



ORAU TEAM Dose Reconstruction Project for NIOSH

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New Total Rewrite Revision Page Change

FOR DOCUMENTS MARKED AS A TOTAL REWRITE, REVISION, OR PAGE CHANGE, REPLACE THE PRIOR REVISION AND DISCARD / DESTROY ALL COPIES OF THE PRIOR REVISION.

PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
02/14/2006	00	New technical basis document for the W.R. Grace and Company in Erwin, Tennessee. First approved issue. Initiated by Paul J. Demopoulos.
07/16/2008	01	Approved revision to incorporate changes made from Task 5 comments. Incorporates formal internal and NIOSH review comments. Incorporates SEC-00082 and a methodology for reconstruction dose during the residual period. Constitutes a total rewrite of the document. Training required: As determined by the Task Manager. Initiated by Karen S. Kent.
09/16/2011	02	This revision was initiated to modify the residual period guidance. This includes a recommendation to not assign plutonium and thorium exposures during the residual period; modified the methodology for assigning external dose and uranium exposure during the residual period; and revised the use of monitoring records during the residual period. Also, an error was corrected for ²³⁸ U in Table 3-3. In addition, references were updated as appropriate and various minor technical edits were made throughout this document. The SEC language in Section 1.0 was moved to a newly created Sections 1.3. These changes were made to Sections 3.1, 3.1.3, 4.7, 5.1, 5.2, and the references. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Karen S. Kent.

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ACRONYMS AND ABBREVIATIONS

ADU	ammonium diuranate
AEC	U.S. Atomic Energy Commission
AP	anterior-posterior
AWE	atomic weapons employer
Bq	becquerel
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
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
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
MOX	mixed oxide
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
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. § 7384I(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 2010):

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

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 2010). 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.

This site profile provides specific information on documentation of historical practices at the W.R. Grace and Company plant.

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 medical X-ray examinations performed for screening. 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 describe 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 U.S. Atomic Energy Commission (AEC) contract years from the late 1950s through 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 were collected in the 1960s 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 used Nuclear Chicago Company and then Landauer, Inc., 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.

1.3 SPECIAL EXPOSURE COHORT PETITION INFORMATION FOR W. R. GRACE

January 1, 1958 through December 31, 1970

NIOSH has determined, and the Secretary of the U.S. Department of Health and Human Services (DHHS) has concurred, that it is not feasible to reconstruct internal radiation dose for (Leavitt 2007):

Atomic Weapons Employer (AWE) employees who were monitored or should have been monitored for potential exposure to thorium while working in any of the 100 series buildings or Buildings 220, 230, 233, 234, 301, or 310 at the W. R. Grace site at Erwin, Tennessee for a number of work days aggregating at least 250 work days from January 1, 1958, through December 31, 1970, or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort.

Therefore, only internal dose from uranium and plutonium is considered in Section 3.0 of this site profile. Because of the identified dose reconstruction infeasibility, all dose reconstructions for all workers having employment during the SEC class period are considered partial dose reconstructions. If monitoring data are available for workers included in the SEC class, this data may be used to refine the dose reconstruction; however, such dose reconstructions are still considered partial dose reconstructions.

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 States 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 in 1986 (Congress 1986, p.108).

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

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



Figure 2-1. Location of Erwin, Tennessee.

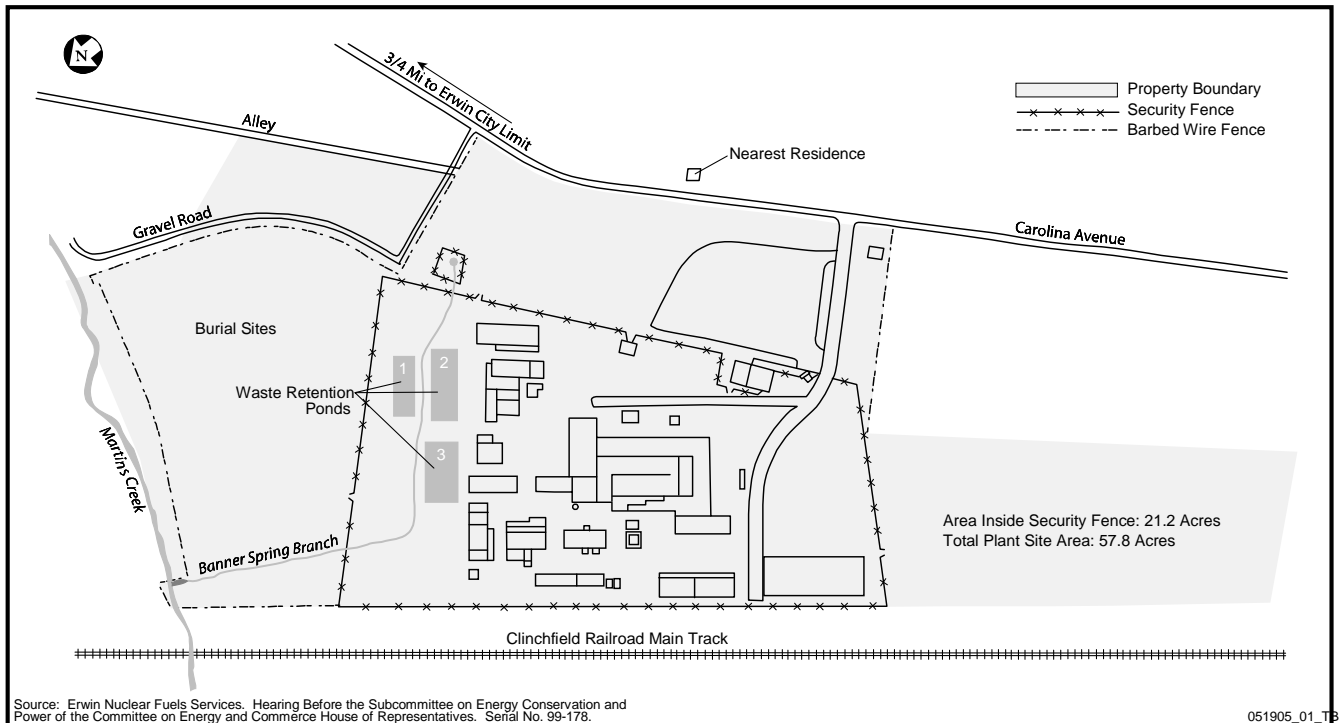


Figure 2-2. Plant site at W.R. Grace/NFS.

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 a series of Feasibility Reports (W.R. Grace 1959; Glauberman 1962; Housholder and Runion 1962; Housholder 1963a,b). Inherent in all the operations is nuclear criticality safety that governs not only the operations and storage but also the movement of material in 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

Table 2-1. W.R. Grace and NFS AEC period and locations.^{a,b}

Operations	Period of AEC work	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	1/1/1960 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 and MOX	1/1/1966 through 12/31/1970	234A, 234B, 234C, 110C, 110D
HEU scrap recovery	1/1/1958 through 12/31/1970 ^c	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

- a. Sources: AEC (1961), Congress (1986, pp. 107-114 and 127-143).
- b. Listed activities include both weapons-related work and non-weapons-related work during the AEC contract period. However, dose from non-weapons-related work is only considered during the AEC contract years, 1958 through 1970.
- c. 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).

whenever possible. For birdcage units, the two control parameters are geometry (birdcage dimensions) and a ²³⁵U mass limit that can be placed in the birdcage. Examples of birdcages that were used include a ²³³U storage birdcage that used a shielded 55-gal drum that contained inserts with spacers; UF₆ cylinders storage birdcage that used a 2-in. x 2-in. frame of metal that held the 5-in. UF₆ cylinder in place at its center with a total dimension of 52 in. high and 30 in. wide; and a U₃O₈ storage birdcage that used a 2-in. x 2-in. metal frame that held a 10.75-in. outside diameter x 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.

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

The UF₆ 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 UF₄, 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% ²³⁵U* (Housholder 1963a, pp. 6-8).

UF₆ was received in solid cylinders and had to be vaporized to transfer it to the UF₆-to-UF₄ reactor (the 6-to-4 unit). Vaporization was accomplished by heating the UF₆ cylinders in an electric oven. As many as six cylinders could be placed in the oven for processing.

The UF₆ was piped into the 6-to-4 unit where it was reduced with hydrogen. The solid UF₄ powder dropped into a product hopper where it was metered into safe-diameter product cans. The offgas flowed through two cyclone separators where entrained UF₄ dust was collected in additional product cans. The offgas was then filtered to remove the last traces of UF₄ dust and 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, derbies were broken into smaller pieces on a large hydraulic press before shipment. On other occasions, derbies were remelted and cast into various shapes such as slugs, rods, and plates. In addition, the uranium could be alloyed with other metals during remelting and casting. Melting occurred in a large vacuum induction furnace. Cast pieces could be pickled as described above.

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 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 g of ^{235}U per liter. There were solutions ranging from 2.8 to 9.9% enrichment at a concentration of 5 g of ^{235}U per liter. From these two types of solutions, a third solution was formed that did not exceed 3% enrichment, and contained a maximum of 88 lb of uranium (39.9 kg) 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 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, pp. 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, 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 apparently 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, pp. 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 ($^{233}\text{UO}_2/\text{ThO}_2$ and $^{233}\text{UO}_2/\text{ZrO}_2$) was in operation from approximately 1961 to 1969 for the light-water breeder reactor fuel for the Shippingport Reactor. This process was more involved than other process lines with 11 separate steps to produce the ^{233}U fuel (Housholder 1963b, pp. 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 in 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 $^{233}\text{UO}_2/\text{ThO}_2$ and $^{233}\text{UO}_2/\text{ZrO}_2$ 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 in 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 enrichments and chemical forms of the processed radionuclides.

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 ₂ F ₂ , & UO ₂ (NO ₃) ₂ (F) UO ₃ & UF ₄ (M) U ₃ O 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 ²³³U fuel composition.

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, 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 to which a worker could have been exposed versus another. 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).

International Commission on Radiological Protection (ICRP) Publication 68 (1995) lists UF₆, UO₂F₂, and UO₂(NO₃)₂ (uranyl nitrate) as type F; UF₄ and UO₃ as type M; and U₃O₈ and UO₂ 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 (0.72%)	IMBA	0.683	0.489	0.023	-	0.489
93.5%	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
DU (0.2%)	IMBA	0.402	0.155	0.011	0.0005	0.834

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 ²³³U 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. However, if only chest count data is available, then ²³³U fuel mixture should not be used since chest counts would not be used to detect the ²³³U fuel mixture given the small abundance of ²³⁵U. In this case the most favorable to claimant mixture (between DU, NU, LEU, and HEU) should be used.

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

Isotope	Mass (g)	Specific activity (Bq/g)	Total activity	% activity	% weight
U-232	3.7795	7.93E+11	3.00E+12	1.63	0
U-233	500,600.60	3.57E+08	1.79E+14	97.5	2
U-234	6,547.90	2.31E+08	1.51E+12	0.82	0
U-235	440.90	8.01E+04	3.53E+07	0.00	0
U-236	112.80	2.40E+06	2.70E+08	0.00	0
U-238	1,900.60	1.25E+04	2.38E+04	0.00	0
Th-232	23,481,000.00	4.06E+03	9.54E+10	0.05	98
Total	23,990,606.58		1.84E+14		100

3.1.1 Uranium Air Sampling

This discussion focuses on documented air-sampling data from two separate Health and Safety Laboratory 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, pp. 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.

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

Operation	Number of samples	Average concentration (dpm/m ³)
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 laboratory	3	3
Spectrographic laboratory	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.

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 glovebox and transferring U ₃ O ₈	4	97
Micropulverizing U ₃ O 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.

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 ₃ O ₈ transfer hood area	3	11
Micropulverizer area	4	62
Accountability room	5	21
Chemistry laboratory	4	4.3
Spectrographic laboratory	2	9.3
Research and development laboratory	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.

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) 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/m³, a geometric standard deviation (GSD) of 2.12, and a 95th-percentile value of 578 dpm/m³. 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.

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 m³/hr, as indicated in ICRP Publication 66 (ICRP 1994). This category assumes 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/m³) 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 ²³³U has also been identified. Airborne ²³³U concentrations for enriched UO₂ decladding and dissolution of ²³³U₃O₈ 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.

For the case in which 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/m³ 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	6.24E+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	1.30E+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 ²³⁴U 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 ²³³U and as such should be applied regardless of whether ²³³U or ²³⁴U is assumed (Tomlinson 1964, p. 6).

3.1.3 Uranium Bioassay

Urinalysis for uranium started in approximately October 1964. Samples were sent to Eberline Services 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

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).

indicated from the review of employee documents from the NFS Health Physics department (Tester 2005, 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 less than 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, the dose reconstructor should assume the MDL is 10 dpm/L [2]. If bioassay data are available, dose reconstructors should use them 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 ²³⁵U was about 120 µg (Tester 2005, p. 2).

A summary of the chest count conversions is shown in Table 3-13.

Table 3-13. Chest count conversions for uranium source terms.

Mixture	Detected isotope	U-235 MDA (ug)	pCi total U per ug U-235 ^b	Total U MDA (pCi)	dpm total U per ug U-235 ^b	Total U MDA (dpm)
Uranium-depleted	U-235	120	201	24,120	446	53,546
Uranium-natural	U-235	120	95	11,377	210	25,257
Uranium-2% enriched	U-235	120	81	9,696	179	21,525
Uranium-3.5% enriched	U-235	120	63	7,543	140	16,745
Uranium-93% enriched	U-235	120	73	8,787	163	19,507
Uranium-233 fuel mixture	U-235	120	1.18E+07	1.41E+09 ^a	2.62E+07	3.14E+09 ^a

a. If only chest count measurements exist in a claimant's records, then U-235 chest count measurements should not be used to assess U-233 fuel mixture intakes. Due to the low U-235 content in the U-233 fuel mixture, the site would not have used U-235 chest counting to detect U-233 intakes.

b. These specific activities can also be used to convert chest counts results reported in U-235 mass to total uranium activity.

3.2 PLUTONIUM EXPOSURES

Many forms of plutonium were possible over the years including metal and oxide. The specific chemical forms of plutonium at the WR Grace site are not known. It is possible that MOX fuel of a plutonium and thorium mixture was processed at some point because UO₂ and ThO₂ 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 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* (ORAUT 2010a).

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 considered reasonable and should be used in the absence of claimant-specific information [3]. A summary of the activity composition of the default reference fuel-grade plutonium mixture is presented in Table 3-14.

Table 3-14. 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 (2010b)

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 ²³⁹Pu was analyzed in urine from 1967 to approximately 1973 [4]. The years of plutonium exposures most likely occurred from 1967 through 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; 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 site profile for the Hanford Site (ORAUT 2010b) reports an MDA of 0.05 dpm/sample. The site profile for the Savannah River Site (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, an assumed DL of 0.03 dpm/L and an MDA of 0.06 dpm/L should be used [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 non-detections or perhaps near the MDA.

3.3 URANIUM-233 CONSIDERATIONS

Thorium oxide as ThO_2 and uranium oxide as $^{233}\text{UO}_2$ were the finished products for the production of $^{233}\text{U}/^{232}\text{Th}/^{228}\text{Th}$ MOX fuel. The ^{233}U was received from Oak Ridge in the form of uranyl nitrate, precipitated to $(\text{NH}_4)_2\text{U}_2\text{O}_7$ (ADU), then calcined to UO_2 . Blending, pressing, and grinding were parts 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 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. 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 - $^{233}\text{UO}_2$ pellet Feasibility Report mentioned 93% enrichment (Housholder 1963b). Isotopic composition of $^{232}\text{Th}/^{233}\text{U}$ 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 reconstruct internal thorium dose adequately 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 during the operational period (1958-1970) should be based on actual bioassay monitoring results for the individual. When uranium bioassay results are not available, the values in Table 3-15 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 a partial internal assessment.

The assumed occupational exposure period ran from January 1, 1958, to December 31, 1970. Table 3-15 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/m³), from Table 3-8, a breathing rate of 1.2 m³/hr (ICRP 1994), and an annual occupational exposure of 2,000 hours. The intake mode is chronic.

Table 3-15. Uranium internal exposure summary for operational period January 1, 1958, to December 31, 1970.^a

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

- 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.

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 UF₆ 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.

4.2 RECORDED EXTERNAL DOSE

Beginning in the late 1950s, Nuclear Chicago 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). In the approximate 1989 timeframe, Landauer supplied W.R. Grace/NFS with TLDs rather than 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 non-penetrating and penetrating dose components. Before 1970, the reported skin dose represented only the non-penetrating dose component. As such, before, the total shallow dose is best estimated by summing the reported skin (i.e., non-penetrating dose) and whole-body (i.e., penetrating dose component) doses.

Table 4-1 lists a more detailed description of the Nuclear Chicago Company or Landauer, Inc., dosimeters used at W.R. Grace and, later, NFS. The 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 <i>whole body</i>	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 <i>whole body</i>	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 <i>whole body</i>	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 <i>whole body</i>	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 <i>wrist</i>	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 <i>wrist</i>	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 <i>wrist</i>	K5 TLD wrist badge	3 TLD-100 chips.
1958–1974	Film badge - <i>finger</i>	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 <i>finger ring</i>	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 types 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 before 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 Non-Penetrating Dose

Dosimeters used at W.R. Grace would measure beta and low-energy photon non-penetrating dose. Before 1970, the reported skin dose represents only the non-penetrating 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 the Hanford Site (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 low in comparison with non-penetrating 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 HEU 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.

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.

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

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

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

4.4 MISSED DOSE

MDL information for photon and beta whole-body dosimeter systems is 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 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, if applicable, based on the location of the cancer and the workplace practices. Non-penetrating dose can be considered for IREP input categories as either electrons greater than 15 keV or photons <30 keV, depending on which selection is more reasonable based on claim information and favorability to the claimant. Typically, non-penetrating 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 non-penetrating 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 non-penetrating 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 $\pm 30\%$ (ORAUT 2006b). Dose reconstructors should use the Exposure (R) to Organ Dose (HT), [8] dose conversion factors from NIOSH for employment during AEC operations, (e.g. January 1, 1958, through December 31, 1970) (2007a). To ensure favorability to claimants, a photon energy range of 100% 30-to-250 keV should be applied (ORAUT 2006c).

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 DCAS-TIB-0010, *Special External Dose Reconstruction Considerations for Glovebox Workers* (DCAS 2010a). This would include chemical operators. Indicators of hands-on plutonium work could be plutonium bioassay results 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 m (Householder 1963b, pp. 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 Oak Ridge Associated Universities (ORAU) Team guidance, which is a posterior-anterior (PA) radiographic chest examination [9]. Preemployment and termination PA chest X-rays are assumed for all site workers based on information from the *Technical Information Bulletin: Guidance on Assigning Occupational X-Ray Dose under EEOICPA for X-Rays Administered Off Site* (ORAUT 2011c). Organ doses can be obtained from *Dose Reconstruction from Occupationally Related Diagnostic X-Ray Procedures* (ORAUT 2011b). 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 in the above sections to determine 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
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
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
Measured dose	1/1/1958–12/31/1970	Photons, 30-250 keV, AP, acute	Penetrating	Recorded value	recorded dose	
Measured dose	1/1/1958–12/31/1970	Photons, <30 keV or electrons >15 keV, AP, acute	Non-penetrating	Recorded value	recorded dose	
Medical X-ray	1/1/1958–12/31/1970	PA radiographic chest exam	Penetrating	Initial plus annual examinations	See Section 4.4.5	See ORAUT (2011b)

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 October 2009 *Report on Residual Radioactive and Beryllium Contamination at Atomic Weapons Employer Facilities and Beryllium Vendor Facilities*, noted that a significant potential for residual contamination remained after 1970. The residual contamination period at W.R. Grace is from January 1, 1971, through October 2009 (NIOSH 2009).

Remediation of the Burial Grounds

Remediation activities of the burial grounds occurred during the residual period at WR Grace. The North site area was used in the past for waste storage and disposal activities related to its nuclear work. NFS has excavated and removed buried wastes and debris. Excavated wastes, debris, and contaminated soils have been packaged for shipment to and disposal at an off-site licensed disposal facility. Three surface impoundments, Ponds 1, 2, and 3 are located within the North site. These impoundments received liquid waste from on-site processing operations from 1957 until 1978. Also low-level, contaminated solid wastes were disposed of in the North site Burial Ground area from 1966 until 1977, as authorized under 10 CFR 20.302. The contents and locations of most disposal pits are well documented. Another area previously used for solid waste disposal is the former Pond 4 area which is located west of the three impoundments. NFS removed waste materials from Ponds 1, 2, and 3 and the Pond 4 area from 1991 through 1996. NFS has also excavated waste and contaminated soil from the North site Burial Ground. Each former disposal area at the site has been identified as a Solid Waste Management Unit (SWMU) in the Hazardous and Solid Waste Amendments (HSWA) permit issued to NFS by the Environmental Protection Agency (EPA 1993a).

Based on this information, these remediation activities are assumed to have occurred between 1991 and 2009 (MACTEC 2009). The residual approach discussed below is to be applied to those workers who had the potential for internal radiation exposure, since they could have worked in the Burial Grounds and Ponds Area. This is a favorable to claimant assumption.

5.1 INTERNAL EXPOSURE TO RESIDUAL ACTIVITY

During the residual radioactivity period at W.R. Grace (1971 through October 2009), personnel might have been internally exposed to uranium in the workplace. Internal monitoring data could exist for plutonium or thorium during the residual period; however, only operations involving uranium were part of the nuclear weapons-related program, and are covered during the residual period for dose reconstruction under EEOICPA (Leiton 2011).

The following information provides a method for estimating exposures during the residual radiation period due to uranium contamination.

Unless the potential for exposure can be ruled out, the default intakes below should be assigned. However, monitoring data can be used instead of the default intake assumptions to limit dose, as appropriate (NIOSH 2008a). These internal exposures can be limited using bioassay data for uranium, if they exist. Internal doses assessed from bioassay data during this period should be evaluated in accordance with the guidance in Section 3.0 for workers with the potential for internal radiation exposure and the depletion factors applied using the values in Table 5-1 of this site profile document.

The uranium intakes (and associated RU components) in Section 3.0 (Table 3-15) can 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 m³/d).

An average daily air concentration is used to estimate the residual surface concentration with guidance from ORAUT (2008a). 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:

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

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 also to depletion (ORAUT 2008a). NIOSH has previously determined the activity removal rates during the residual radiation periods for the four sites as summarized in Table 5-1 (DCAS-TKBS-0002 (DCAS 2010b); Battelle-TBD-6000 Appendix C (NIOSH 2008b); ORAUT-TKBS-0045 (ORAUT 2008b); ORAUT-TKBS-0032 (ORAUT 2011a). In these cases, contemporary estimates of airborne radioactivity at the beginning and end of each site's residual radiation period were used to estimate the effective exponential clearance of the contamination over an extended period of time for each site (ORAUT-OTIB-0070 (ORAUT 2008a)).

The source-term depletion rates calculated for these sites are provided in Table 5-1.

Table 5-1. Source-term depletion rates during residual period.

Facility	Depletion rate (per day)
Blockson	0.00076
Dow Madison	0.00027
General Atomics	0.00116
Simonds Saw and Steel	0.00049
Average	0.00067

Because these values were empirically derived at operating facilities, NIOSH considers them more appropriate than a value based solely on theoretical considerations. As such, the average value of 0.067% per day, as calculated using the data in Table 5-1, will be used to determine source term depletion effects. The intakes in Table 5-2 and Table 5-3 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. The internal dose analysis should include the potential inadvertent ingestion of uranium activity and be based on guidance in NIOSH (2004).

Table 5-2. Uranium (and RU components) annual inhalation rates for the residual period.

Year	Inhalation (pCi/day)						
	U234	Pu238	Np237	Tc-99	Th232	Th228	Ru-106
1971	1.71E+03	1.03E+02	8.55E+00	6.84E+02	3.42E+01	3.42E+01	6.84E+01
1972	1.34E+03	8.03E+01	6.70E+00	5.36E+02	2.68E+01	2.68E+01	5.36E+01
1973	1.05E+03	6.29E+01	5.24E+00	4.19E+02	2.10E+01	2.10E+01	4.19E+01
1974	8.21E+02	4.93E+01	4.11E+00	3.28E+02	1.64E+01	1.64E+01	3.28E+01
1975	6.43E+02	3.86E+01	3.21E+00	2.57E+02	1.29E+01	1.29E+01	2.57E+01
1976	5.03E+02	3.02E+01	2.52E+00	2.01E+02	1.01E+01	1.01E+01	2.01E+01
1977	3.94E+02	2.37E+01	1.97E+00	1.58E+02	7.88E+00	7.88E+00	1.58E+01
1978	3.09E+02	1.85E+01	1.54E+00	1.23E+02	6.17E+00	6.17E+00	1.23E+01
1979	2.42E+02	1.45E+01	1.21E+00	9.67E+01	4.83E+00	4.83E+00	9.67E+00
1980	1.89E+02	1.14E+01	9.46E-01	7.57E+01	3.79E+00	3.79E+00	7.57E+00
1981	1.48E+02	8.89E+00	7.41E-01	5.93E+01	2.96E+00	2.96E+00	5.93E+00
1982	1.16E+02	6.96E+00	5.80E-01	4.64E+01	2.32E+00	2.32E+00	4.64E+00
1983	9.09E+01	5.45E+00	4.54E-01	3.64E+01	1.82E+00	1.82E+00	3.64E+00
1984	7.12E+01	4.27E+00	3.56E-01	2.85E+01	1.42E+00	1.42E+00	2.85E+00
1985	5.57E+01	3.34E+00	2.79E-01	2.23E+01	1.11E+00	1.11E+00	2.23E+00
1986	4.36E+01	2.62E+00	2.18E-01	1.75E+01	8.73E-01	8.73E-01	1.75E+00
1987	3.42E+01	2.05E+00	1.71E-01	1.37E+01	6.83E-01	6.83E-01	1.37E+00
1988	2.68E+01	1.61E+00	1.34E-01	1.07E+01	5.35E-01	5.35E-01	1.07E+00
1989	2.10E+01	1.26E+00	1.05E-01	8.38E+00	4.19E-01	4.19E-01	8.38E-01
1990	1.64E+01	9.85E-01	8.20E-02	6.56E+00	3.28E-01	3.28E-01	6.56E-01
1991	1.28E+01	7.71E-01	6.42E-02	5.14E+00	2.57E-01	2.57E-01	5.14E-01
1992	1.01E+01	6.04E-01	5.03E-02	4.02E+00	2.01E-01	2.01E-01	4.02E-01
1993	7.88E+00	4.73E-01	3.94E-02	3.15E+00	1.58E-01	1.58E-01	3.15E-01
1994	6.17E+00	3.70E-01	3.08E-02	2.47E+00	1.23E-01	1.23E-01	2.47E-01
1995	4.83E+00	2.90E-01	2.42E-02	1.93E+00	9.66E-02	9.66E-02	1.93E-01
1996	3.78E+00	2.27E-01	1.89E-02	1.51E+00	7.57E-02	7.57E-02	1.51E-01
1997	2.96E+00	1.78E-01	1.48E-02	1.18E+00	5.92E-02	5.92E-02	1.18E-01
1998	2.32E+00	1.39E-01	1.16E-02	9.28E-01	4.64E-02	4.64E-02	9.28E-02
1999	1.82E+00	1.09E-01	9.08E-03	7.27E-01	3.63E-02	3.63E-02	7.27E-02
2000	1.42E+00	8.53E-02	7.11E-03	5.69E-01	2.84E-02	2.84E-02	5.69E-02
2001 on	1.11E+00	6.68E-02	5.57E-03	4.46E-01	2.23E-02	2.23E-02	4.46E-02

Table 5-3. Uranium (and RU components) annual ingestion rates for the residual period.

Year	Ingestion (pCi/day)						
	U234	Pu238	Np237	Tc-99	Th232	Th228	Ru-106
1971	3.56E+01	2.14E+00	1.78E-01	1.42E+01	7.12E-01	7.12E-01	1.42E+00
1972	2.79E+01	1.67E+00	1.39E-01	1.12E+01	5.58E-01	5.58E-01	1.12E+00
1973	2.18E+01	1.31E+00	1.09E-01	8.73E+00	4.37E-01	4.37E-01	8.73E-01
1974	1.71E+01	1.03E+00	8.55E-02	6.84E+00	3.42E-01	3.42E-01	6.84E-01
1975	1.34E+01	8.03E-01	6.69E-02	5.35E+00	2.68E-01	2.68E-01	5.35E-01
1976	1.05E+01	6.29E-01	5.24E-02	4.19E+00	2.10E-01	2.10E-01	4.19E-01
1977	8.21E+00	4.92E-01	4.10E-02	3.28E+00	1.64E-01	1.64E-01	3.28E-01
1978	6.43E+00	3.86E-01	3.21E-02	2.57E+00	1.29E-01	1.29E-01	2.57E-01
1979	5.03E+00	3.02E-01	2.52E-02	2.01E+00	1.01E-01	1.01E-01	2.01E-01
1980	3.94E+00	2.36E-01	1.97E-02	1.58E+00	7.88E-02	7.88E-02	1.58E-01
1981	3.09E+00	1.85E-01	1.54E-02	1.23E+00	6.17E-02	6.17E-02	1.23E-01
1982	2.42E+00	1.45E-01	1.21E-02	9.67E-01	4.83E-02	4.83E-02	9.67E-02
1983	1.89E+00	1.14E-01	9.46E-03	7.57E-01	3.78E-02	3.78E-02	7.57E-02
1984	1.48E+00	8.89E-02	7.41E-03	5.93E-01	2.96E-02	2.96E-02	5.93E-02
1985	1.16E+00	6.96E-02	5.80E-03	4.64E-01	2.32E-02	2.32E-02	4.64E-02
1986	9.09E-01	5.45E-02	4.54E-03	3.63E-01	1.82E-02	1.82E-02	3.63E-02
1987	7.11E-01	4.27E-02	3.56E-03	2.85E-01	1.42E-02	1.42E-02	2.85E-02
1988	5.57E-01	3.34E-02	2.79E-03	2.23E-01	1.11E-02	1.11E-02	2.23E-02
1989	4.36E-01	2.62E-02	2.18E-03	1.74E-01	8.72E-03	8.72E-03	1.74E-02
1990	3.42E-01	2.05E-02	1.71E-03	1.37E-01	6.83E-03	6.83E-03	1.37E-02
1991	2.67E-01	1.60E-02	1.34E-03	1.07E-01	5.35E-03	5.35E-03	1.07E-02
1992	2.09E-01	1.26E-02	1.05E-03	8.38E-02	4.19E-03	4.19E-03	8.38E-03
1993	1.64E-01	9.84E-03	8.20E-04	6.56E-02	3.28E-03	3.28E-03	6.56E-03
1994	1.28E-01	7.71E-03	6.42E-04	5.14E-02	2.57E-03	2.57E-03	5.14E-03
1995	1.01E-01	6.03E-03	5.03E-04	4.02E-02	2.01E-03	2.01E-03	4.02E-03
1996	7.88E-02	4.73E-03	3.94E-04	3.15E-02	1.58E-03	1.58E-03	3.15E-03
1997	6.17E-02	3.70E-03	3.08E-04	2.47E-02	1.23E-03	1.23E-03	2.47E-03
1998	4.83E-02	2.90E-03	2.41E-04	1.93E-02	9.66E-04	9.66E-04	1.93E-03
1999	3.78E-02	2.27E-03	1.89E-04	1.51E-02	7.56E-04	7.56E-04	1.51E-03
2000	2.96E-02	1.78E-03	1.48E-04	1.18E-02	5.92E-04	5.92E-04	1.18E-03
2001 on	2.32E-02	1.39E-03	1.16E-04	9.27E-03	4.64E-04	4.64E-04	9.27E-04

The daily ingestion rate in activity per day is estimated as 0.2 times the average daily air concentration expressed in units of activity/m³ (NIOSH 2004). The annual ingestion rates for the residual period are shown in Table 5-3.

Remediation of the Burial Grounds

Remediation activities of the burial grounds should be assessed based on individual bioassay records per guidance in Section 3.

5.2 ESTIMATION OF EXTERNAL EXPOSURE FROM RESIDUAL RADIOACTIVITY

The default exposure rates below should be assigned, during the applicable years. External dosimetry data might exist during the residual period; however, operations were not part of the nuclear weapons-related program, and are not covered for dose reconstruction under EEOICPA (Leiton 2011). However, these monitoring data can be used to limit dose, as appropriate (NIOSH 2008a).

In the absence of claim-specific information, the following assumptions in Table 5-4 should be made about external exposures [11]:

Table 5-4. External default assumptions during the residual period.

Residual period	Sources of external exposure	Default assumptions
1971–Present (Applicable to all workers pre-1991 and all workers 1991 on with no monitoring data)	Photons, 30–250 keV	Ambient H * (10) to organ dose (HT)
	Electrons, > 15 keV or <30 keV	Follow ORAUT (2005d) requirements
1991–2009 (Applicable to Burial Grounds Remediation Workers with monitoring data)	Photons, 30–250 keV	<i>Deep Dose Equivalent (Hp(10)) to Organ Dose (HT)</i>
	Electrons, > 15 keV or <30 keV	Follow ORAUT (2005d) requirements

The 95th-percentile value of the maximum average breathing-zone air concentration for the 1961 AEC air sample results (from Table 3-8) was calculated using Crystal Ball and assuming a lognormal distribution calculation yielded a distribution with a GM of 167 dpm/m³ and a GSD of 2.12. This resulted in a 95th-percentile value of 578 dpm/m³.

It is assumed that the material was deposited on the floor with a deposition velocity of 0.00075 m/s for a period of 1 year without cleanup. This would result in a contamination level of 1.37×10^7 dpm/m². External dose rates were calculated for exposure to contaminated surfaces and submersion from resuspended materials based on the DCFs in Federal Guidance Report No. 12 (EPA 1993).

The external doses are from penetrating photons with energies between 30 keV and 250 keV and >15 keV for penetrating exposures. NIOSH has previously determined the activity removal rates during the residual radiation periods for the four sites as summarized in Table 5-1 (DCAS-TKBS-0002 (DCAS 2010b); Battelle-TBD-6000 Appendix C (NIOSH 2008b); ORAUT-TKBS-0045 (ORAUT 2008b); ORAUT-TKBS-0032 (ORAUT 2011a)). In these cases, contemporary estimates of airborne radioactivity at the beginning and end of each site's residual radiation period were used to estimate the effective exponential clearance of the contamination over an extended period of time for each site (ORAUT-OTIB-0070 (ORAUT 2008a)). The source-term depletion rates calculated for these sites are provided in Table 5-1. Because these values were empirically derived at operating facilities, NIOSH considers them more appropriate than a value based solely on theoretical considerations. As such, the average value of 0.067% per day, as calculated using the data in Table 5-1, will be used to determine source term depletion effects.

Table 5-5 provides the external exposure rates for the residual period using the average source term depletion value.

Table 5-5. External annual exposure rates for the residual period.

Year	External (rem/year)	
	Pen	Non-Pen
1971	0.007	1.444
1972	0.005	1.131
1973	0.004	0.885
1974	0.003	0.693
1975	0.003	0.543
1976	0.002	0.425
1977	0.002	0.333
1978	0.001	0.261
1979	0.001	0.204
1980	0.001	0.160
1981	0.001	0.125
1982	0.001*	0.098
1983	0.001*	0.077
1984	0.001*	0.060
1985	0.001*	0.047
1986	0.001*	0.037
1987	0.001*	0.029
1988	0.001*	0.023
1989	0.001*	0.018
1990	0.001*	0.014
1991	0.001*	0.011
1992	0.001*	0.008
1993	0.001*	0.007
1994	0.001*	0.005
1995	0.001*	0.004
1996	0.001*	0.003
1997	0.001*	0.003
1998	0.001*	0.002
1999	0.001*	0.002
2000	0.001*	0.001
2001 - 2009	0.001*	0.001

*As a favorable to claimant assumption, values were rounded up to 0.001 rem per year.

Remediation of the Burial Grounds

Remediation activities of the burial grounds should be assessed based on individual monitoring records per guidance in Section 4 and Table 5-4.

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. Senior Health Physicist. 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, the dose reconstructor should assume the MDL is 10 dpm/L.
- [3] Sharfi, Mutty M. ORAU Team. Principal AWE Dosimetrist. June 2011.
An assumption of 10-year-old fuel-grade plutonium is considered reasonable and should be used in the absence of claimant-specific information.
- [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 an 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 that 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. 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, M., and Winslow, R.. ORAU Team. Senior Health Physicists. 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] Fix, J. J., Smith, M., and Winslow, R. ORAU Team. Senior Health Physicists. March 2008. Default DCFs from NIOSH (2007a) were identified for photons and electrons for the operational and residual periods.

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