution Concentration

The received uranyl nitrate solution had a concentration of approximately 150 g/L ^{233}U. Before precipitation, a higher concentration was required and was obtained through evaporation.

2.1.4.3 Precipitation

The uranyl nitrate solution was measured into safe batches and the uranium was precipitated by the addition of a precipitating agent such as NH_4OH.

2.1.4.4 Drying and Grinding

The resultant precipitate was dried at a low temperature and ground to a fine powder.

2.1.4.5 Calcination to Uranium Dioxide

The dried and ground uranium precipitate was calcined to UO_2 in a continuous muffle furnace under a hydrogen atmosphere. The resultant oxide was stored in birdcages to await blending.

2.1.4.6 Blending

To ensure homogeneity of the oxide, the precipitation batches were accumulated and blended before addition of the diluents (ThO_2 or ZrO_2). After a homogenous UO_2 blend was obtained, it was sorted in birdcages to await blending with the diluents.

2.1.4.7 Diluent Addition and Blending

Safe batches of UO_2 and diluents were weighed out and blended together in a twin-shell blender. To ensure thorough mixing, each batch was also ball-milled.

2.1.4.8 Binder Addition

A binder was mixed with the oxide blend, and the wet mixture was granulated and dried. After drying, the granules were broken up by screening.

2.1.4.9 Lubricant Addition and Pressing

To improve pressability, a die lubricant was added to the granules and blended in. The oxides were then compacted into small pellets on a 40-ton press.

2.1.4.10 Binder Removal and Sintering

The resultant pellets were loaded into trays and heated in an oven to drive off the binder. They were then sintered in a continuous muffle furnace under a hydrogen atmosphere.

2.1.4.11 Physical Measurement and Grinding

The sintered pellets were inspected and measured to see that they met customer requirements. Before packaging, the sintered pellets could have required grinding to the correct diameter. If grinding was necessary, the pellets were cleaned by washing them in water, drying them, and remeasuring them.

2.1.4.12 Tube Loading

The finished pellets were stacked, weighed, and loaded into Zircaloy tubes. The tubes were welded closed and loaded into shielded 55-gal-drum birdcages for temporary storage and eventual shipment to the customer.

2.1.5 Production of Mixed Oxide Fuel

Documentation of the process steps for the production of MOX fuel (combination of PuO_2 with LEU UO_2) for SEFOR at the facility was not available. However, other DOE sites have performed similar operations. Production of MOX fuel would have followed similar process steps for the production of $^{233}UO_2/ThO_2$ and $^{233}UO_2/ZrO_2$ fuel from the blending step onto tube loading. The process steps were completely within shielded and filtered gloveboxes for worker health and safety. It is assumed that the plutonium sent to the facility was in approved shipping packages as PuO_2 powder in sealed cans. The final product would have had approximately 5% ^{239}Pu in fuel pin assemblies loaded into shielded 55-gal-drum birdcages for temporary storage and shipment to the customer.

2.1.6 Uranium Hexafluoride Cylinder Washing

UF_6 cylinder washing was performed in the Building 200 complex to recover uranium in a ventilated glovebox using water or steam. The removed wash solution was transferred to the HEU scrap recovery process to recover the uranium.

3.0 ESTIMATION OF INTERNAL EXPOSURE

The primary sources of internal radiation exposure at W.R. Grace were uranium and thorium dust produced from the manipulation and chemical processing of those materials during recovery and fuel fabrication processes. Beginning in approximately 1966, there was also the potential for internal dose from plutonium-containing dust.

Uranium enrichment levels included DU, natural uranium (NU), LEU (3.5%), and HEU (93%) as well as ^{232}U and ^{233}U. Uranium-233 contains ^{232}U as an impurity due to nuclear reactions from the neutron irradiation of ^{232}Th. The ^{232}U impurity results in elevated gamma and beta dose rates due to the ingrowth of ^{232}U progeny (^{228}Th and its progeny). Uranium from recycling operations would have included small activities of non-uranium isotopes such as ^{99}Tc, ^{237}Np, ^{232}Th, ^{106}Ru, and ^{238}Pu. There was one indication of RU processing in the case of a recycled ^{233}U pellet (Housholder 1963b).

Table 3-1 lists the various enrichments and chemical forms of the processed radionuclides.

3.1 URANIUM EXPOSURES

The chemical form and the uranium enrichment varied over time at W.R. Grace. The manufacture of uranium occurred in most of the buildings at W.R. Grace, with the exception of Buildings 110 and 234 where plutonium was primarily processed.

In most cases, it will not be known how much of a specific uranium enrichment versus another, to which a worker could have been exposed. Due to the presence of many forms of uranium, the most favorable to claimant of the absorption types (F, M, and S) should be assumed (ORAUT 2007a).

Table 3-1. Fuel types, chemical forms, isotopes, and enrichments of W.R. Grace process material.

Radionuclide or fuel	Chemical form and solubility type(s)	Isotope	Enrichment
Uranium	UF_6, UO_2F_2, & $UO_2(NO_3)_2$ (F) UO_3 & UF_4 (M) U_3O_8 & UO_2, (S)	U-233 U-234 U-235 U-236 U-238	DU, NU, LEU (3.5%), HEU (93%)
Plutonium	PuO_2 (S)	Pu-238 0.8%, Pu-239 2.6%, Pu-240 1.33%, Pu-241 93.4%, Am-241 1.9%, Pu-242 0.0002% (% activity)	Fuel grade aged 10 yr (12% Pu-240)
Technetium or other transuranic elements	Same as the U or Pu matrix	Tc-99, Np-237	NA
MOX; Pu U-235 fuel	PuO_2 /UO_2 (S)	20% PuO_2 and 80% UO_2 by weight (Sharma et al. 2002, p.40)	About 3.5% U-235
MOX; Th U-233 fuel[a]	ThO_2 /$^{233}UO_2$, ZrO_2 /$^{233}UO_2$ and ZrO_2 /$^{235}UO_2$ (S)	1% UO_2 and 99% ThO_2 or ZrO_2 by weight (Housholder and Runion 1962, p.189)	HEU (likely >20%)

a. See Table 3-3 for ^{233}U fuel composition.

ICRP (1995) lists UF_6, UO_2F_2, and $UO_2(NO_3)_2$ (uranyl nitrate) as type F; UF_4 and UO_3 as type M; and U_3O_8 and UO_2 as type S. Table 3-2 identifies uranium source term information. .

Table 3-2. Uranium source term information.

Uranium source term	Reference	Specific activity (pCi/µg)	Activity fractions			
			U-234	U-235	U-236	U-238
NU	IMBA	0.683	0.489	0.023	-	0.489
93.%	IMBA	68.1	0.968	0.030	0.002	0.0003
3.5%	IMBA	2.20	0.818	0.034	-	0.147
2%	HPS[a]	1.616	0.648	0.041	0.0009	0.311
Typical DU	IMBA	0.402	0.155	0.011	0.0005	0.834
			Specific constituent activity in mixture (µCi/g, nCi/mg, or pCi/µg)			
NU	IMBA	0.683	0.334	0.016	-	0.334
93.%	IMBA	68.1	65.9	2.04	0.136	0.020
3.5%	IMBA	2.20	1.80	0.075	-	0.323
2%	HPS[a]	1.616	0.778	0.049	0.001	0.373
Typical DU	IMBA	0.402	0.062	0.004	0.0002	0.335

a. American National Standards Institute Standard N13.22 (HPS 1995).

The composition of the Shippingport Reactor fuel, total core is shown in Table 3-3. (Olson, McCardell, and Illum 2002, p.37). For dose reconstruction, the composition of the ^{233}U fuel mixture in Table 3-3

should be used for assessing uranium intakes beginning on January 1,1961 (Congress 1986, p. 107) because it is more favorable to claimants.

Table 3-3. Uranium-233 Shippingport Reactor fuel activity fractions, total core).

	G	Bq/g	Total activity	% activity	% weight
U-232	3.7795	7.93E+11	3.00E+12	1.56172%	0%
U-233	500600.60	3.57E+08	1.79E+14	93.18272%	2%
U-234	6547.90	2.31E+08	1.51E+12	0.78607%	0%
U-235	440.90	8.01E+04	3.53E+07	0.00002%	0%
U-236	112.80	2.40E+06	2.70E+08	0.00014%	0%
U-238	1900.60	4.47E+09	8.49E+12	4.41967%	0%
Th-232	23481000.00	4.06E+03	9.54E+10	0.04966%	98%
Total	23990606.58		1.92E+14		100

3.1.1 Uranium Air Sampling

This discussion focuses on documented air-sampling data from two separate Health and Safety Laboratory (HASL) reports by the AEC. The first air sampling was performed at W.R. Grace during the recovery of 93% HEU from uranium-aluminum alloy scrap in the storage, ceramics, and chemical buildings (AEC 1959, p.5-6). The second air sampling was performed during the recovery of 3.6% LEU from slag scrap (AEC 1961). Tables 3-4 to 3-9 list the results of the sampling analyses.

Table 3-4. Occupational exposures for 93% uranium-aluminum alloy recovery.[a]

Operator	Number of persons	Average daily weighted exposures (dpm/m^3)
Column operator	6	31
Ceramic building operator	3	45
Charge makeup	3	14
Accountability	3	15
Chemist and technicians	8	3.1
Spectrographic operator and technician	3	2.5
Machine shop	5	12
Laundry	1	7
GM[b]		10.84
GSD[b]		2.76
95% confidence level value[b]		57.63
90% confidence level value[b]		39.84
50% confidence level value[b]		10.84

a. Data from AEC (1959, p.11).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

The air samples consisted of collection on filters of radioactive particulates from breathing zones and general areas during processing. The measured alpha activity on the filter was used to determine the airborne alpha activity concentrations. When multiple samples at a location were collected, AEC used the mean air concentration in subsequent calculations. AEC matched air concentration determinations with information about worker categories, locations, tasks, and times at each location or task.

Although it is unlikely that workers would have been exposed to the same air concentrations from the many other processes that were conducted at NFS, this is the only currently available air sample information. In addition, changes in the processes and the safety controls could have resulted in both increased and decreased exposure for any given period. In this document, an estimate of the intake was made by calculating the 95th percentile value of the maximum average breathing-zone concentration for the 1961 AEC air sample results (from Table 3-8 later in this section) by assuming a

lognormal distribution of the data in the AEC report (AEC 1961). Using Crystal Ball, a lognormal distribution calculation yielded a distribution with a geometric mean (GM) of 167 dpm/m^3,a geometric

Table 3-5. Average breathing-zone samples for 93% uranium-aluminum alloy recovery.[a]

Operation	Number of samples	Average concentration (dpm/m^3)
Shearing U-Al alloy for charge makeup and weighing	3	43
Charging digester with batch of U-Al alloy	2	170
Running material from digester through filter press and column	3	50
Cleaning residue from filter press	2	19
Securing O.K. Liquor from columns	1	65
Precipitation of O.K. Liquor	2	16
Filtering ppt. on Buchner funnel	2	15
Removing filter cake, placing in tray	1	1
Transferring tray from furnace to cooling area	2	65
Transferring tray from furnace to dry box, weighing and unloading tray in dry box, cleaning residue tray	3	280
Digestion of organic ashes in hood	1	1
Filtration of digested organic ashes	1	65
GM[b]		26.65
GSD[b]		5.84
95% confidence level value[b]		485.13
90% confidence level value[b]		255.57
50% confidence level value[b]		26.65

a. Data from (AEC 1959, p.12).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

Table 3-6. Average general air-sampling concentrations for 93% uranium-aluminum alloy recovery.[a]

Area	Number of samples	Average concentration dpm/m^3
Shearing and weighing	2	9
Solvent extraction area	6	19
Chemical building–oven area	3	23
Ceramics building	9	2
Machine shop	4	16
Wet chemistry lab	3	3
Spectrographic lab	3	2
Laundry	3	6
Lunch room	3	7
Clean locker room	3	2
Contaminated locker room	3	4
GM[b]		5.81
GSD[b]		2.52
95% confidence level value[b]		26.55
90% confidence level value[b]		18.98
50% confidence level value[b]		5.81

a. Data from AEC (1959, p.13).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

standard deviation (GSD) of 2.12, and a 95th-percentile value of 578 dpm/m^3. The calculated 95th-percentile air concentration was used to calculate upper estimates of internal exposures. The most conservative air concentrations were generated from the radiological task-oriented information in Table 3-8. If actual operator information from Tables 3-4 and 3-7 were to be used, the calculated intakes would be about a factor of 4 to 10 less. Task-oriented weighted average samples involve the

highest encountered air concentrations because of the closest proximity to the actual work and no accounting of the time away from the actual or lower exposure rate activities. The use of the task-oriented time-weighted average air concentrations is favorable to claimants.

Table 3-7. Occupational exposures for 3.6% uranium recovery from slag.[a]

Operator	Number of persons	Average daily weighted exposures (dpm/m^3)
Williams Roll Mill	3	170
Digestion	3	45
Solvent extraction	3	10
ADU precipitation	3	45
ADU oxide (ceramics)	3	17
Micropulverizer-drifter	3	71
Accountability-shipping and receiving	2	22
Laundry	1	9.4
Janitor	1	37
Health physics technician	2	20
GM[b]		30.72
GSD[b]		2.46
95% confidence level value[b]		132.11
90% confidence level value[b]		95.55
50% confidence level value[b]		30.22

a. Data from AEC (1961, p.33).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

Table 3-8. Average breathing-zone samples for 3.6% uranium recovery from slag.[a]

Operation	Number of samples	Average concentration (dpm/m^3)
Operating jaw crusher	4	230
Loading top of Williams roll mill with crushed slag	3	500
Removing drum of 325 mesh MgF_2 from hopper	2	70
Loading and unloading pot (oxidation furnace) with slag metal heavies	2	140
Charging digester with slag	3	150
Cleaning ADU from plate and frame filter press	3	130
Placing tray of ADU inside of oven	2	84
Removing tray of oxide from furnace-placing inside of glove box and transferring U_3O_8	4	97
Micropulverizing U_3O_8 (bag not working properly)	3	590
GM[b]		167.83
GSD[b]		2.12
95% confidence level value[b]		578.38
90% confidence level value[b]		440.08
50% confidence level value[b]		167.83

a. Data from AEC (1961, p. 34).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

As seen in Table 3-4, the uranium exposure by occupation differed with the column and ceramic building operators with the largest daily weighted exposures. This exposure variability between occupations is also seen in Table 3-7, where the Williams roll mill operators were clearly the highest exposed group, with the micropulverizer-drifter, digestion and ammonium diuranate (ADU) precipitation operators being the next highest groups.

The general air sample results in Tables 3-6 and 3-9 result in the lowest calculated intakes. This is to be expected because general air samples are typically lower than breathing-zone samples. Bioassay results are expected to be at levels between breathing-zone and general air sample results.

The breathing rate is based on the default for light work, 1.2 m^3/hr, as indicated in International Commission on Radiological Protection (ICRP) Publication 66 (ICRP 1994). This category assumes

Table 3-9. Average general air-sampling concentrations for 3.6% uranium recovery from slag.[a]

Area	Number of samples	Average concentration dpm/m^3
Crushing room	5	110
Digester area	6	11
Leach area	3	20
Solvent extraction area–all levels	8	6.4
Boil-down feed tank area	3	17
O.K. Liquor storage	3	9
Raffinate storage area	3	9
Neutralizer tank storage	4	10
ADU precipitation tanks	3	16
ADU filter area	4	17
U_3O_8 transfer hood area	3	11
Micropulverizer area	4	62
Accountability room	5	21
Chemistry lab	4	4.3
Spectrographic lab	2	9.3
Research and development lab	4	6.6
Laundry	4	2.3
Locker room–clean side	2	17
Locker room–dirty side	2	45
Lunch room	4	21
Health physics room	2	1.5
GM[b]		12.57
GSD[b]		2.7
95% confidence level value[b]		64.52
90% confidence level value[b]		44.95
50% confidence level value[b]		12.57

a. Data from AEC (1961, p.35).
b. Statistics calculated using a lognormal distribution and the Crystal Ball program.

an activity distribution of one-third sitting and two-thirds light exercise. The intakes in picocuries are calculated by dividing the 95th-percentile value of the air concentration (578 dpm/m^3) by 2.22 dpm/pCi and multiplying this result by the breathing rate and the assumed number of hours of exposure at a given concentration. The organ doses are assumed to be a constant distribution. Several assumptions in the intake and dose reconstruction are likely to be overestimating assumptions. This includes the use of a lognormal distribution, the 95% confidence level concentration, the task-related versus occupation time-weighted average, and the assumption of constant work activity and worker exposure during the entire work period.

Air sampling for ^{233}U has also been identified. Airborne concentrations for enriched UO_2 decladding and dissolution of $^{233}U_3O_8$ pellets from the immediate work area resulted in an average concentration of less than 1.8% of the maximum air concentration (1×10^{-10} µCi/ml) (Householder 1963b, p.51). This would result in calculations of intake less than that calculated above.

In the case where inhalation intakes are calculated from air concentrations, ingestion intakes are also to be considered. NIOSH (2004) indicates that the ingestion rate, in terms of picocuries for an 8-hour workday, can be estimated by multiplying the air concentrations in picocuries per cubic meter by a factor of 0.2. The uranium ingestion rate based on air concentration of 260 pCi/m^3 would be 52 pCi/workday. The daily inhalation and ingestion intake rate from LEU recovery is estimated from the 95% confidence level air concentration in Table 3-8. Table 3-10 lists the inhalation intake rate per year. Table 3-11 lists the ingestion intake rate by year.

Table 3-10. Estimated uranium inhalation intake rates based on measured time-weighted air concentrations during uranium recovery operations.

Work period	Number of years	Number of potential AEC work hours per work period	Air concentration (pCi/m^3)	Breathing rate (m^3/hr)	Intake (pCi)
1 yr	1	2,000	260	1.2	6.24×10^5

Table 3-11. Estimated uranium ingestion rates based on measured time-weighted air concentrations during uranium recovery operations.

Work period	Number of years	Number of potential AEC work days per work period	Ingestion rate pCi/workday[a]	Intake (pCi)
1 yr	1	250	52	1.30×10^4

a. Ingestion values were calculated according to NIOSH (2004). Choose the same f_1-value as that used for inhalation in accordance with NIOSH (2004).

Bioassay data should be used first rather than the intakes that were calculated based on air sampling results unless the bioassay data are deficient. Most potentially exposed workers at W.R. Grace/NFS were monitored. Skin contamination was a recurrent issue at W.R. Grace/NFS. The contamination levels in the claimant records should be bounded by the assumption of 10% of the skin contamination equilibrium activity levels being ingested in accordance with *Estimation of Ingestion Intakes* (NIOSH 2004). A study at the Oak Ridge Gaseous Diffusion Plant to determine the intake from hand contamination indicated that the amount of uranium that was transferred from the hand to the cigarette while smoking was approximately 1% of the material on the surface of the hand (Bailey 1959, p.166). The ingestion calculation included both contamination of food or drink from contaminated air settling and the transfer from contaminated surfaces to the hands to food or drink on a chronic basis.

3.1.2 Enriched and Recycled Uranium

For a given uranium process, the mass of (long-lived) uranium that is released to air does not change because of enrichment (ORAUT 2006a). Because the AEC air samples were counted with alpha detectors, which detect radioactivity rather than mass, there is no need to adjust measured air concentration results for assumed uranium enrichment. AEC air sample results taken during the recovery of HEU were below the measured concentrations during the recovery of LEU. Because various levels of enrichment occurred during the processing and recovery operations of uranium, this document assumes that intakes are ^{234}U for the purpose of internal dose calculation.

It should be assumed that exposure to RU potentially occurred during the entire operational period (January 1,1958, to December 31, 1970) [1]. When assigning dose due to contaminants in RU, the same material type should be applied to the contaminants as that for the uranium (ORAUT 2007a, p.21). For RU, the dose reconstructor should add the intakes from Table 3-12 (ORAUT 2005a), or those derived from bioassay results. The RU contaminant ratios are bounding for recycled components in uranium 233 and as such should be applied regardless of whether ^{233}U or ^{234}U is assumed.(Tomlinson 1964, p.6)

Table 3-12. Intakes of contaminants in RU as fractions of uranium intake.[a]

Radionuclide	Activity fraction of contaminant (e.g., pCi X/pCi U)
Uranium	1.0
Pu-238	0.06
Np-237	0.005
Tc-99	0.4
Th-232	0.02
Ru-106	0.04

a. Source: Based on ORAUT (2005a, p.11).

3.1.3 Uranium Bioassay

Urinalysis for uranium started in approximately October 1964. Samples were sent to Eberline until about 1974. From about that time, onsite analysis was performed; details of the analysis are not known. The recording level was 1 dpm/L at the 2-sigma or 95% confidence level as indicated from the review of employee documents as indicated from the NFS Health Physics department (Tester 2005a, p.2). This is likely the assumed decision level (DL). The minimum detectable level (MDL) should be twice the critical level or 2 dpm/L. Some random samples were sent off the site for fecal analysis of uranium and urine analysis for EU. Some bioassay records from W.R. Grace/NFS have some of the radiometric uranium bioassay results with a minimum recording level of <10 dpm/L. Other records have bioassay results reported on two forms: one handwritten form that indicated laboratory results with actual results recorded down to 1.0 dpm/L, and another typed report in which the lowest values are recorded as <10 dpm/L (i.e., all values <10 dpm/L were recorded as <10 dpm/L on the typed form). If the recorded result is between 2 and 10 dpm/L, 2 dpm/L should be used as the MDL. If zeros or <10 dpm/L were recorded for uranium results, then the dose reconstructor should assume the MDL is 10 dpm/L [2]. When bioassay data are available, dose reconstructors should use bioassay data to estimate worker intakes. Because the uranium urine sample results are reported in units of activity (dpm/L), it is not necessary to adjust these results to a particular uranium enrichment. Intakes of the RU components in Table 3-12 should be added based on the given ratios.

Lung counts have been performed from 1970 to the present for uranium, as indicated from a cursory review of claimant documents. The minimum detectable activity (MDA) for ^{235}U was about 120 µg (Tester 2005a, p.2).

3.2 PLUTONIUM EXPOSURES

Many forms of plutonium were possible over the years including metal and oxide. Because not enough information for the recovery or manufacture of plutonium was found, the exact chemical forms are not known. It is possible that MOX fuel of a plutonium and thorium mixture was processed at some point because UO_2 and ThO_2 mixtures were produced on the site and plutonium production capacity existed at the same time. Most of the manufacture of plutonium occurred in Buildings 234A, 234B, and 234C.

In general, plutonium oxides, carbides, and hydroxides are absorption type S; nitrates and other compounds are type M (ICRP 1995). Older materials, even when starting out as soluble, can have a tendency to oxidize when left in contact with air. Oxides, metals, and old contamination should be treated as type S. If the chemical form of plutonium is unknown, either type M or S can be used to maximize the dose to the organ of concern (ORAUT 2007a). Americium-241 is a component of plutonium contamination and should be modeled in the lung the same as the plutonium matrix in which it has grown. In other words, the americium should be treated as absorption type S if the plutonium is type S (ORAUT 2007a). It is possible that some forms of plutonium at W.R. Grace could be retained in the lung longer than is predicted by the normal material type S model (commonly referred to as type Super S). Organ doses based on intake of plutonium should be evaluated in

accordance with the guidance in the technical information bulletin *Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT2007c).

There are essentially three types of plutonium-based material – reactor, fuel, and weapons grade – with fuel grade falling between reactor and weapons grade. Although the origin of the plutonium at W.R. Grace is currently unknown, without any specific information on the actual composition of the processed plutonium an assumption of 10-year-old fuel-grade plutonium is favorable to claimants and reasonable [3]. This is noted in the Hanford site profile (ORAUT 2007b) because Hanford processed much of the DOE complex plutonium. A summary of the activity composition of the default reference fuel-grade plutonium mixture is presented in Table 3-13.

Table 3-13. Activity composition of Hanford Site reference fuel-grade plutonium mixture (12%).[a]

Radionuclides: 10 year aged[b]	**Specific activity in mixture (Ci/g)[a]**
Pu-238	1.58E-02
Pu-239	5.26E-02
Pu-240	2.72E-02
Pu-241	1.91E+00
Pu-242	3.93E-06
Am-241	3.89E-02
Pu-239+240	7.98E-02
Activity ratios	
Pu-239+240:Am-241	2.05
Pu-239+240:Pu-238	5.05
Pu-241:Pu-239+240	24.0

a. Source: ORAUT (2007b, p.19)
b. Time since separation of the Am-241 from the plutonium mix.

3.2.1 Plutonium Bioassay

A review of plutonium records during the operational period showed that ^{239}Pu was analyzed in urine from 1967 to approximately 1973 [4]. The years of plutonium exposures most likely occurred from 1967 though February 1973; however, a plutonium exposure period of 1966 through 1970 should be assumed (for the operational period), because it is possible that some startup plutonium operations could have occurred in 1966 (Claimant records; Congress 1986, p.107; and NFS 2005, p.2). Bioassay data should be used for assessing a worker's plutonium dose. No attempt should be made to estimate plutonium dose for unmonitored workers during the operational period.

Historical detection limits for W.R. Grace are not available, so a review of detection limits for the 1960s and 1970s at other AEC sites was made. The internal dosimetry section of the DOE Hanford site profile (ORAUT 2007b) reports an MDA of 0.05 dpm/sample. The Savannah River Site profile (ORAUT 2005b) has a plutonium recording level and MDA of 0.1 dpm/1.5 L. These values are reasonably consistent with the observed reporting limits from W.R. Grace. From a review of bioassay records, the lowest observed nonzero recorded plutonium results at W.R. Grace was 0.03 dpm/L, although some results were reported in units of dpm/sample, with a given sample volume. If additional information on detection capabilities is not available from the records, dose reconstructors should assume a DL of 0.03 dpm/L and an MDA of 0.06 dpm/L [5].

Chest counting for ^{239}Pu started at W.R. Grace around 1987. A germanium detector system had the ability to detect ^{239}Pu but had a variable MDA of about 168 nCi (July 6, 1989) to 481 nCi (October 22,

1987). The chest-counting detection limits are based on a cursory review of claimant files and are not useful because the MDAs are high. Most results were nondetections or perhaps near the MDA.

3.3 URANIUM-233 CONSIDERATIONS

Thorium oxide as ThO_2 and uranium oxide as $^{233}UO_2$ were the finished products for the production of $^{233}U/^{232}Th/^{228}Th$ MOX fuel. The ^{233}U was received from Oak Ridge in the form of uranyl nitrate, then precipitated to $(NH_4)_2$-U_2O_7 (ADU), then calcined to UO_2. Blending, pressing, and grinding were also a part of the process in addition to chemical processing. Particle sizes varied. Because of criticality concerns, a safe-dry batch did not exceed 4 kg of ^{233}U during blending operations; this amount of ^{233}U is nuclearly safe with a safety factor of 2.3 for a $H/^{233}U$ ratio of 1.5 and in a nominally reflected system. It should be noted that the limitation of 4 kg of ^{233}U at low $H/^{233}U$ ratios is for a metal-water system and the campaign tested was for a low-density oxide-water system. With ^{235}U, where more data are available, almost three times as much low-density ^{235}U than high density ^{235}U is nuclearly safe. No enrichment in ^{233}U was stated in the Feasibility Report, but the ZrO_2-$^{233}UO_2$ pellet Feasibility Report mentioned 93% enrichment (Housholder 1963b). Isotopic composition of $^{232}Th/^{233}U$ fuel is given in Table 3-3.

3.4 INTERNAL DOSE RECONSTRUCTION ASSUMPTIONS

Because of the SEC determination (NIOSH 2007b) that it is infeasible to adequately reconstruct internal thorium dose during the AWE operational period that is covered by the SEC (January 1, 1958 through December 31, 1970), dose estimates for this period are considered partial dose estimates.

Uranium and plutonium (and associated radionuclides) intakes should be based on actual bioassay monitoring results for the individual. When uranium bioassay results are not available, the values in Table 3-14 should be assigned for estimation of uranium dose. If a worker has positive thorium bioassay results during the operational period, these positive results can be used to reconstruct a partial thorium dose, as an approach that is favorable to claimants; however, this would still be considered a partial dose assessment because such dose is known to not be the entire possible thorium dose. In addition, if an individual was not monitored for plutonium exposures through bioassay, no attempt should be made to estimate plutonium dose during the operational period, and the dose reconstruction is to be considered to be a partial internal assessment.

Table 3-14. Uranium internal exposure summary for operational period January 1, 1958, to December 31, 1970.[a]

Internal	Start	End	Exposure	Intake (pCi/d)	IREP distribution
Uranium	1/1/1958	12/31/1970	Inhalation Ingestion	1.71E+03 3.56E+01	Constant
Pu-238	1/1/1958	12/31/1970	Inhalation Ingestion	1.03E+02 2.14E+00	Constant
Np-237	1/1/1958	12/31/1970	Inhalation Ingestion	8.55E+00 1.78E-01	Constant
Tc-99	1/1/1958	12/31/1970	Inhalation Ingestion	6.84E+02 1.43E+01	Constant
Th-232	1/1/1958	12/31/1970	Inhalation Ingestion	3.42E+01 7.13E-01	Constant
Th-228[b]	1/1/1958	12/31/1970	Inhalation Ingestion	3.42E+01 7.13E-01	Constant
Ru-106	1/1/1958	12/31/1970	Inhalation Ingestion	6.84E+01 1.43E+00	Constant

a. Ingestion values were calculated according to NIOSH (2004). Choose the same f_1-value as that for inhalation in accordance with NIOSH (2004).
b. Th-232 is assumed to be in equilibrium with Th-228.

The assumed occupational exposure period ran from January 1, 1958, to December 31, 1970. Table 3-14 lists the intake assumptions for uranium and associated compounds. These intake rates are based on information from Tables 3-10 and 3-11, which were derived from the 95th-percentile value of the average breathing-zone samples for 3.6% uranium recovery from slag (578.38 dpm/m^3), from Table 3-8, a breathing rate of 1.2 m^3/hr (ICRP 1994), and an annual occupational exposure of 2,000 hours. The intake mode is chronic.

4.0 ESTIMATION OF EXTERNAL EXPOSURE

During operations at the facility, occupational exposure occurred from handling of received material, standing near stored fissile material (either as feed or product), and airborne radioactivity with the resultant buildup of surface contamination. The fissile material was enriched UF_6 that arrived in approved shipping and storage cylinders, solid fissile material in various forms (liquid, powder, or metal) to be converted into or made into nuclear fuel, or scrap material containing LEU or HEU.

4.1 LIMITATIONS

The potential for external radiation dose existed at all locations where radioactive materials were handled or stored. Based on site operations, sources of potential exposure included beta, photon, and neutron radiation emitted from materials containing uranium, thorium, and plutonium. Personnel beta/photon dosimeters were assigned to workers. Claimant-specific data should be used for analysis of the external radiation dose. No attempt should be made to estimate external radiation doses for unmonitored workers during the operational period and, if this is done, the dose reconstruction should be considered to be a partial dose estimate. The only exception to assessing the unmonitored worker dose concerns the estimation of neutron doses to chemical operators using a neutron-to-photon dose ratio as discussed further in this section.

4.2 RECORDED EXTERNAL DOSE

Beginning in the late 1950s, Chicago Nuclear supplied all dosimetry badges and performed the necessary calibrations. At one time, everyone in the W.R. Grace plant was badged. At a later time, office workers were not assigned dosimeters (ORAUT 2005c). Landauer supplied dosimeter services beginning in 1961. The reported doses were normally adjusted for results of transit control dosimeters (i.e. control dosimeters were sent with dosimeters to be assigned to workers, and the control dosimeter results, were deducted from the personnel dosimeter results to arrive at the reported occupational dose). Minimal beta or soft X-ray nonpenetrating doses were not routinely reported until a positive skin dose was recorded (Gordon 2004, p.3). In the approximate 1989 timeframe, Landauer supplied W.R. Grace/NFS with TLDs instead of film.

External radiation exposure records at W.R. Grace/NFS utilized AEC form AEC 5, which is similar to the NRC form 5 of today. Gamma, beta, and neutron doses are listed in separate columns. In 1961, Landauer started to report the external dose in the same format as AEC form 5. In 1964, Landauer still reported the gamma, beta, and neutron dose components in separate columns but had the same forms filled out separately for whole body, skin, and extremity dose components. Code 1 was for the whole body, code 2 for the skin of the whole body, and code 3 for the extremities (i.e. hands and forearms). Beginning January 1, 1970, the reported skin dose included the shallow dose from nonpenetrating and penetrating dose components. Prior to 1970, the reported skin dose represented only the nonpenetrating dose component. As such, prior to 1970, the total shallow dose is best estimated by summing the reported skin (i.e. nonpenetrating dose) and whole body dose (i.e. penetrating dose component) doses.

Table 4-1 lists a more detailed description of the Nuclear Chicago or Landauer dosimeters used at W.R. Grace and subsequently, NFS. W.R. Grace/NFS' distribution of low-energy photons is

dependent on the amounts, separation and enrichments of uranium, and the age and type of plutonium used at the site.

Table 4-1. Dosimetry for external whole-body, wrist, and extremity exposures.

Period	Monitoring technique	Dosimeter description
Beta/photon dosimeters		
1958–June 1974 *whole body*	Photographic film badge	Nuclear-Chicago or similar film badges. Nuclear-Chicago film badge contained single film packet. Three filters (front and back) were incorporated into film badge for energy dependence: cadmium, aluminum, and lead.
July 1974–about 1983[a] *whole body*	Landauer J (beta and gamma)	Type J dosimetry were film badges. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
About 1974[a]–April 1990 *whole body*	Landauer G1	Film emulsion packaged placed in standard Gardray holder/badge for monitoring beta, X-ray, and gamma exposure. Insensitive to neutron radiation. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
May 1990–present *whole body*	Landauer Z1 dosimeter	3 TLD-700 chips for monitoring beta, X-ray, and gamma exposure. Insensitive to neutron radiation. Replaced Landauer G1.
July 1974–about 1983[a] *wrist*	Landauer Type M (wrist beta-gamma) badges.	Type M dosimetry was a film badge. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
About 1983[a]–1990 *wrist*	G5 wrist film badge	Responded to beta, X-ray, and gamma exposure to provide data on extremity dose. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.
1991–present *wrist*	K5 TLD wrist badge	3 TLD-100 chips.
1958–1974	Film badge-*finger*	Nuclear-Chicago or similar film badges. Nuclear-Chicago film badge contained single film packet. Three filters (front and back) were incorporated into film badge for energy dependence: cadmium, aluminum, and lead.
About 1983[a]–present *finger ring*	U3 TLD (LiF)	Responded to beta, X-ray, and gamma exposure to provide data on extremity dose. Gamma and X-ray: 30 keV to 20 MeV; beta: over 1.5 MeV.

a. This analysis found no documentation that shows the start of G1, U3, G5, I8, and E1 dosimeter use and the end of Type K, J, and M badge use.

The NRC has reported the annual occupational radiation dose for nuclear power reactors since 1974. Consistent annual dose reporting could not be found in NRC records for the W.R. Grace Plant until 1982. The only information found prior to 1982 was for whole-body doses greater than 1.25 rem for employees with employment of less than 90 days. The reporting format varied over the years. In the early years, the format included: the number of monitored individuals, measurable dose, collective dose, and average measurable dose in rem. In later years, the format included the annual whole-body doses segregated into dose bins with the number of workers in each bin.

4.2.1 Nonpenetrating Dose

Dosimeters used at W.R. Grace would measure beta and low-energy photon nonpenetrating dose. Prior to 1970, the reported skin dose represents only the nonpenetrating dose component and beginning January 1, 1970, the reported skin dose includes the shallow dose from the penetrating dose component. For this reason, determination of the skin dose beginning on January 1, 1970 must be calculated as the difference between the reported skin and total-body dose [6]. For low energy photon irradiation such as to 16-keV X-rays and 59-keV photons typical of plutonium facilities, there is an over-response by factors of 8.5 to 12 and 14 to 19, respectively based on measurements at Hanford (Wilson et al, 1990) when the dosimeter shallow dose is calibrated to uranium.

4.2.2 Penetrating Dose

Commercially provided personnel dosimeters used at W.R. Grace would measure the photon penetrating dose. Exposure to photon radiation from natural uranium metals and solutions would be comparatively low compared to the nonpenetrating exposure. The photon radiation is predominantly in the 30-to-250 keV energy range. As uranium becomes more enriched, the photon spectrum increases because of the higher specific activity of ^{235}U, and the energy of the photon emissions (DOE 2004). Photon radiation from plutonium would be expected to include <30 and 30-250 keV components.

4.3 NEUTRON DOSE

There were no documented neutron exposures at W.R. Grace. However, the use of uranium hexafluoride and uranium fluoride, and the possible presence of plutonium can generate neutrons through an alpha-neutron reaction between the uranium and the fluorine (DOE 2004). Neutrons can also arise from highly enriched uranium and during plutonium processing operations (DOE 2006).

Personnel exposure records are used to evaluate radiation exposure. No attempt should be made to estimate neutron dose for workers not monitored for neutrons during the operational period. If a dose reconstruction is performed, the dose reconstruction is considered to be a partial dose reconstruction and should be noted as such.

4.3.1 Extremity Doses

Extremity dosimeters for monitoring exposures to the forearm and hand were used at W.R. Grace/NFS from at least the 1960s to the 1970s. The MDL information in Table 4-2 for wrist (forearm) and finger (hand or extremity) is from Landauer. Ring and wrist badges were calibrated for high-energy gamma (for ^{137}Cs at 0.662 MeV) and high-energy beta (1.5 MeV) unless special arrangements were made (Gordon 2004, p.3). DOE (2006) provided results for measured extremity photon dose rates from plutonium glovebox operations and discusses that: “Doses to the extremities are usually dominated by gamma rays in typical glovebox operations. The extremity dose is more limiting than a whole body dose if the dose gradient is greater than 10:1 over a distance of 1 meter, the maximum distance from the fingers to the trunk of the body. In most cases, the source is not at arm’s length and the dose gradient needs to be 10:1 or 20:1 for the extremity dose to be limiting (NUREG/CR-4297; Reece et al. 1985). But in highly shielded gloveboxes, it is possible to have a very high extremity dose from dust layer on gloves; the dose to the torso can be much lower because of shielding applied to the glovebox.” Dose reconstructors should use guidance from DOE (2006) if the calculation of extremity dose is needed.

4.4 MISSED DOSE

(MDL) information for photon and beta whole-body dosimeter systems are listed in Table 4-2. The potential annual missed doses listed in Table 4-2 are considered default values and are to be used, unless claimant dosimetry records suggest that a different dosimeter exchange frequency occurred [7].

4.5 DOSE RECONSTRUCTION RECOMMENDATIONS

4.5.1 Beta Dose

Measured and missed non-penetrating doses from beta radiation should be corrected to account for attenuation by clothing or personal protective equipment (PPE), if applicable, based on the location of the cancer and the workplace practices. Nonpenetrating dose can be considered for IREP input

Table 4-2 MDL and maximum potential annual missed photon or beta dose.[a]

Dosimeter	Period of use	MDL (rem)	Maximum annual missed dose (rem) (MDL/2 × frequency)
Nuclear Chicago film– *whole body*	1957–5/1959[b]	0.04 photons 0.04 beta	0.520 beta-photons (every 2 weeks)
Nuclear Chicago film– *whole body*	6/1959–12/1960[b]	0.04 photons 0.04 beta	0.240 beta-photons (monthly)
Landauer film– *whole body*	1/1961–12/1963[b]	0.04 photons 0.04 beta	0.240 beta-photons (monthly)
Landauer film– *whole body*	1/1964–12/1988[c]	0.04 photons 0.04 beta	0.240 beta-photons (monthly)
Landauer TLD– *whole body*	1/1989–12/1998[c]	0.04 photons 0.04 beta	0.080 beta-photons (quarterly)
Landauer OSL– *whole body*	1/1/1999–12/2004[c]	0.001 photons 0.001 beta	0.008 beta-photons (quarterly)
Landauer TLD– *whole body*	1/2005–present[c]	0.02 photons 0.02 beta	0.040 beta-photons (quarterly)
Film-badge– *wrist*	1957–June 1974[b]	0.04 photons 0.04 beta	0.240 beta-photons (monthly)
Landauer Type M– *wrist*	July 1974–about 1983[c]	0.02 photons 0.04 beta	0.120 photons (monthly) 0.240 beta (monthly)
G5 wrist film badge– *wrist*	About 1983–1990[c]	0.02 photons 0.04 beta	0.120 photons (monthly) 0.040 photons (quarterly) 0.240 beta (monthly) 0.080 beta (quarterly)
K5 TLD– *wrist*	1991–present[c]	0.04 photons 0.04 beta	0.240 photons (monthly) 0.240 beta (monthly)
Film badge– *finger*	1957–1982[c]	0.04 photons 0.04 beta	0.240 beta -photons (monthly)
U3 TLD (LiF)– *finger ring*	About 1983–present[c]	0.03 photons 0.04 beta	0.180 photons (monthly) 0.060 photons (quarterly) 0.240 beta (monthly) 0.080 beta (quarterly)

a. TLD = thermoluminescent dosimeter.
b. MDLs from Tester (2005a, p.2-3).
c. Koperski (2004). MDL information as communicated by Landauer.

categories as either electrons greater than 15 keV or photons <30 keV, depending upon which selection is more reasonable based on claim information and favorability to the claimant. Typically, nonpenetrating doses for plutonium workers are assigned as <30 keV photons for deep organ cancers (e.g. nonskin cancers) using the dose conversion factors (DCFs) for plutonium workers in accordance with the *External Dose Reconstruction Implementation Guide* (NIOSH 2007a). If a nonpenetrating dose is assigned as <30 keV photons, attenuation factors for clothing are not applicable (ORAUT 2005d). A Dose Reconstructor should apply the guidance in ORAUT-OTIB-0017 for assignment of skin dose from penetrating and nonpenetrating radiation (ORAUT 2005d).

4.5.2 Photon Dose

Measured penetrating doses from photon radiation should be reasonably correct. It is possible to read a photon dose of 100 mrem to within ±15 mrem if the exposure involved photons with energies between several keV and several MeV (Morgan 1961, p.13). The estimated standard error in recorded film badge doses from photons of any energy is ±30% (ORAUT 2006c). Dose Reconstructors should include an uncertainty factor of 1.3 multiplied by the measured photon dose, assume a constant distribution for overestimates, and utilize the Exposure (R) to Organ Dose (HT), [8] dose conversion factors from IG-001. To ensure favorability to claimants, a photon energy range of 100% 30 to 250 keV should be applied (ORAUT 2006b).

4.5.3 Glovebox Geometry Assumptions

Consideration of glovebox geometry is necessary for workers with significant glovebox or similar benchtop geometry work activities with plutonium, as noted in OCAS-TIB-0010, "*Special External Dose Reconstruction Considerations for Glovebox Workers.*" (NIOSH 2005). This would include the Chemical Operators. Indicators of hands-on plutonium work may be plutonium bioassay results and/or extremity monitoring. For these work activities, adjustment to assigned doses for organs of the lower torso in relation to the measured whole-body dosimeter dose is necessary to account for potential geometry effects.

4.5.4 Uranium-233

In the spring of 1961, NFS processed 28 kg of ^{233}U in used pellets. The radiation levels reached 3,000 mR/hr at 1 in. for 15 g of U_3O_8 pellets. Twenty operators were exposed to these operations with none exceeding the 10 CFR Part 20 limits; the average exposure for the operation was 377 mrem. Rubber gloves were worn during operations in shielded gloveboxes with filtration. Birdcages were used for storage of the materials. The exposure rate for the surface of each container did not exceed 200 mR/hr, and the contamination level did not exceed 500 dpm/100 cm^2. The reading from each birdcage did not exceed 1 mR/hr at 1 meter (Householder 1963b, p.51-55). Doses from the processing of ^{233}U were measured through external monitoring. Therefore, dose reconstructors should use reported dosimetry results to assign measured dose, along with the assignment of missed dose.

4.6 OCCUPATIONAL MEDICAL DOSE

To date, no site-specific information is available for W.R. Grace workers in relation to the type or frequency of occupationally required medical X-ray examinations. The type and projection of X-ray examination should be based on current Oak Ridge Associated Universities (ORAU) Team guidance, which is a posterior-anterior (PA) radiographic chest examination [9]. Preemployment and annual PA chest x-rays are assumed for all site workers. Organ doses can be obtained from the current revision of ORAUT-OTIB-0006, *Dose Reconstruction from Occupationally Related Diagnostic X-Ray Procedures* (ORAUT 2005e). Photofluorography (PFG) could have been possible, but unless there is evidence in the claimant files that PFG was performed; it is reasonable to assume it was not [10]. Occupational medical X-ray doses should be entered into IREP as the annual dose to an acute exposure to photons with an energy range of 30 to 250 keV. The distribution is assumed to be normal with a standard deviation of 30%.

4.7 SUMMARY OF EXTERNAL DOSE RECONSTRUCTION RECOMMENDATIONS

Guidance presented in the foregoing sections to determine IREP input categories for annual external doses to be assigned is summarized in Table 4-3.

Table 4-3. IREP input summary.

Dose category	Period of exposure	Exposure category	Exposure type	Basis	Annual exposure	IREP distribution
Photon missed dose	1/1/1958–12/31/1970	Photons, 30-250 keV, AP acute	Penetrating	Table 4-2	Use MDL/2 × exchange frequency)	Lognormal GSD 1.52
Photon missed dose	1/1/1958–12/31/1970	Photons, <30 keV or electrons >15 keV, AP, acute	Non-penetrating	Table 4-2	Use MDL/2 × exchange frequency & OTIB-0017	Lognormal GSD 1.52
Photon measured dose	1/1/1958–12/31/1970	Photons, 30-250 keV, AP acute	Penetrating	Recorded value	1.3 * recorded dose	Constant

Dose category	Period of exposure	Exposure category	Exposure type	Basis	Annual exposure	IREP distribution
Photon measured dose	1/1/1958–12/31/1970	Photons, <30 keV or electrons >15 keV, AP, acute	Non-penetrating	Recorded value	1.3 * recorded dose	Constant
Medical X-ray	1/1/1958–12/31/1970	PA radiographic chest exam	Penetrating	Initial plus annual examinations	See Section 4.4.6	See ORAUT (2005e)

5.0 ESTIMATION OF EXPOSURE TO RESIDUAL ACTIVITY

The W.R. Grace/NFS plant continued to operate after the covered AWE period, which ended on December 31, 1970. The *Report on Residual Radioactive and Beryllium Contamination at Atomic Weapons Employer Facilities and Beryllium Vendor Facilities*, from December 2006 noted that a significant potential for residual contamination remained after 1970. The residual contamination period at W.R. Grace is from January 1, 1971, to the present (NIOSH 2006). Differentiation between residual contamination during the operational period at W.R. Grace and contamination due to ongoing operations is not feasible.

5.1 INTERNAL EXPOSURE TO RESIDUAL ACTIVITY

5.1.1 Introduction

During the residual radioactivity period at W.R. Grace (1971 to the present), personnel might have been internally exposed to uranium, plutonium, and thorium in the workplace. This section describes the methdology for the assignment of internal dose.

Internal exposures should be estimated using bioassay data, if they exist. For situations where no internal monitoring was performed or monitoring information is not available, the default intakes for uranium, plutonium, and thorium should be assigned, unless the potential for exposure to these radionuclides can be ruled out. In situations where there are bioassay data, but gaps exist, the default intakes should be assigned from the day following the date of the last bioassay sample, through the end of the potential exposure period (ORAUT 2007a, p. 14).

The following information provides a method for estimating exposures during the residual radiation period due to uranium, thorium, and plutonium contamination, in cases where bioassay data is unavailable.

5.1.2 Uranium

If bioassay data are available for the residual period, then that data should be used to estimate and assign internal dose for the worker. Internal doses from bioassay data for this period should be evaluated with the guidance in Section 3.0. Intakes that are calculated in this manner are considered an overestimate because only the contribution from residual contamination should be applied. If a best estimate must be calculated, source term depletion factors from Table 3-1 of ORAUT (2008) may be applied as appropriate.

When bioassay data are not available, and the worker might have worked in areas with residual activity, the following method may be used to assign internal dose.

The uranium intakes (and associated RU components) in Section 3.0 (Table 3-14) may be used as a basis for intakes during the residual period. These tabulated values are used to calculate an

associated average daily air concentration (assuming 250 workdays per calendar year and a breathing rate of 9.6 m^3/d).

An average daily air concentration is used to estimate the residual surface concentration with guidance from ORAUT (2008). The annual deposition amount is estimated with a deposition velocity of 0.00075 m/s and with deposition assumed to occur for 1 year. Using this approach a surface concentration of uranium is estimated as follows:

$$\textit{air concentration}\ \text{pCi/m}^3 \times 31{,}536{,}000\ \text{s/yr} \times 0.00075\ \text{m/s} = \textit{surface activity}\ \text{dpm/m}^2 \quad \text{(Eq. 1)}$$

This mean surface concentration is favorable to claimants at the end of the operating period. The deposited material is assumed to have been resuspended and inhaled during the residual period. The amount of resuspension is assumed to reduce with time due to fixing of the material on surfaces and to also due to depletion (ORAUT 2008). The depletion factors for each year are described in Table 3-1 of ORAUT (2008). The depletion factors indicate that the residual concentration at the end of the operational period is to be used for the first year, the second year is reduced by a factor of 0.03, and the third and remaining years are reduced by a factor of 0.0007 (which represents a constant concentration after the third year).

The intakes in Table 5-1 can be used to estimate the internal dose to the target organ for the years of employment for the worker. The estimated internal doses are assigned as a constant distribution.

Table 5-1. Uranium (and RU components) air concentration and annual intake in the residual period.

Nuclide	Intake (pCi/yr)		
	1971	**1972**	**1973–present**
U-234	1.5E+04 Inhalation 3.12E+02 Ingestion	4.4E+02 Inhalation 9.2E+00 Ingestion	1.0E+01 Inhalation 2.1E-01 Ingestion
Pu-238	8.9E+02 Inhalation 1.9E+01 Ingestion	2.7E+01 Inhalation 5.6E-01 Ingestion	6.2E-01 Inhalation 1.3E-02 Ingestion
Np-237	7.4E+01 Inhalation 1.5E+00 Ingestion	2.2E+00 Inhalation 4.6E-02 Ingestion	(a)
Th-232	3.0E+02 Inhalation 6.2E+00 Ingestion	8.9E+00 Inhalation 1.9E-01 Ingestion	(a)
Th-228	3.0E+02 Inhalation 6.2E+00 Ingestion	8.9E+00 Inhalation 1.9E-01 Ingestion	(a)
Tc-99	(a)	(a)	(a)
Ru-106	(a)	(a)	(a)

a. No intake assigned, <0.001 rem/yr.

The internal dose analysis should include the potential inadvertent ingestion of uranium activity and be based on guidance in NIOSH (2004). The daily intake rate in activity per day is estimated as 0.2 times the average daily air concentration expressed in units of activity/m^3. Using the air concentration for the first year after the end of operations (1971) from Table 5-1, this value should be applied for all years of the residual period using the depletion factors as described in Table 3-1 of ORAUT (2008). For ingestion intakes, it is not necessary to include intakes of ^{228}Th and ^{237}Np from 1973 through the present, because these intakes result in < 0.001 rem per year. It is also not necessary to include inhalation and ingestion intakes of ^{99}Tc and ^{106}Ru from 1971 to the present, because these intakes result in < 0.001 rem per year.

5.1.3 Plutonium

Plutonium urine sample results from the operational period were reviewed, sorted from lowest to highest, ranked and statistically analyzed (ORAUT 2005f) [11]. A summary of the urine sample

results is presented in Table 5-2. The highest reported level was 2.856 dpm/d (after normalizing the result) from October 3, 1967. Seventy-five sample results were taken during the operational period (October 1, 1958, to December 31, 1970). Thirty-six (approximately 49%) of these sample results were less than the assumed DL of 0.03 dpm/L. A review of the available plutonium data (to date) suggests that they are representative of the types of workers most likely to be exposed to plutonium and includes: chemical operators, operators, production and maintenance foreman, front line supervisors, and radiation technicians. Table 5-3 summarizes the 95th-percentile excretion rate, which was derived from the plutonium urinalysis results during the operational period.

Table 5-2. Plutonium urinalysis results from operational period (January 1, 1958 to December 31, 1970) as of April 17, 2007.

Sample date(s)	Result (dpm/d)[a,b]	Sample #'s
10/11/67–12/05/70	<LOD[c]	1–36
12/4/1967	0.042	37
12/5/1967	0.042	38
11/3/1967	0.056	39
9/3/1968	0.056	40
5/29/1968	0.070[d]	41
5/19/1969	0.070	42
12/7/1967	0.084	43
10/15/1968	0.084	44
7/31/1969	0.084	45
12/7/1967	0.098	46
12/13/1967	0.098[d]	47
8/30/1970	0.098	48
12/5/1967	0.126	49
5/20/1969	0.126	50
11/17/1970	0.126	51
8/6/1968	0.140	52
9/7/1967	0.154	53
8/27/1970	0.154	54
12/28/1967	0.182	55
2/20/1969	0.182	56
8/23/1970	0.182	57
11/10/1967	0.224	58
2/5/1968	0.238	59
11/19/1967	0.266[d]	60
2/21/1969	0.266	61
8/1/1969	0.266	62
10/5/1967	0.280	63
10/8/1967	0.350	64
10/5/1967	0.392	65
12/13/1967	0.392	66
11/10/1967	0.434[d]	67
3/4/1968	0.476[d]	68
8/30/1968	0.602	69
10/4/1970	0.742	70
8/6/1968	0.966	71
8/5/1968	1.344	72
7/28/1968	1.400	73
8/5/1968	1.624	74
10/3/1967	2.856[d]	75

a. Results were normalized using 1.4 L/d.

b. Assume DL = 0.03 dpm/L (normalized = 0.042 dpm /d).
c. LOD = limit of detection.
d. Positive sample results all from one individual (a production worker/laboratory technician), who also had the highest result. This same individual had two <LOD results on October 11, 1967, and August 8, 1968.

Table 5-3. Summary of excretion rates from plutonium urinalysis results from operational period (dpm/d).[a]

GM	GSD	95th percentile
0.058	5.6	1.01

a. Excretion rates were determined by log-transforming the data, calculating the *z*-score, plotting the data, associated R^2 fit parameter to estimate a 50th-and 95th-percentile values (ORAUT 2005f).

The excretion rates from Table 5-3 were modeled in IMBA, assuming a chronic intake from the date of the first bioassay sample on September 7, 1967, through the date of the last bioassay sample on November 21, 1970, and using an effective bioassay sample date of the midpoint of this intake period. Based on this analysis, an intake rate of 563 dpm/d was calculated (based on the GM). The intake rate at the 95th-percentile value was 9,760 dpm/d.

The plutonium intake during the operational period as calculated above was used as the basis for plutonium intakes at the start of the residual period (1971). The 95th-percentile intake rate (dpm/d) was used to calculate an associated daily air concentration (assuming 250 workdays per calendar year and a breathing rate of 9.6 m^3/d). This daily air concentration was then used to estimate the residual surface concentration with guidance from the technical information bulletin (ORAUT 2008). The annual deposition amount was estimated using a deposition velocity of 0.00075 m/s, with deposition assumed to occur for 1 year. Using this approach a surface concentration of plutonium is estimated as follows.

$$\textit{air concentration}\ \text{pCi/m}^3 \times 31{,}536{,}000\ \text{s/yr} \times 0.00075\ \text{m/s} = \textit{surface activity}\ \text{dpm/m}^2 \quad \text{(Eq. 2)}$$

The deposited material was assumed to have been resuspended and inhaled during the residual period. The calculated intake rate was 84,000 dpm/yr. Plutonium intakes during subsequent residual period years was calculated based on air monitoring data available for the period from 1976 through 1989 (Table 5–4). The highest sitewide air concentration for the period was used to estimate an intake rate starting in 1976 that was assumed to remain constant. Because the data is in fractions of the maximum permissible concentration (MPC), it was necessary to assume that all of the gross alpha air activity was plutonium and using the plutonium MPC value (2×10^{-12} µCi/ml). The intake rate that was calculated for 1976 and subsequent years was 5,168 dpm/yr. Intake rates for 1972 through 1975 were calculated based on the 1971 and 1976 values by fitting them to an exponential function.

Plutonium intakes during the residual period are summarized in Table 5-5.

The internal dose analysis should include the potential inadvertent ingestion of plutonium activity based on the guidance in NIOSH (2004). The daily intake rate (dpm/d) was estimated as 0.2 times the daily air concentration. The estimated internal doses are assigned as a constant distribution in IREP.

Table 5-4. Air-sampling results in percentage of MPCs[a] during the residual period, 1976 to 1989.[b]

Building/area/process	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	Average
105 (302) Laboratories	3.90	4.60	3.20	3.90	4.55	5.15	3.00	1.75	1.48	0.58	1.42	1.46	0.71	0.62	2.59
100 Decontamination Laundry	1.40	0.90	1.50	2.20	1.70	1.75	1.85	1.44	1.76	0.83	1.25	1.14	0.31	0.43	1.32
105 Hall									1.39	0.68	2.09	2.18	1.01	0.94	1.38
110A, B LEU Scrap Rec, Store & Staging	1.60	2.20	2.10	3.20	4.00	4.56	0.95	0.95	1.01	0.56	0.60	1.48	0.24	0.14	1.68
110C, D Pu U-233 Labs	19.20	22.00	22.60	14.30	24.00	15.35	6.30	6.25	17.13	11.88	22.57	8.19	2.76	1.64	13.87
110D-1 U Spectrographic lab	4.70	3.00	1.50	5.00	2.40	1.80	0.65	0.41	0.43	0.43	0.64	0.40	2.39	1.27	1.79
111 Chemical LEU scrap recovery	3.10	4.60	3.60	5.10	5.65	4.45	4.35	1.58	3.64	3.38	2.24	1.21	0.15	0.13	3.08
120 Maintenance	1.40	1.30	3.70	3.20	2.10	2.45	1.95	2.02	1.22	0.50	1.42	0.77	0.19	0.14	1.60
130 (Metals) LEU UF6 Cyl wash	6.50	1.70	3.80	5.60	16.05	43.80	9.80	0.59	0.69	0.95	1.92	1.95	0.32	0.85	6.75
131 Pilot Plant–R & D	2.20	0.60	1.90	3.00	1.85	2.25	0.95	0.51	0.51	0.34	0.57	0.40	0.15	0.05	1.09
220 NDA Scanning	1.10	1.10	1.80	4.20	1.35	1.90	1.35	1.16	0.81	0.67	1.98	1.09			1.54
230 Complex									2.11	0.70	1.86	1.76	0.66	0.79	1.31
233 HEU Scrap Recovery	31.70	14.60	15.20	12.50	10.40	9.35	7.50	5.54	4.31	3.10	5.50	3.61	3.61	3.27	9.30
234A Pu Production Areas	3.20	6.30	18.00	20.40	10.00	11.90	8.40	7.15	26.13	11.88	10.63	0.00	0.00	0.00	9.57
234B U-233 Process Area	0.30	0.30	0.50	0.40	0.45	0.50	0.35	0.45	0.52	0.25	0.00	0.00	0.00	0.00	0.29
234C Pu Dissolution Area	26.00	6.40	21.00	12.70	10.50	10.00	7.15	8.75	18.38	11.46	9.00	0.00	0.00	0.00	10.10
234C Fabrication Area	48.50	6.40	18.10	13.50	10.75	11.55	8.55	10.13	22.38	11.47	9.75	0.00	0.00	0.00	12.22
234 building											8.25	7.79	3.62	1.63	5.32
300 Nuclear Materials Warehouse	0.60	0.80	1.20	2.40	1.55	2.10	0.85	0.60	0.47	0.33	0.33	0.31	0.00	0.00	0.82
301 LEU Oxide Production	0.40	0.50	4.00	10.70	2.95	3.30	1.20	1.39	0.66	0.46	2.00	0.98	0.36	0.58	2.10
302 Finished Fuel Production	38.60	25.10	20.40	24.70	12.25	14.35	13.90	16.06	6.52	3.59	7.46	4.39	5.97	7.92	14.37
302 Retort Tube Cleaning	32.30	32.70	40.00	31.20	23.05	22.90	22.15	31.36	11.75	5.47	0.00	0.00	0.00	0.00	18.06
302 Incinerator	9.80	10.50	10.80	30.00	15.55	31.40	10.50	9.19	4.27	1.90	0.00	0.00	0.00	0.00	9.56
302 Lunchroom	9.50	8.50	5.00	4.80	3.25	2.75					0.00	0.00	0.00	0.00	3.38
303 Finished Fuel Production	27.20	27.00	14.50	29.90	18.05	25.80	18.55	13.04	5.16	7.63	11.11	9.68	8.07	10.22	16.14
304 Finished Fuel Warehouse	3.50	3.00	4.50	7.40	6.40	7.70	3.60	3.27	1.98	1.22	2.21	1.58	0.95	1.06	3.45
304 Warehouse excluding storage									2.15	1.22	3.34	3.54	0.00	0.00	1.71
305 Lunchroom									1.78	0.69	0.86	0.57	0.26	0.18	0.72
306 Complex									0.99	1.07	1.74	2.18	0.00	0.00	1.00
310 Scrap Metal Warehouse	0.50	0.40	1.80	4.00	2.75	3.65	1.10	0.39	0.51	0.35	0.51	0.29	0.00	0.00	1.16

a. MPC is considered to be the most restrictive NRC limit whether soluble or insoluble (i.e., 2×10^{-12} µCi/ml for plutonium).

b. Data adapted from Tester (2005b,c,d,e).

Table 5-5. Plutonium air concentration and annual intake in the residual period.

Year	Air concentration (dpm/m^3)	Inhalation (dpm/yr)	Ingestion (dpm/yr)
1971	35	8.4E+04	1.8E+03
1972	29	7.0E+04	1.5E+03
1973	20	4.8E+04	1.0E+03
1974	11	2.8E+04	5.5E+02
1975	5.4	1.3E+04	2.7E+02
1976	2.1	5.2E+03	1.1E+02
1977–present	2.1	5.2E+03	1.1E+02

5.1.4 Thorium

Air-sampling data from thorium processing facilities, which is summarized in ORAUT-OTIB-0070 (ORAUT 2008), were used as the basis for airborne thorium concentrations at the end of the operational period. Data from the Lindsay Light facility were used as a surrogate for W.R. Grace. The Lindsay Light facility was selected as a basis for the upper bound of the radiological conditions in thorium process areas at W.R. Grace due to the nature of the activities at Lindsay (operations with thorium compounds such as thorium nitrate and thorium oxide) and the likely presence of these types of materials at W.R. Grace. Radiological conditions during the meteorological processing of thorium are bounded by those during the processing of the precursor materials. Since W.R. Grace handled both the source materials (thorium compounds) and produced thorium metals, the use of the data from Lindsay is believed to be bounding for all of the activities performed at W.R. Grace. The 95th-percentile air concentration (411 dpm/m^3) was assumed to be bounding for the general area air concentration at the end of the operational period (1971). This air concentration was then used to estimate the residual surface concentration with the guidance from the technical information bulletin (ORAUT 2008). The annual deposition amount was estimated using a deposition velocity of 0.00075 m/s, with deposition assumed to occur for 1 year. Using this approach, a surface concentration of thorium was estimated as follows.

air concentration pCi/ m^3 × 31,536,000 s/yr × 0.00075 m/s = *surface activity* dpm/m^2 (Eq. 3)

This mean surface concentration is favorable to claimants at the end of the operating period. The deposited material was assumed to have been resuspended and inhaled during the residual period. The amount of resuspension was assumed to be reduced with time due to fixing of the material on surfaces and to also due to depletion (ORAUT 2008). The depletion factors that were applied to each year are described in Table 3-1 of ORAUT (2008). The depletion factors indicate the residual concentration at the end of the operational period is to be used for the first year, the second year concentration is reduced by a factor of 0.03, and the concentration for the third and remaining years is reduced by a factor of 0.0007 (representing a constant concentration after the third year).

Tabulated values represent total thorium activity; as such, they should be distributed between ^{232}Th and ^{228}Th in a manner favorable to claimants (West 1965; GGA 1971) [12]. An equal intake of ^{228}Ra should be added.

The intakes in Table 5-6 can be used to estimate the internal dose to the target organ for the years of employment for the worker. The estimated internal doses should be assigned as a constant distribution.

Table 5-6. Thorium air concentration and annual intake in the residual period.

Nuclide	Intake (pCi/yr)		
	1971	1972	1973–present
Th-232/Th228	1.1E+04 Inhalation 2.3E+02 Ingestion	3.2E+02 Inhalation 6.7E+00 Ingestion	7.4E+00 Inhalation 1.5E-01 Ingestion
Ra-228	1.1E+04 Inhalation 2.3E+02 Ingestion	3.2E+02 Inhalation 6.7E+00 Ingestion	7.4E+00 Inhalation 1.5E-01 Ingestion

The internal dose analysis should include the potential inadvertent ingestion of thorium activity using the guidance in NIOSH (2004). The daily intake rate is estimated as 0.2 times the average daily air concentration. Using the air concentration for the first year after the end of operations (1971) from Table 5-6, this value should be applied for all years of the residual period using the depletion factors as described in Table 3-1 of ORAUT (2008).

5.2 ESTIMATION OF EXTERNAL EXPOSURE FROM RESIDUAL RADIOACTIVITY

Radiation exposure records from 1971 to the present should be used to estimate external exposure to residual activity, as described in Section 4.0. The measured dose includes the NFS non-DOE operational dose plus whatever DOE residual dose may exist. As such, the measured dose will over-estimate the dose from DOE operations only.

The lack of a neutron personnel dosimetry program during the residual period is an indication of the low potential for neutron exposure. It is possible, however, that some minimal exposure to neutron radiation (from contaminated surfaces or held-up material) did occur. Any additional dose from unmonitored neutron exposure is likely bounded by the overall measured photon dose which includes dose from non-EEOICPA activities in addition to any residual dose covered under the EEOICPA program.

In the absence of claim-specific information, the following assumptions should be made about external exposures [13]:

Residual period	Sources of external exposure	Default assumptions
1971–to the present	Photons, 30–250 keV Electrons, > 15 keV or <30 keV	Exposure (R) to Organ Dose (HT) Follow ORAUT-OTIB-0017 (ORAUT 2005d) requirements

For unmonitored workers, dose information from site external radiation summary reports (available from 1976 through 1988) can be used as a basis for external dose. The penetrating exposure of 100 mrem (Table 5-7 under the average row for 1978) was the maximum listed value from 1976 through 1988. This 100-mrem penetrating exposure value between the external dose equivalent values was calculated from possible contamination source terms. It is not likely that unmonitored coworkers would have received more external dose from residual AEC sources than that recorded for coworkers who were monitored for NRC- or DOE-related activities [14].

For unmonitored workers the dose reconstructor should take the following steps:

1. For the penetrating dose estimate, take the highest annual coworker average penetrating exposure and add a missed dose calculation. The highest annual exposure of 100 mrem should be applied to all years of unmonitored residual years. The 100 mrem is based on the recorded coworker exposure history from Table 5-7 for 1978.

   An example calculation is as follows for the year:

Use the coworker penetrating exposure estimate of 100 mrem/yr, and add the missed dose calculation (40 mrem per badge cycle is the limit of detection from Table 4-2 for the Landauer film whole-body period of January 1964 to December 1988), which is 40 mrem ÷ 2 × 11 months = 220 mrem (The 100-mrem coworker dose counts as a recorded dose and therefore there are only 11 months of unrecorded or missed dose.)

100 mrem (high average coworker) + 220 mrem (missed dose) = 320 mrem

A value of 320 mrem/yr for penetrating dose should be assigned to unmonitored workers, assigned as a constant distribution, and a 30-250 keV photons energy range.

2. For the nonpenetrating dose estimate, take the highest average annual coworker average nonpenetrating exposure and do not add a missed dose calculation.

   An example calculation is as follows for the year:

   Coworker nonpenetrating exposure estimate, which is 390 mrem/yr (390 mrem based on the highest average recorded coworker nonpenetrating exposure history in Table 5-7, 1976 row). There is no necessity to add a missed dose calculation to the nonpenetrating dose estimate because it has already been accounted for in the missed dose penetrating estimate (ORAUT 2005d).

   390 mrem (high average coworker)

   A value of 390 mrem/yr for nonpenetrating dose should be assigned to unmonitored workers, assigned as a constant distribution, and as electrons > 15 keV or < 30 keV photons, consistent with the approach described in Section 4.4.1.

Table 5-7. W.R. Grace/NFS external exposure summary (NFS 1980, 1980-1989).[a]

Penetrating exposure range	**1976**	**1977**	**1978**	**1979**	**1980**	**1981**	**1982**	**1983**	**1984**	**1985**	**DDE range**	**1986**	**1987**	**1988**	
No measurable exposure	320	322	297	364	149	316	147	353	278	309	M	500	532	603	
<100 mrem	175	144	165	150	504	496	943	546	594	560	>M-50	253	319	348	
>100 mrem <250 mrem	7	44	60	1	101	107	70	44	31	2	>50-100	127	24	56	
>250 mrem <500 mrem	0	1	8	0	5	6	1	1	1	0	>100-150	19	2	2	
>500 mrem <1,500 mrem	0	0	3	0	1	0	0	0	0	0	>150	5	4	1	
											Max	180	340	160	
Total	502	511	533	515	760	925	1,161	944	904	871	**Total**	904	881	1,010	**Total average**
High	0.25	0.50	1.50	0.25	1.50	0.50	0.50	0.50	0.50	0.25		0.18	0.34	0.16	0.53
Average	0.05	0.08	0.10[b]	0.05	0.07	0.08	0.06	0.06	0.06	0.05		0.04	0.03	0.03	0.06
Nonpenetrating exposure range											**SDE range**				
No measurable exposure	381	404	417	431	149	316	946	799	277	307	M	493	522	592	
<600 mrem	117	107	113	84	609	606	218	145	627	564	>M-50	237	320	358	
>600 mrem <1,500 mrem	1	0	3	0	2	3	0	0	0	0	>50-100	147	33	57	
>1,500 mrem <3,000 mrem	1	0	0	0	0	0	0	0	0	0	>100-150	19	2	2	
>3,000 mrem <4,500 mrem	1	0	0	0	0	0	0	0	0	0	>150	8	4	1	
>4,500 mrem	1	0	0	0	0	0	0	0	0	0					
											Max	290	340	160	
											Total	904	881	1,010	**Total average**
High	4.52	0.60	1.50	0.60	1.50	1.50	0.60	0.60	0.60	0.60		0.29	0.34	0.16	1.03
Average	0.39[c]	0.30	0.32	0.30	0.30	0.30	0.30	0.30	0.30	0.30		0.04	0.03	0.03	0.25

a. W.R. Grace did not provide averages and collective dose before 1977; the average has to be approximated. This will sometimes overestimate the dose. NFS Radiation Exposure Summary Reports are available for 1977-2005.

b. This represents the highest value of the average penetrating dose for years 1976-1988.

c. This represents the highest value of the average non-penetrating dose for years 1976-1988.

6.0 ENVIRONMENTAL DOSE

It is not necessary to include an environmental dose component for W.R. Grace worker dose because all workers are assumed to have been exposed to operational conditions, and dose has been assigned accordingly.

7.0 ATTRIBUTIONS AND ANNOTATIONS

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

[1] Guido, Joseph. ORAU Team Member. March 2008
Because it is unknown, it is favorable to assume RU radionuclides were present during the entire operational period.

[2] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. March 2006.
Based on personal observations during review of claimant uranium urine results, if the recorded result is between 2 and 10 dpm/L, it is reasonable to assume 2 dpm/L as the MDL. If there are zeros or "<10 dpm/L" recorded for uranium results, then the dose reconstructor should assume the MDL is 10 dpm/L.

[3] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. March 2006.
An assumption of 10-year-old fuel-grade plutonium is favorable to claimants and reasonable in the absence of claimant-specific information. This is also noted in the Hanford site profile (ORAUT 2007b).

[4] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. March 2006.
Personal observations during review of plutonium bioassay records show in general that plutonium urine sampling occurred from 1967 through approximately 1973.

[5] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. March 2006.
Personal observations during a review of plutonium bioassay records and plutonium DLs and MDAs from other sites show that a plutonium DL of 0.03 dpm/L and a MDA of 0.06 dpm/L is reasonable.

[6] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. March 2006.
The observation that the reported skin dose represents only the shallow dose component before 1970, and the dose after January 1, 1970, includes the penetrating gamma component, is from a cursory review of claimant dosimetry records.

[7] Demopoulos, Paul J. ORAU Team Member. Senior Health Physicist. March 2006.
Based on personal observations during a review of dosimetry records. Actual recorded badge frequencies should always be used in lieu of these default assumptions.

[8] Smith, Matthew, and Winslow, Rob, ORAU Team Members. June 2007.
In the absence of further information, it is appropriate to apply these DCFs.

[9] Thomas, Elyse M. ORAU Team. Principal Medical Dosimetrist, February 2007.
The default assumption for the frequency of chest X-rays for AWE sites is a PA chest X-ray at preemployment, annually, and at termination, as stated in an e-mail from E. Thomas to C. Bloom (February 23, 2007).

[10] Thomas, Elyse M. ORAU Team. Principal Medical Dosimetrist. February 2007
It is reasonable to assume that PFG was not performed unless there is information in the claimant files to suggest otherwise, as stated in an e-mail from E. Thomas to C. Bloom (February 23, 2007).

[11] Labone, Thomas. ORAU Team. Deputy Principal Internal Dosimetrist. April 2007.
Statistical analysis was performed by Thomas Labone.

[12] Olsen, Bernard M. ORAU Team. Senior Health Physicist. February 2008.
The ratio of ^{232}Th to ^{228}Th is variable and ranges from equal activity concentrations minimum condition of ^{228}Th being present at approximately 40% the activity concentration of ^{232}Th (West 1965; GGA 1971). The relative ratio of ^{232}Th to ^{228}Th is significant for dose reconstruction. Organ dose that is determined by applying the DCFs of ICRP Publication 68 (ICRP 1995) is maximized for nonrespiratory cancers by assuming all thorium is ^{232}Th, while the dose is maximized for respiratory cancers by assuming that all thorium is ^{228}Th. For a nonrespiratory cancer, it is favorable to the claimant to assume that all the thorium is ^{232}Th. For a respiratory cancer, it is favorable to the claimant to assume that all the thorium is ^{228}Th. For a best-estimate evaluation, dose reconstructors should assume that ^{232}Th and ^{228}Th are present in ratios of 1 to 1 for a respiratory cancer, or 7 to 3for a nonrespiratory cancer (the minimum condition of ^{228}Th present at 40% that of ^{232}Th) .

[13] Fix, Jack, Smith, Matthew, and Winslow, Rob. ORAU Team. ORAU Team Members. March 2008.
Default DCFs from NIOSH (2007a) were identified for photons and electrons for the respective operational and residual periods.

[14] Demopoulos, Paul J. ORAU Team. Senior Health Physicist. April 2007.
The residual period AEC source term was not likely to cause exposures above those during the AEC operational period because contamination levels would likely decrease, not increase, with the cessation of AEC-related operations. The amount of this decrease at this time cannot be defined.

REFERENCES

AEC (U.S. Atomic Energy Commission), 1959, *Davison Chemical Company, Nuclear Reactor Materials Plant, Erwin, Tennessee: Occupational Exposure to Radioactive Dust*, HASL-75, Health and Safety Laboratory, Environmental Sciences Division, Field Services Branch, November. [SRDB Ref ID: 11771]

AEC (U.S. Atomic Energy Commission), 1961, *Davison Chemical Company, Nuclear Reactor Materials Plant, Erwin, Tennessee: Occupational Exposure to Radioactive Dust*, HASL Technical Memorandum 61-9, Health and Safety Laboratory, Health Protection Engineering Division, August. [SRDB Ref ID: 11771]

Bailey, J. C., 1959, "Personnel Contamination as a Uranium Hazard," *Symposium on Occupational Health Experience and Practices in the Uranium Industry, Held in New York City, October 15 – 17, 1958*, HASL-58, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York Operations Office, New York, New York, September, p. 157. [SRDB Ref ID: 7886]

Congress (U.S. Congress), 1986, *Erwin Nuclear Fuel Services – Hearing before the Subcommittee on Energy Conservation and Power of the Committee on Energy and Commerce House of Representatives, Ninety-ninth Congress, Second Session*, Serial No. 99-178, September 18, Washington, D.C. [SRDB Ref ID: 18808]

DOE (U.S. Department of Energy), 2004, *Guide of Good Practices for Occupational Radiological Protection in Uranium Facilities*, DOE-STD-1136-2004, Washington, D.C., December. [SRDB Ref ID: 4617]

DOE (U.S. Department of Energy), 2006, *Guide of Good Practices for Occupational Radiological Protection in Plutonium Facilities*, DOE-STD-1128-98, Change Notice 2, Washington, D.C., December. [SRDB Ref ID: 34061]

GGA (Gulf General Atomics), 1971, *Estimating Airborne Radioactivity from Thorium and its Daughters at FOD*, Issue 1, San Diego, California, February 4. [SRDB Ref ID: 20394]

GAO (General Accounting Office), 1967, *Report to Joint Committee on Atomic Energy, Congress of the United States, Review of Accountability Controls Over Special Nuclear Materials, Nuclear Fuel Services, Inc*, ; Comptroller General of the United States, September. [SRDB Ref ID: 7214]

Glauberman, H., 1962, "Feasibility Report for the New Highly Enriched Scrap Recovery Facilities at NRMP (Davison Chemical Company)," internal memorandum to H. Werner (Technical Services Division), June 27. [SRDB Ref ID: 11783]

Gordon, M., 2004, "Landauer Dosimetry Info from 1970s," documented communication with B. Koperski (R. S. Landauer), Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, October 27. [SRDB Ref ID: 23570]

Housholder, W. R., 1963a, *Feasibility Report for the Production of Uranium Metal and Uranium Metal Alloys Enriched Up To 12% ^{235}U*, W.R. Grace, Davison Chemical Division, Erwin, Tennessee, October. [SRDB Ref ID: 11783]

Housholder, W. R., 1963b, *Revised Feasibility Report for the Production of $^{233}UO_2/ThO_2$ and $^{233}UO_2/ZrO_2$ Pellets*, W.R. Grace, Davison Chemical Division, Erwin, Tennessee, November. [SRDB Ref ID: 11772]

Housholder, W. R., and Runion, H. L., 1962, *Feasibility Report for New Highly Enriched Scrap Recovery Facilities at NRMP*, W.R. Grace, Davison Chemical Division, Erwin, Tennessee. [SRDB Ref ID: 11783, p. 177]

HPS (Health Physics Society), 1995, *An American National Standard for Bioassay Programs for Uranium*, ANSI Standard N13.22-1995, McLean, Virginia, October.

ICRP (International Commission on Radiological Protection), 1994, *Human Respiratory Tract Model for Radiological Protection*, Publication 66, Pergamon Press, Oxford, England.

ICRP (International Commission on Radiological Protection), 1995, *Dose Coefficients for Intakes of Radionuclides by Workers*, Publication 68, Pergamon Press, Oxford, England.

Katine, E., 1960, "Davison Chemical Company Scrap Recovery Process under AEC Contract AT(30-1)2659," memorandum to H. V. Werner (Nuclear Safety Review Board), December 21, Davison Chemical Company, Erwin, Tennessee. [SRDB Ref ID: 11732, p. 18]

Meiners, S. 1999, *Paducah UF_6 Cylinder Painting Project*, Bechtel Jacobs Company, July 7. [SRDB Ref ID: 13682]

Morgan, K. Z., 1961, "Dosimetry Requirements for Protection from Ionizing Radiation," *Selected Topics in Radiation Dosimetry, Proceedings of the Symposium on Selected Topics in Radiation Dosimetry, Sponsored by and Held in Vienna 7-11 June 1960*, International Atomic Energy Agency, Vienna, Austria, pp. 3–23. [SRDB Ref ID: 8345]

NFS (Nuclear Fuel Services), 1980, *Radiation Exposure Summary 1977-1979*, January 3. [SRDB Ref ID: 19315]

NFS (Nuclear Fuel Services), 1980-1989, *Radiation Exposure Summary 1980-1989*. [SRDB Ref ID: 19316]

NFS (Nuclear Fuel Services), 2005, "NFS Timeline," http://www.nuclearfuelservices.com/timeline-more.htm, accessed April 11, 2005. [SRDB Ref ID: 18804]

NIOSH (National Institute for Occupational Safety and Health), 2004, *Estimation of Ingestion Intakes*, OCAS-TIB-009, Rev. 0, Office of Compensation Analysis and Support, Cincinnati, Ohio, April.

NIOSH (National Institute for Occupational Safety and Health), 2005, *Best Estimate External Dose Reconstruction for Glovebox Workers,* OCAS-TIB-0010, Rev. 02, Office of Compensation Analysis and Support, Cincinnati, Ohio, December 30.

NIOSH (National Institute for Occupational Safety and Health), 2006, *Report on Residual Radioactive and Beryllium Contamination at Atomic Weapons Employer Facilities and Beryllium Vendor Facilities*, December. [Appendix A-3 p. 222 of 236]

NIOSH (National Institute for Occupational Safety and Health), 2007a, *External Dose Reconstruction Implementation Guideline*, OCAS-IG-001, Rev. 3, Office of Compensation Analysis and Support, Cincinnati, Ohio, November 21.

NIOSH (National Institute for Occupational Safety and Health), 2007b, *SEC Petition Evaluation Report SEC-00082*, Office of Compensation Analysis and Support, Cincinnati, Ohio, March 26.

Olson, G. L., R. K. McCardell, and D. B. Illum, 2002, *Fuel Summary Report: Shippingport Light Water Breeder Reactor*, INEEL/EXT-98-00799, Rev. 2, Bechtel BWXT Idaho, Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, September. [SRDB Ref ID: 43085]

ORAUT (Oak Ridge Associated Universities Team), 2005a, *Internal Dose Overestimates for Facilities with Air Sampling Programs*, ORAUT-OTIB-0018, Rev. 01, Oak Ridge, Tennessee, August 9.

ORAUT (Oak Ridge Associated Universities Team), 2005b, *Savannah River Site – Occupational Internal Dose*, ORAUT-TKBS-0003, Rev. 03, Oak Ridge, Tennessee, April 5.

ORAUT (Oak Ridge Associated Universities Team) 2005c, Public Outreach Meeting minutes, July 21.

ORAUT (Oak Ridge Associated Universities Team), 2005d, *Interpretation of Dosimetry Data for Assignment of Shallow Dose,* ORAUT-OTIB-0017, Rev. 01, Oak Ridge, Tennessee, October 11.

ORAUT (Oak Ridge Associated Universities Team), 2005e, *Dose Reconstruction from Occupationally Related Diagnostic X-Ray Procedures,* ORAUT-OTIB-0006, Rev. 03 PC-1, Oak Ridge, Tennessee, December 21.

ORAUT (Oak Ridge Associated Universities Team), 2005f, *Analysis of Coworker Bioassay Data for Internal Dose Assignment,* ORAUT-OTIB-0019, Rev. 01, Oak Ridge, Tennessee, October 7.

ORAUT (Oak Ridge Associated Universities Team), 2006a, *Estimating the Maximum Plausible Dose to Workers at Atomic Weapons Employer Facilities,* ORAUT-OTIB-0004, Rev. 03 PC-2, Oak Ridge, Tennessee, December 6.

ORAUT (Oak Ridge Associated Universities Team), 2006b, *External Dose Reconstruction*, ORAUT-PROC-0006, Rev. 01, Oak Ridge, Tennessee, June 5.

ORAUT (Oak Ridge Associated Universities Team), 2006c, *A Standard Complex-Wide Methodology for Overestimating External Doses Measured with Film Badge Dosimeters*, ORAUT-OTIB-0010, Rev. 01, Oak Ridge, Tennessee, June 5.

ORAUT (Oak Ridge Associated Universities Team), 2007a, *Internal Dose Reconstruction*, ORAUT-OTIB-0060, Rev. 00, Oak Ridge, Tennessee, February 6.

ORAUT (Oak Ridge Associated Universities Team), 2007b, *Hanford Site – Occupational Internal Dose*, ORAUT-TKBS-0006-5, Rev. 02, Oak Ridge, Tennessee, June 22.

ORAUT (Oak Ridge Associated Universities Team), 2007c, *Estimating Doses for Plutonium Strongly Retained in the Lung*, ORAUT-OTIB-0049, Rev. 01, Oak Ridge, Tennessee, December 18.

ORAUT (Oak Ridge Associated Universities Team), 2008, *Dose Reconstruction during Residual Radioactivity Periods at Atomic Weapons Employer Facilities*, ORAUT-OTIB-0070, Rev. 00, Oak Ridge, Tennessee, March 10.

Runion, T.C., 1959, Davison Chemical Company, memorandum to J.O. Clarke, US Atomic Energy Commission, July 28. [SRDB Ref ID: 11920]

Sharma, A. R., D. Chandra, S. Chaturvedi, S. Ganesan, and P. K. Kaw, 2002, *Integral Validation and Use of Improved Nuclear Data in Fusion Blanket Studies, Final Report*, Institute for Plasma Research, Gujrat, India, February. [SRDB Ref ID: 18706]

Tester, M., 2005a, "OCAS conference call," documented communication with NFS, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, August 11. [SRDB Ref ID: 18803]

Tester, M., 2005b, "Radiation Exposure Summary 1977-1979," documents received from NFS/W.R. Grace Safety Department, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, October 24. [SRDB Ref ID: 19315]

Tester, M., 2005c, "Radiation Exposure Summary 1980-1989," documents received from NFS/W.R. Grace Safety Department, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, October 24. [SRDB Ref ID: 19316]

Tester, M., 2005d, "Radiation Exposure Summary 1990-1999", documents received from NFS/W.R. Grace Safety Department, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, October 24. [SRDB Ref ID: 19317]

Tester, M., 2005e, "Radiation Exposure Summary 2000-2005", documents received from NFS/W.R. Grace Safety Department, Oak Ridge Associated Universities Team, Oak Ridge, Tennessee, October 24. [SRDB Ref ID: 19318]

Tomlinson, R. E., 1964, "Tentative U-233 and Thorium Nitrate Specifications," document received from Hanford Atomic Products Operation, Richland, Washington, December 18. [SRDB Ref ID: 20258]

West, C. M., 1965, *Health Physics Considerations Associated With Thorium Processing*, Y-KB-53, Union Carbide Corporation, Y-12 Plant, Oak Ridge, Tennessee, March 10. [SRDB Ref ID: 11596]

Wilson, R. H., J. J. Fix, W. V. Baumgartner, and L. L. Nichols, 1990, *Description and Evaluation of the Hanford Personnel Dosimeter Program from 1944 Through 1989,* PNL-7447, Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Washington, September. [SRDB Ref ID: 4793]

WNA (World Nuclear Association), 2005, *Information and Issue Briefs – Thorium*,. http://www.world-nuclear.org/info/inf62.htm, (accessed June 29, 2005). [SRDB Ref ID: 17596]

W.R. Grace, 1959, *Feasibility Report: Processing of Highly Enriched Scrap Material for the Recovery of Contained Uranium under Contract No. AT-30-1 2377*," Davidson Chemical Division, Erwin, Tennessee, May 11. [SDRB Ref ID: 11740]