

Internal Dosimetry Coworker Intake and Exposure Model for the W. R. Grace Company, Erwin, Tennessee

White Paper

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INTRODUCTION

A review by Sanford Cohen and Associates (SC&A) and Salient of the W. R. Grace, Erwin, Tennessee, site profile (ORAUT 2011) in February 2013 resulted in several findings, of which Finding 3 is associated with intakes of plutonium. SC&A noted the need for intakes for unmonitored workers and questioned why intakes of plutonium were not considered in dose reconstructions during the residual contamination period (SC&A and Salient 2013).

W. R. Grace was an Atomic Weapons Employer (AWE) facility from 1958 through 1970 with a residual contamination period from 1971 through March 1, 2011 (DOE 2017). The current site profile (ORAUT 2011) provides instructions to reconstruct plutonium intakes for workers with plutonium bioassay data during the AWE contract period. The site profile does not provide plutonium intakes for individuals with no internal monitoring results. In addition, the site profile states that plutonium intakes are not reconstructed for the residual contamination period.

The National Institute for Occupational Safety and Health (NIOSH) has reviewed the use of plutonium at W. R. Grace and now presumes dose from intakes of plutonium is covered under the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA) in both the AWE operational and residual periods.

PURPOSE

Some employees were not monitored for potential intakes of plutonium, or the records of such monitoring are incomplete or unavailable. In such cases, data from monitored coworkers was used to develop intake rates to address potential internal exposures of radioactive material during operations. This white paper provides plutonium coworker intake rates for the period of plutonium production at the site. These intake rates may be used to fill in gaps in internal monitoring data or used for workers presumed to have been exposed but not monitored for intakes of plutonium. In addition, alpha airborne concentration results from plutonium areas were used to develop plutonium intake rates for the post-operations period.

The plutonium facilities were shut down in 1973. Decontamination and decommissioning (D&D) of the facilities occurred from 1990 through 1993. Individual bioassay data is presumed sufficient for all workers exposed to plutonium during the D&D period. This paper provides intake rates for 1974 through 1989 based on general area air sampling data; those intake rates are for incidental exposure to plutonium contamination for workers who entered the building but were not monitored for plutonium. Some workers were monitored for plutonium exposure during this period, and they are presumed to have been exposed to potentially higher levels of plutonium. A discussion of the plutonium bioassay program is provided in Attachment D.

BACKGROUND

A summary of plutonium activities at W. R. Grace is shown in Table 1.

Table 1. Timeline of plutonium activities at W. R. Grace.

Timeframe	Activity	Reference
1964-1965	Plutonium facilities were constructed in Buildings 110 and 234. This was also known as the Mixed Oxide (MOX) UO ₂ /PuO ₂ fuel fabrication facility.	NFS 1988, ca. 2000; EG&G 1994
Late 1960s	Additional plutonium facilities added for plutonium scrap dissolution.	EG&G 1994, PDF p. 389
1965-1972	Operation of the MOX facilities (Buildings 110 Lab and 234), Fabrication of MOX fuel elements discontinued in 1972.	NFS 1994; NRC 1991, PDF p. 27; Higinbotham 1994, PDF p .4
1973	MOX facility placed in safe standby.	Haskins 1995; EG&G 1994
1973-1985	Nuclear Fuel Services (NFS) unsuccessful in finding a disposal site for wastes that would be generated from decommissioning activities.	NFS 1988
04/1986	Contract signed which allowed NFS to ship transuranic (TRU) waste to U.S. Department of Energy (DOE)-Idaho Falls.	NFS 1988
06/1988- 09/1988	MOX facilities were characterized.	NFS 1994
01/1989- 07/1989	MOX facility safety systems upgrades/prepare for decommissioning.	NFS 1994
06/20/1989	U.S. Nuclear Regulatory Commission (NRC) approval of decommissioning plan.	NFS 1994; NFS and EkoTec 1990
06/28/1990	Completed Decontamination and Volume Reduction Facility and started cold testing.	NFS 1994
09/12/1990	MOX facility D&D began.	NFS 1994
1991	Building 110 preparation for final release.	NFS 1991
06/1993- 09/1993	Excavation of ~2,310 ft ³ TRU contaminated soil from under Area B of Building 234 (Wet Chem Cell) w/~1700 ft ³ remaining.	NFS 1994
10/1993- 12/1993	Final cleanup of facilities; contaminated soils under the Wet Cell remained for future decommissioning.	NFS 1994
As of 2009	Contaminated material remains (as of 2009) in the soil underground where the Building 234 Wet Cell used to exist. A large tent was placed over the building site.	NFS 2009

Plutonium operations began in 1965 and were permanently shut down in 1973 when the uranium oxide/plutonium oxide MOX fuel fabrication facility was placed into safe standby (Haskins 1995). Between 1965 and 1973, approximately 812 kg of plutonium were processed for essentially four customers, as shown in Tables 2 and 3. Table 2 illustrates the plutonium activity processed by project. Table 3 provides a listing of job orders involving plutonium. The largest order covered the manufacture of about 2,000 PuO₂-UO₂ MOX fuel rods for the Southwest

Experimental Fast Oxide Reactor (SEFOR). This was a joint undertaking of General Electric (GE), the U.S. Atomic Energy Commission (AEC), and several utility companies. The GE-SEFOR order and the DuPont/Savannah River Operations Office (SROO) order comprised 94% of the plutonium introduced to the facility (NFS 1988).

Table 2. Plutonium activity processed by project at W. R. Grace.

Project	Original kg Pu	Ci/g TRU	Total Curies
SEFOR	762 ^a	0.648	493,776
Halden	3	3.610	10,830
Consumers	23.5	1.470	34,545
West Valley	23.5	2.800	65,800
Total	812.0	Not applicable	604,951

Source: NFS 1988.

Note: Isotopes present: ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu and ²⁴¹Am.

a. Includes 16 kg of plutonium used on the DuPont project for which no isotopic data is available.

Table 3. Summary of plutonium processes at W. R. Grace.

Timeframe	Customer	Product	kg Pu	U (Mass)	Pu (Mass)	U Enrichment
1965-1966 ^a	DuPont/SROO	MOX fuel rods	16	99.7%	~0.315%	Depleted
1967-1971 ^a	SEFOR/GE/AEC	MOX; 2,000 fuel rods and scrap dissolution	746	80.0%	20.0%	Depleted
1972 ^b	Halden/NFS-Research Facilities Design (RFD)	MOX fuel rods	3	98.5% to 97.5%	1.5% to 2.5%	Depleted and 1.0% to 5.0%
1972-1973 ^b	Big Rock Point (Consumers)/NFS-RFD	MOX fuel assemblies	47	97.73%	Average 2.27%	Average 2.41%
Total	Not applicable	Not applicable	812.0	Not applicable	Not applicable	Not applicable

Sources: NFS 1988; 1989a, PDF p. 119.

a. AEC Programs (16 kg + 746 kg = 762 kg, which is 94% of the total plutonium processed).

b. NFS Programs (3 kg + 47 kg = 50 kg, which is 6% of the total plutonium processed).

The plutonium facilities at W. R. Grace were involved with the dissolution of plutonium metal and oxide; coprecipitation of uranium-plutonium; wet or dry blending of MOX powders; pellet production and inspection; rod loading, welding, and inspection; scrap dissolution (for which NFS added facilities in the late 1960s); and full laboratory service (NFS 1988).

Building 110 plutonium areas included: Area C (Wet Lab Chemistry) and Area D (Spectrographic Lab). Building 234 plutonium areas were: Area A (Batch Weigh), Area B

(Former ^{233}U Process), Area C (Pelleting), Area D (Fabrication), Area E (Lab), Area F (Office), Area G (Clean Change), Area H (Process Change), Area I (Material Unloading), Area M (Air Lock), Areas 67 and 68 (Condensate Stations), and the Wet Cell area (NFS 1988).

Equipment in the facilities was located primarily in gloveboxes or in a single limited-entry cell adjacent to the conversion area. In addition to gloveboxes, the plutonium plant contained equipment such as metal tanks; glass columns; pumps; mixing vessels; blenders; drying, conversion, and sintering furnaces; pellet press, cut-off machine and centerless grinder; outgassing equipment; inspection jigs; welders; leak test gear; liquid and air high-efficiency particulate filters; miscellaneous laboratory equipment; ventilation fans; wet scrubbers; and piping (NFS 1988).

The SEFOR project processed the greatest majority of the MOX fuel rods and scrap dissolution at W. R. Grace. This project also had the largest percentage of plutonium in its MOX. It is favorable to claimants to assume the SEFOR MOX as a default assumption.

The AEC projects account for 94% of the plutonium fuel W. R. Grace produced. Of that, the SEFOR project accounted for 98%, with the DuPont/SROO project accounting for the other 2%. No information is available on the MOX fuel produced for DuPont/SROO. The composition of the plutonium used in the SEFOR project was used for determination of the composition of the various plutonium radionuclides. The original order for SEFOR plutonium was prepared as plutonium nitrate solution by the Atlantic Richfield Hanford Company between November 1965 and December 1966. This nitrate solution was then shipped to NFS where the plutonium nitrate solution was blended with depleted uranium nitrate solution. The original order for plutonium from Hanford totaled 545 kg plutonium (Higinbotham 1994, PDF pp.32-33). Details of date of separation and preparation of the remaining source material are unknown.

NFS (1988, PDF p.10) provides an isotopic composition of the SEFOR project material type adjusted for radioactive decay for 1970 and 1988. The SEFOR material is also identified by a ^{240}Pu assay of ~8.3% and a Pu:U ratio of 1:4 (NFS 1989b, PDF p. 107). The SEFOR mixture is the default plutonium assumption, as shown in Table 4. The year of separation was selected at a time without americium activity. This resulted in an estimate that the 1970 material was aged 14.313 years. These assumptions were used to complete the remainder of Table 4. The resulting ^{238}Pu value is not identical to that specified in NFS 1988 (0.036%). The ^{238}Pu value is adjusted to increase the wt % sum to 100.

The SEFOR MOX is also assumed during the time of the DuPont/Savannah River project (which occurred before the SEFOR project), as the DuPont/Savannah River source material is unknown.

Table 4. Default plutonium assumptions, SEFOR MOX.

Material Age	²³⁸ Pu (wt %)	^{239/240} Pu (wt %)	²⁴¹ Pu (wt %)	²⁴² Pu (wt %)	²⁴¹ Am (wt %)	Depleted U (wt %)
Fresh (2 Weeks)	0.050	98.948	0.948	0.052	0.002	400
5 Year	0.048	98.951	0.747	0.052	0.202	400
10 Year	0.047	98.955	0.587	0.052	0.360	400
20 Year	0.043	98.965	0.363	0.052	0.577	400

Source: NFS 1988, PDF p.10.

Note: For Material Age, 1970 is assumed to be representative of 14.313-year-aged material. Therefore, because the production of the plutonium nitrate solution for use at the W. R. Grace site occurred around the 1965 to 1966 timeframe (Higinbotham 1994), the fresh and 5-year-aged ratios are not applicable to W. R. Grace. Therefore, the 10-year-aged ratios were applied from 1965 through 1974, and the 20-year-aged ratios were applied to all intakes starting in 1975.

BIOASSAY DATA

NIOSH does not have a database of bioassay data from the site, but the NIOSH-Division of Compensation Analysis Claims Tracking System (NOCTS) has a sufficient number of claims with plutonium bioassay data to perform a coworker intake analysis for the period of plutonium production at W. R. Grace from 1967 to 1973. The derived intake rates for 1967 were extended back to include January 1, 1965, through 1966, when plutonium startup operations began (NFS 1988).

Although some plutonium bioassay data are available for other years, they are insufficient for the statistical analysis needed for a coworker study. Average alpha airborne concentration data from 1976 through 1993 was used to develop intake rates for plutonium; these data are from routine general area air sampling in the plutonium facilities. These intake rates are applicable for workers entering the building for various reasons, but are not applicable for D&D activities or other work that might have disturbed contamination. For the period between the end of the coworker data in 1973 and the beginning of the airborne concentration data in 1976, guidance in ORAUT-OTIB-0070, *Dose Reconstruction during Residual Radioactivity Periods at Atomic Weapons Employer Facilities* (ORAUT 2012), was used to connect the intake rates between the two periods, using an exponential decay curve. The intake rates developed from the airborne concentration data ending in 1993 were then extended from 1994 through March 1, 2011.

One of the MOX Fuel Fabrication Facilities Decommissioning Project objectives was to restore the existing facilities and site to levels of contamination which would permit “unrestricted” use. In November 1992, the project work scope was modified to include the removal of significantly more uranium and plutonium material in contaminated soil beneath Area B of Building 234 than was originally envisioned. Area B, the Wet Chemistry Cell area, was used for nitric acid dissolution and mixing of plutonium and depleted or low-enriched uranium. Much of the soil was excavated from under the Wet Cell for several months in 1993 and packaged as TRU waste. However, some soil contaminated with plutonium and uranium at TRU levels remained beneath the Wet Cell area and in a cell below ground (NFS 2009), and removal was planned to be

conducted as part of plantwide decommissioning efforts (NFS 1994). Because contamination still exists, it follows that the derived intake rates ending in 1993 should be extended from 1994 through March 1, 2011.

COWORKER INTAKE RATES BASED ON BIOASSAY (1965-1973)

Attachment A, *Plutonium Coworker Statistical Analysis*, provides the detailed method and statistical analysis for the development of the plutonium coworker intake rates based on NOCTS data. Attachment B, *Plutonium Coworker Statistical Analysis Instructions*, provides the instructions for the analysis for the development of the plutonium coworker intake rates.

The recommended coworker intake rates are shown in Tables 5 and 6. Table 5 summarizes the plutonium intake rates that correspond to an intake of Type M materials. Table 6 summarizes the plutonium intake rates that correspond to an intake of Type S materials. Dose from plutonium Type Super S materials will be estimated according to the methods described in ORAUT-OTIB-0049, *Technical Information Bulletin: Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT 2010).

Table 5. Plutonium intake rates based on bioassay (Type M) (dpm/d).

Start	End	50 th Percentile	GSD	95 th Percentile
01/01/1965	12/31/1968	49.93	4.65	626.64
01/01/1969	12/31/1971	10.96	3.39	81.50
01/01/1972	12/31/1973	2.567	6.59	57.10

Note: A SEFOR PuO₂-UO₂ MOX should be assumed.

Table 6. Plutonium intake rates based on bioassay (Type S) (dpm/d).

Start	End	50 th Percentile	GSD	95 th Percentile
01/01/1965	12/31/1968	1,692	5.07	24,431
01/01/1969	12/31/1971	291.9	3.30	2,076
01/01/1972	12/31/1973	107.2	6.59	2,384

Note: A SEFOR PuO₂-UO₂ MOX should be assumed.

Plutonium coworker intakes should be assigned to workers who were not monitored via bioassay, or to fill in gaps in monitoring data, as specified in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2018a). Plutonium workers with a hands-on operations job categories should be assigned the 95th-percentile coworker intake rates as a constant distribution. Radiological support workers should be assigned the coworker intake rate geometric mean (GM) as a lognormal distribution with a geometric standard deviation (GSD). Supervisors should be assigned 50% of the radiological support worker intake rates assuming the same lognormal distribution and GSD. Administrative workers should be assigned 10% of supervisor intake rates assuming the same lognormal distribution and GSD.

EXPOSURE MODEL TO DEVELOP PLUTONIUM INTAKE RATES BASED ON AIRBORNE CONCENTRATION DATA (1976-1993)

Attachment C, *Exposure Model to Develop Plutonium Intakes based on Airborne Concentration Data*, provides the detailed method used to develop the plutonium intakes based on airborne concentration data.

Table 7 summarizes the recommended total gross alpha inhalation and ingestion intake rates. The material type (M, S, Super S) that is most favorable to the claimant should be assigned.

Table 7. Total alpha intake rates based on airborne concentration data (dpm/d).

Start	End	Inhalation 95 th Percentile	Ingestion 95 th Percentile
01/01/1976	12/31/1987	8.74	1.75
01/01/1988	12/31/1993	1.13	0.23
01/01/1994	03/01/2011	1.13	0.23

Although the airborne concentration data are actually reporting total alpha, treating them as total plutonium simplifies the calculations and results in an assessment favorable to the claimant. A SEFOR PuO₂-UO₂ MOX should be assumed. Ingestion intakes should also be assigned when airborne concentration data are used to assign inhalation intakes as described in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004).

For the post-plutonium production period, workers presumed to have exposure in plutonium buildings should be assigned the 95th percentile intake rates as a constant distribution. In addition, workers with plutonium bioassay data should be assigned intakes based on their data. All other workers who are not assumed to have been working in the plutonium buildings should receive only environmental intakes.

D&D workers should be evaluated for intakes of plutonium based on their bioassay measurements.

ORAUT-OTIB-0070 METHOD TO DEVELOP PLUTONIUM INTAKE RATES (1974-1975)

For the gap between the end of the coworker data in 1973 and the start of the airborne concentration data in 1976, guidance in ORAUT (2012) was used to derive exposure rates for 1974 and 1975 using an exponential decay curve fit between the intake rates in 1973 and 1976.

A decay correction rate was calculated between the midpoint of the 1973 derived coworker intake rates for plutonium Types S and M as Year 1 and the midpoint of the 1976 intake rates derived from airborne concentration data. This was done for the 50th and 95th percentiles. The

higher of the two GSDs was applied to be favorable to the claimant. However, it is favorable to the claimant to assume the 95th percentile intake rates.

Though the airborne concentration data are reporting total alpha, treating them as total plutonium simplifies the calculations and results in an assessment favorable to the claimant.

Table 8 summarizes the plutonium intake rates that correspond to an intake of Type M materials. Table 9 summarizes the plutonium intake rates that correspond to an intake of Type S materials.

Table 8. Plutonium intake rates (Type M) (dpm/d).

Start	End	Inhalation 95 th Percentile	Ingestion 95 th Percentile
01/01/1973	12/31/1973	57.1	11.4
01/01/1974	12/31/1974	30.6	6.11
01/01/1975	12/31/1975	16.4	3.27
01/01/1976	12/31/1976	8.74	1.75

Note: Super S intake rates are assumed to be equal to Type S.

Note: A SEFOR PuO₂-UO₂ MOX should be assumed.

During the post-plutonium production period, workers presumed to have exposure in plutonium buildings will be assigned the 95th percentile intake rates as a constant distribution. All other workers that are not assumed to be working in the plutonium buildings will receive environmental intakes.

Table 9. Plutonium intake rates (Type S) (dpm/d).

Start	End	Inhalation 95 th Percentile	Ingestion 95 th Percentile
01/01/1973	12/31/1973	2384.0	476.8
01/01/1974	12/31/1974	367.9	73.6
01/01/1975	12/31/1975	56.8	11.4
01/01/1976	12/31/1976	8.74	1.75

Note: Super S intake rates are assumed to be equal to Type S.

Note: A SEFOR PuO₂-UO₂ MOX should be assumed.

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ORAUT (Oak Ridge Associated Universities Team), 2012, *Dose Reconstruction during Residual Radioactivity Periods at Atomic Weapons Employer Facilities*, ORAUT-OTIB-0070, Rev. 01, Oak Ridge, Tennessee, March 5. [SRDB Ref ID: 108851]

ORAUT (Oak Ridge Associated Universities Team), 2014, *Analysis of Stratified Coworker Datasets*, ORAUT-RPRT-0053, Rev. 02, Oak Ridge, Tennessee, October 8. [SRDB Ref ID: 136245]

ORAUT (Oak Ridge Associated Universities Team), 2016, *Use of Claimant Datasets for Coworker Modeling*, ORAUT-OTIB-0075, Rev. 01, Oak Ridge, Tennessee, June 17. [SRDB Ref ID: 157060]

ORAUT (Oak Ridge Associated Universities Team), 2017a, *W. R. Grace Plutonium Coworker Support Calculations, Updated Pu Sort Excel Spreadsheet*, Oak Ridge, Tennessee, May 31. [SRDB Ref ID: 172467]

ORAUT (Oak Ridge Associated Universities Team), 2017b, *W. R. Grace Plutonium Coworker Support Calculations, Pu Corrections Excel Spreadsheet*, Oak Ridge, Tennessee, August 23. [SRDB Ref ID: 172466]

ORAUT (Oak Ridge Associated Universities Team), 2018a, *Internal Dose Reconstruction*, ORAUT-OTIB-0060, Rev. 02, Oak Ridge, Tennessee, April 20. [SRDB Ref ID: 171554]

ORAUT (Oak Ridge Associated Universities Team), 2018b, *Applications of Regression in External Dose Reconstruction*, ORAUT-RPRT-0087, Rev. 00, Oak Ridge, Tennessee, March 13. [SRDB Ref ID: 170100]

SC&A (Sanford Cohen and Associates) and Salient, Inc., 2013, *Review of the NIOSH Site Profile for the W. R. Grace and Company in Erwin, Tennessee*, SCA-TR-SP2013-0041, Rev. 0-Draft, Vienna, Virginia, and Jefferson, Maryland, January 16. [SRDB Ref ID: 166040]

Attachment A, Plutonium Coworker Statistical Analysis Methods

ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005), describes the general process NIOSH uses to analyze bioassay data for the assignment of doses to individuals based on coworker results. ORAUT-PLAN-0014, *Coworker Data Exposure Profile Development* (ORAUT 2004), describes the approach and processes to develop reasonable exposure profiles based on available dosimetric information for workers at DOE sites. The ORAUT (2004) approach is assumed to be applicable to W. R. Grace, an AWE site.

Bioassay Data

Bioassay data in NOCTS for W. R. Grace employees were used to develop a representative database of coworker bioassay data using the guidance of ORAUT (2016). Plutonium gross alpha ($^{238}\text{Pu} + ^{239}\text{Pu}$) urinalysis data were extracted from the NOCTS data for employees at W. R. Grace for the period from 1967 through 1973. After extraction, these data were subjected to a 100% review for transcription errors and corrected as necessary. The data from one employee was excluded beginning in 1972 due to an incident.

The analytical results were converted to units of dpm/day based on an assumed daily urinary excretion of 1.4 L/day and the assumption that sample results with no volume noted (i.e., “dpm” or “dpm/sample” type) had a volume of 1 L. Results recorded as zero were assumed to be censored at 0.03 dpm/L, the decision level noted in the site technical basis document (ORAUT 2011).

Analysis

Statistical analysis of the plutonium gross alpha bioassay data was performed in accordance with ORAUT (2005, 2014) using the time-weighted one person-one statistic (TWOPOS) method. The data were analyzed on an annual basis except for 1972 and 1973, which were merged due to the small amount of data available for each year and the assumption that the exposure potential was similar in both years. In 1972, plutonium processing activities were completed (Congress 1986, PDF p. 72) and the fabrication of fuel elements with a mixture of uranium-plutonium MOX was discontinued (NRC 1991, PDF p. 27). Safe shutdown of the MOX facility occurred in 1973, as previously mentioned.

Table A-1 provides the results of the statistical analysis.

Intake Modeling

Each result that was used in the intake calculations was assumed to have a normal distribution. A uniform absolute error of 1 was applied to all results, thereby assigning the same weight to each result. Because of the nature of work at W. R. Grace, intakes could have been chronic or

Table A-1. 50th and 84th percentile urinary excretion rates of plutonium gross alpha, 1967 to 1973 (dpm/d).

Effective Bioassay Date	50th Percentile	84th Percentile	GSD	Number of Individuals
07/01/1967	0.183	0.472	2.58	34
07/01/1968	0.118	0.763	6.48	36
07/01/1969	0.065	0.202	3.11	25
07/01/1970	0.033	0.172	5.23	28
07/01/1971	0.032	0.070	2.18	37
01/01/1973	0.007	0.046	6.44	29

acute. However, a series of acute intakes can be approximated as a chronic intake. Therefore, intakes were assumed to be chronic and to occur through inhalation with a default breathing rate of 1.2 m³/hour and a 5- μ m activity median aerodynamic diameter particle size distribution.

The results were entered in the Integrated Modules for Bioassay Analysis (IMBA) computer software to obtain intake rates for the assignment of dose distributions. IMBA was then used to fit the bioassay results to a series of chronic inhalation intakes. The intake assumptions were based on observed patterns in the bioassay data. Periods with constant chronic intake rates were chosen by the selection of periods in which the bioassay results were similar. A new chronic intake period was started if the data indicated a significant sustained change in the bioassay results. By this method, the years from 1967 through 1973 were divided into multiple chronic intake periods. For intake modeling purposes only, the plutonium gross alpha results were assumed to be ²³⁹Pu.

The plutonium intake periods were independently fit using only the bioassay results from the single intake period. This method likely results in an overestimate of intakes for exposures that extend through multiple assumed intake periods.

The solid lines in Figures A-1 to A-6 (see Attachment A) show the individual fits to the 50th and 84th percentile excretion rates for Type M materials. Excluded results are shown in red (dark gray in grayscale) in the figures; included results are shown in blue (light gray in grayscale). Figures A-7 and A-8 show the 50th-and 84th percentile predicted excretion rates, respectively, from all Type M intakes. Figures A-9 to A-14 show the 50th-and 84th-percentile predicted excretion rates for Type S materials. Figures A-15 and A-16 show the 50th-and 84th-percentile predicted excretion rates, respectively, from all Type S intakes.

Intake Assignment

For each intake period below, the GSDs were determined by dividing the 84th-percentile intake rates by the 50th-percentile rate. For the calculation of doses to individuals from bioassay data, a GSD of three is used to account for biological variation and uncertainty in the models (ORAUT

2014). The same models were used for fitting the coworker data, so the same uncertainty applies. Therefore, a minimum GSD of 3 was assigned for each of the intake periods.

Multiple intake periods were fit to the derived 50th- and 84th-percentile plutonium excretion data. The 95th-percentile intake rates were calculated using the following equation:

$$95\text{th-percentile intake} = 50\text{th-percentile intake} \times \text{GSD}^{1.645} \quad \text{Equation A-1}$$

Table A-2 summarizes the 50th- and 95th-percentile plutonium intake rates that correspond to an intake of Type M materials that were calculated from the excretion rates. Table A-3 provides the same information for Type S materials.

Table A-2. Type M plutonium gross alpha intake rates (dpm/d).

Start	End	50th Percentile	GSD	95th Percentile
01/01/1967	12/31/1968	49.93	4.65	626.64
01/01/1969	12/31/1971	10.96	3.39	81.50
01/01/1972	12/31/1973	2.567	6.59	57.10

Table A-3. Type S plutonium gross alpha intake rates (dpm/d).

Start	End	50th Percentile	GSD	95th Percentile
01/01/1967	12/31/1968	1,692	5.07	24,431
01/01/1969	12/31/1971	291.9	3.30	2,076
01/01/1972	12/31/1973	107.2	6.59	2,384

Plutonium Statistical Analysis Results

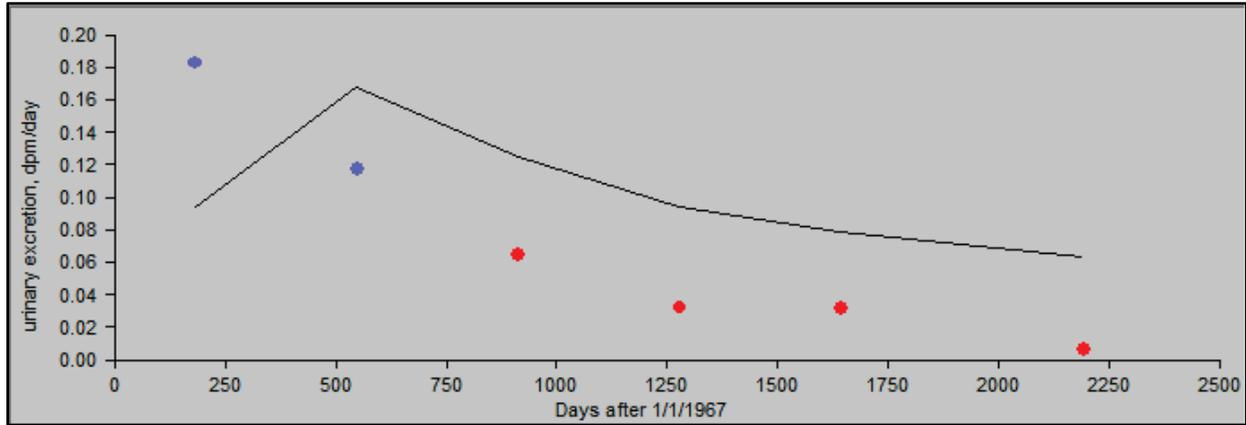


Figure A-1. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1967 to 1968, Type M

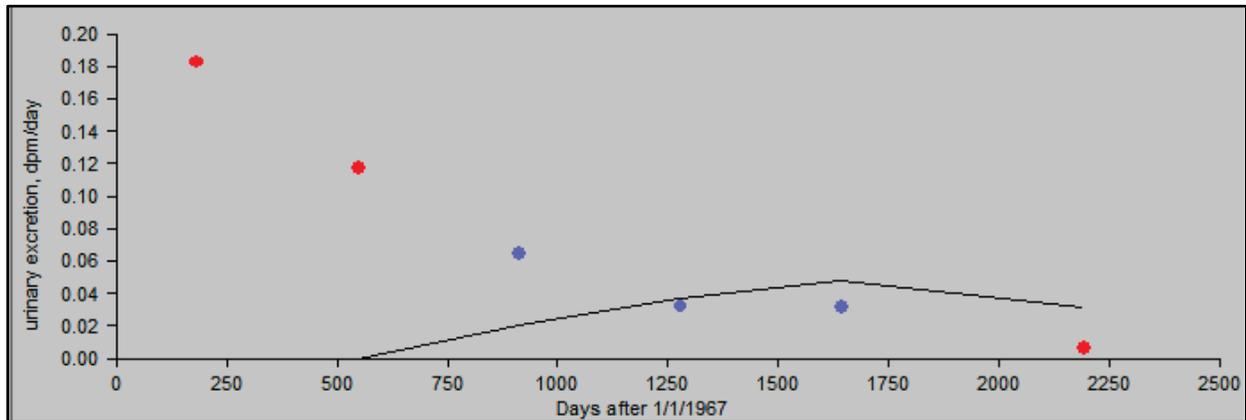


Figure A-2. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1969 to 1971, Type M

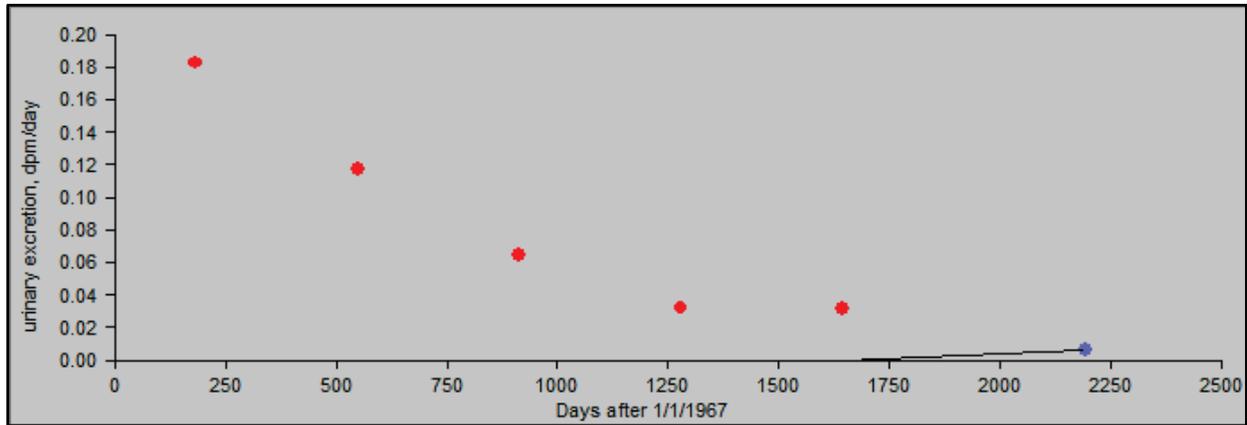


Figure A-3. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1972 to 1973, Type M

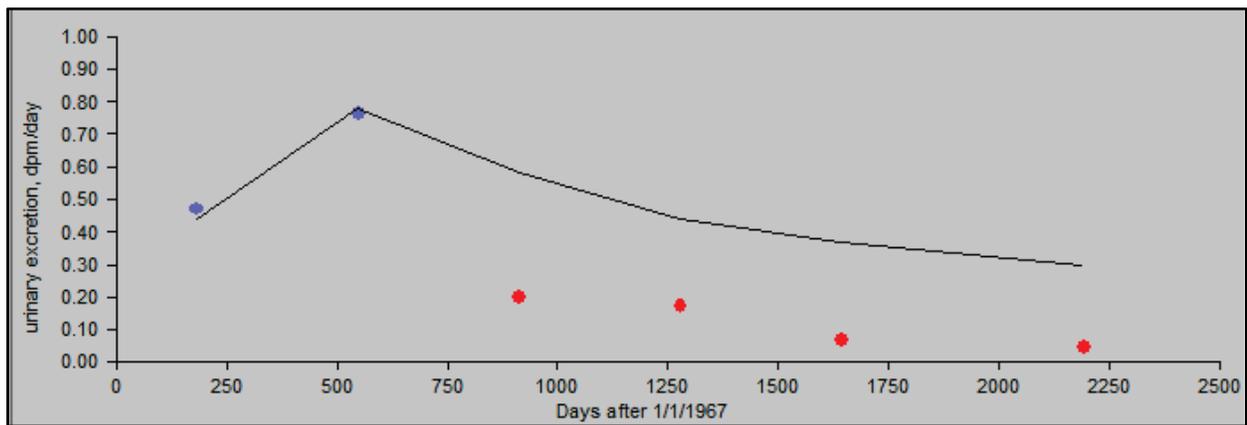


Figure A-4. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1967 to 1968, Type M

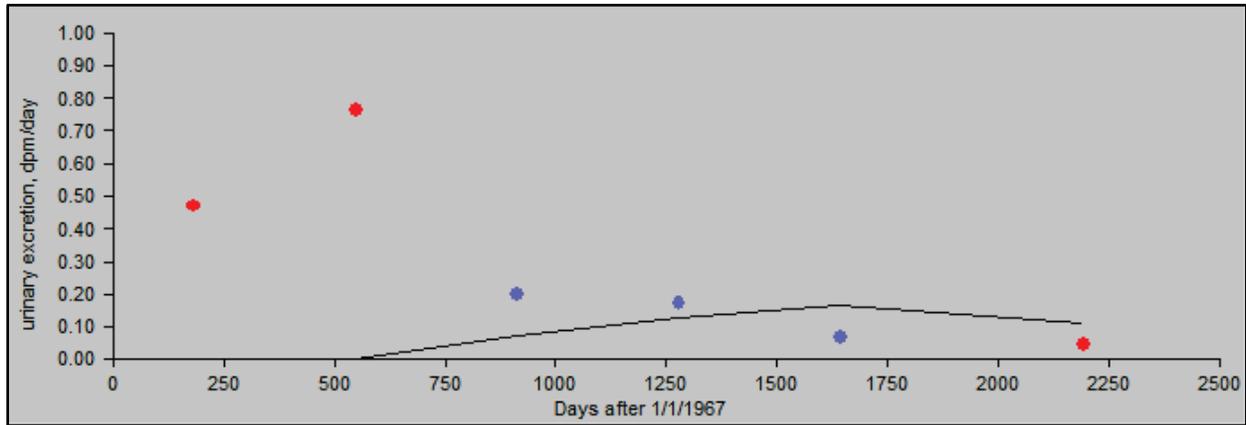


Figure A-5. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1969 to 1971, Type M

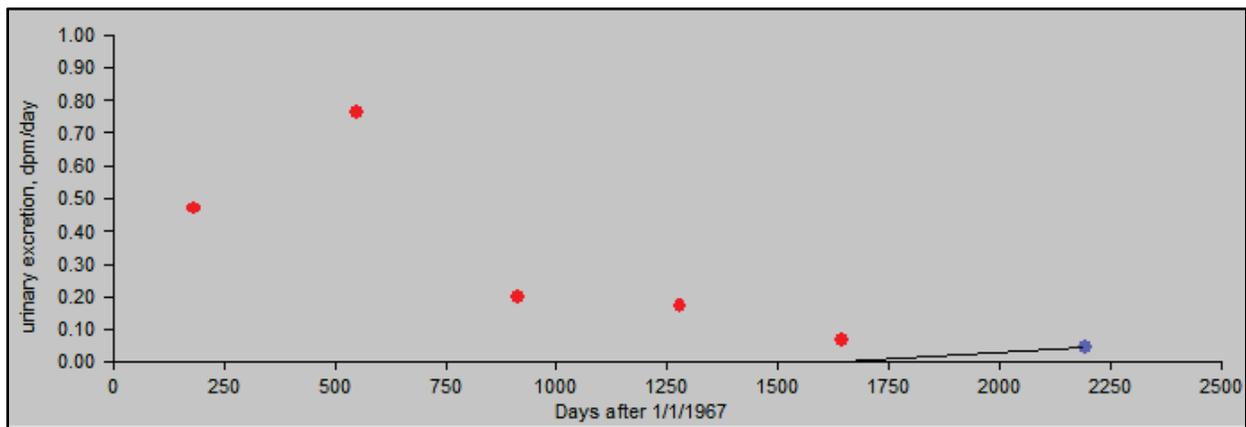


Figure A-6. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1972 to 1973, Type M

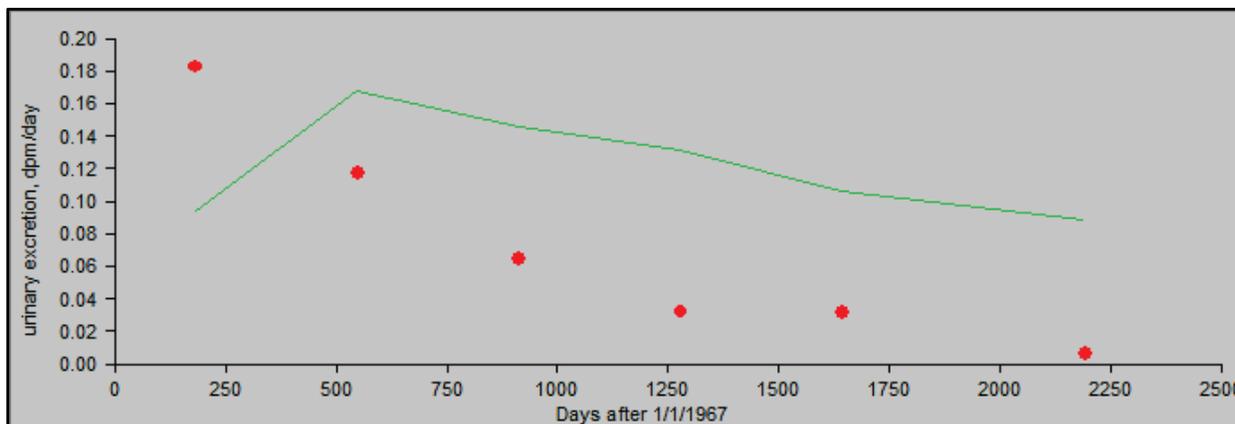


Figure A-7. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, all years, Type M

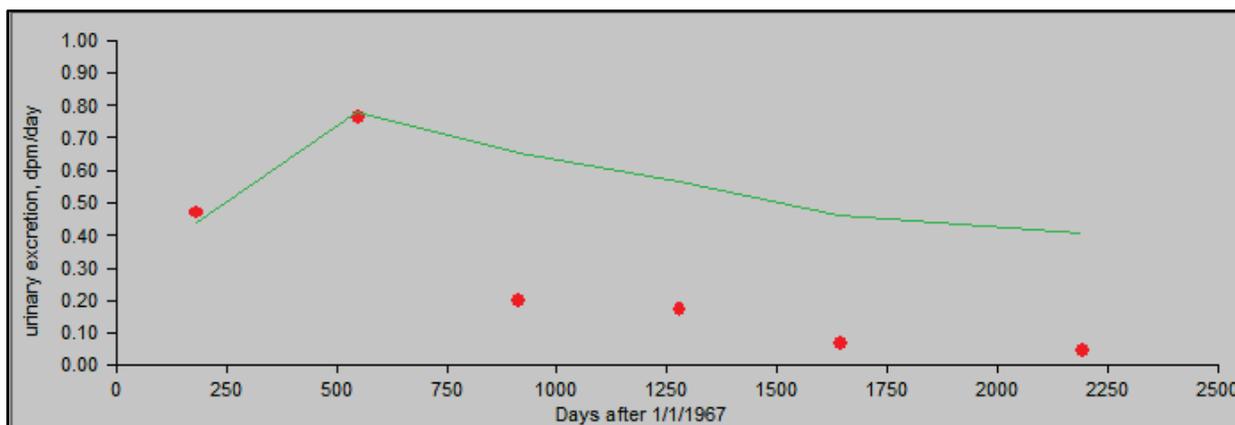


Figure A-8. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, all years, Type M

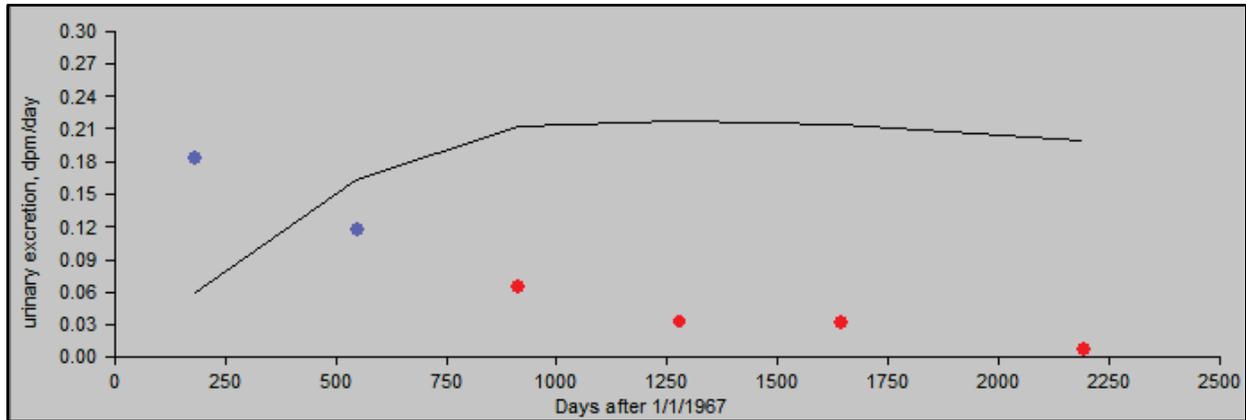


Figure A-9. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1967 to 1968, Type S.

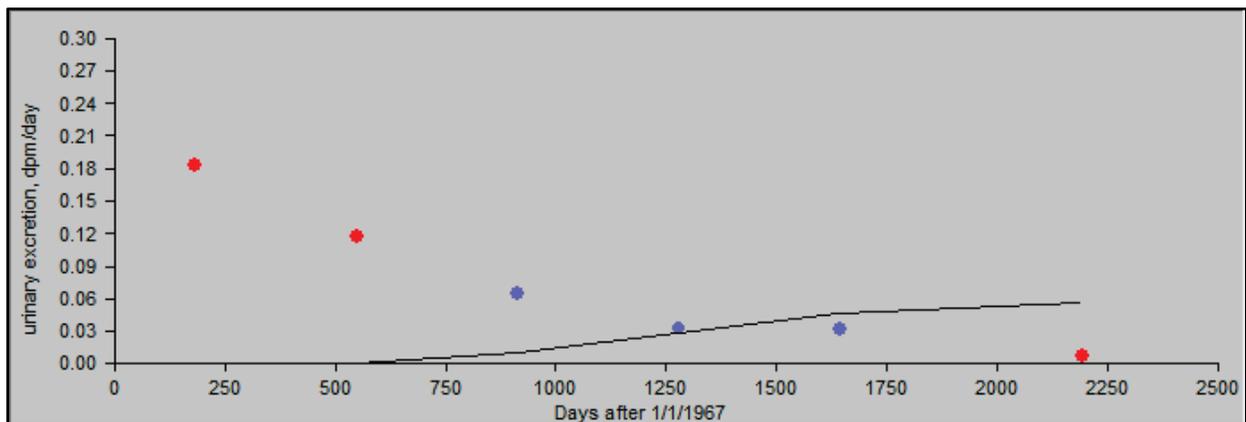


Figure A-10. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1969 to 1971, Type S.

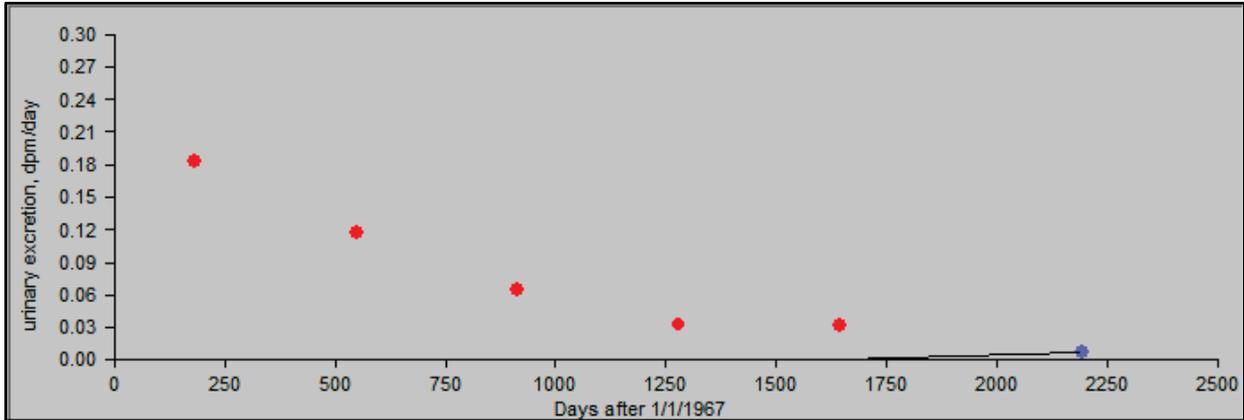


Figure A-11. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, 1972 to 1973, Type S.

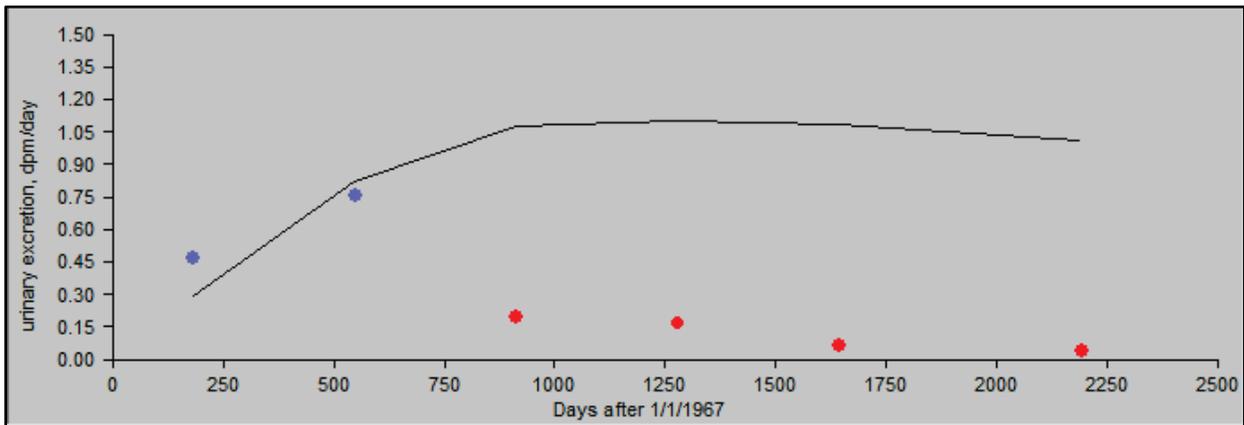


Figure A-12. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1967 to 1968, Type S.

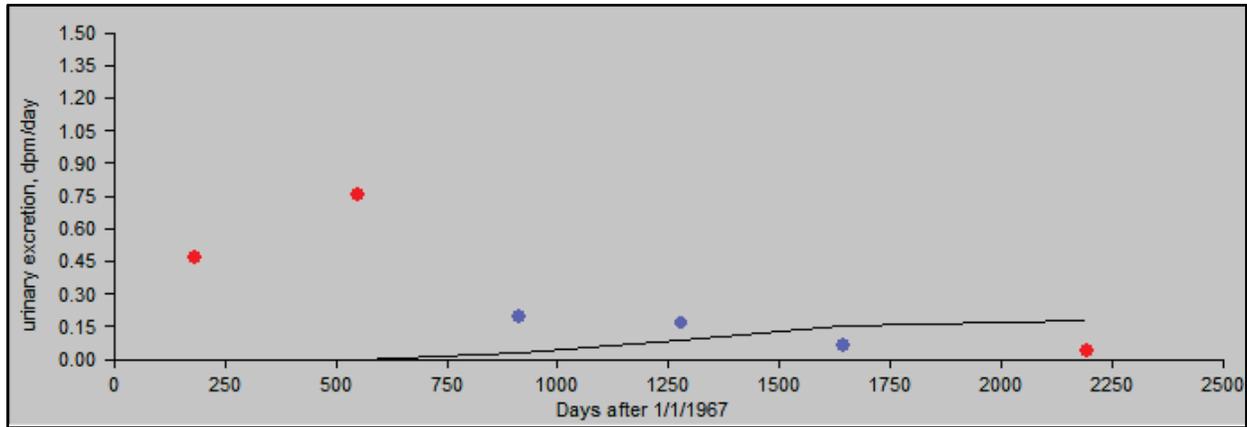


Figure A-13. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1969 to 1971, Type S.

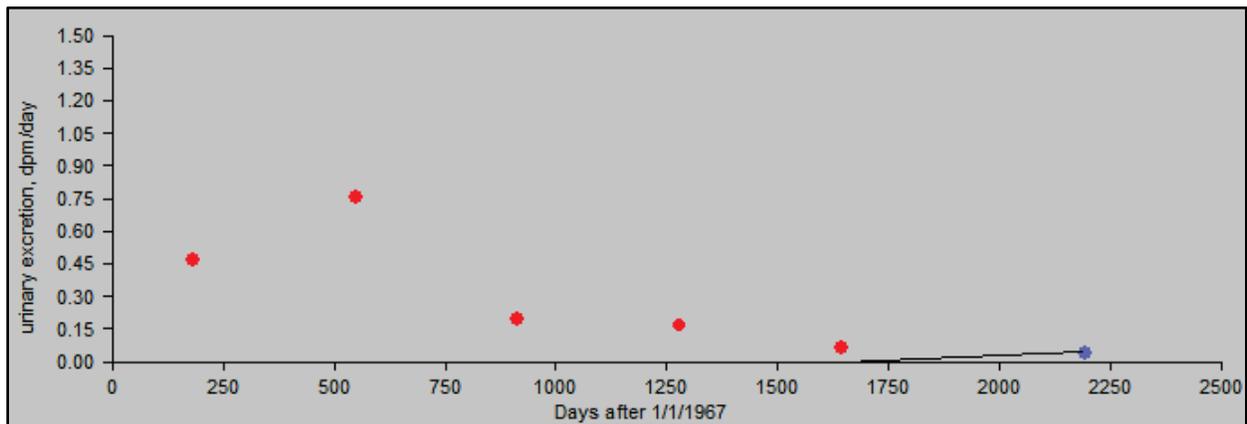


Figure A-14. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, 1972 to 1973, Type S.

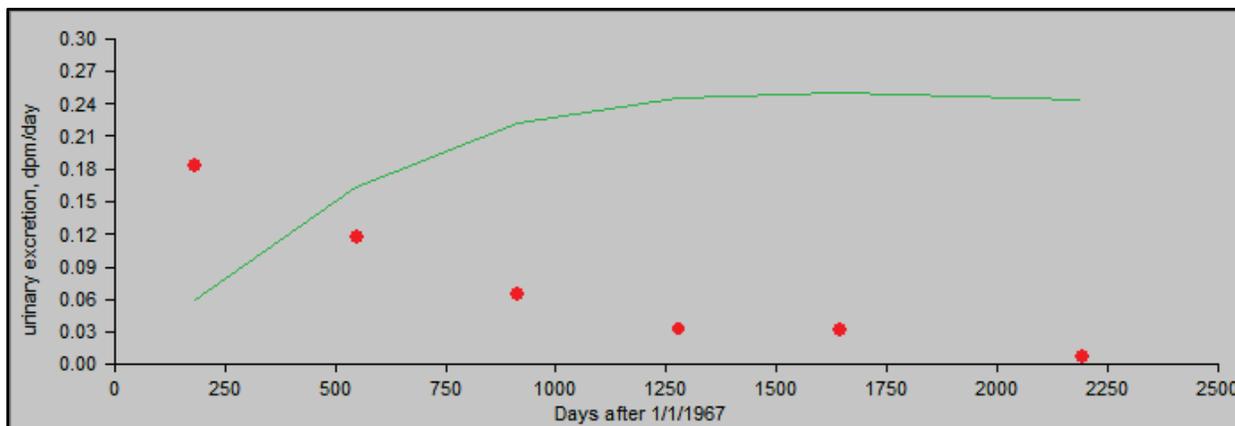


Figure A-15. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 50th percentile, all years, Type S.

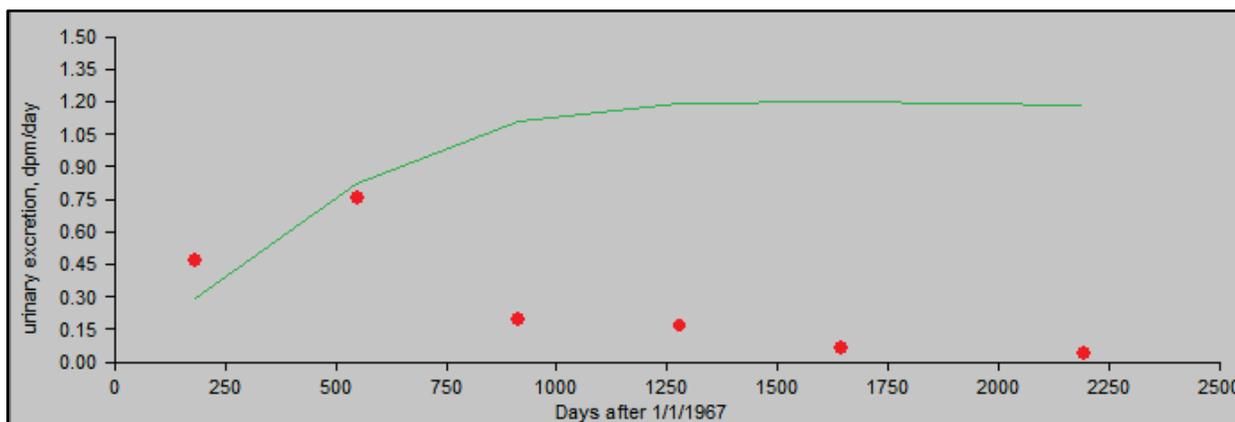


Figure A-16. Predicted plutonium gross alpha bioassay results calculated using IMBA-derived plutonium gross alpha intake rates (line) compared with measured bioassay results (dots), 84th percentile, all years, Type S.

Attachment B, Plutonium Coworker Statistical Analysis Instructions

This attachment provides the instructions used to perform the analysis for the development of the plutonium coworker intake rates.

Data Sources:

- *W. R. Grace Plutonium Coworker Support Calculations, Updated Pu Sort Excel Spreadsheet (ORAUT 2017a).*
- *W. R. Grace Plutonium Coworker Support Calculations, Pu Corrections Excel Spreadsheet (ORAUT 2017b).*

Instructions

- Update the source data file using the source data corrections file and identifying lines by the “Unique ID#” column.
 - Replace individual cell contents based on cell contents in the corrections file.
 - If a cell in the corrections file contains “blank,” then delete that cell’s contents in the source data file.
 - If the corrections file “HP Review Comments” column contains the word “exclude” or “remove” then do not use that line for the statistical analysis.
- “Use the Sample Date” column (Column J) as the date of sample collection.
 - If the month and/or day of the date are illegible (“XX” characters), substitute in the month and/or day most claimant-favorable for the TWOPOS calculation.
 - Do not use results with an illegible year.
- “Use the Claim #” field (Column A) as the individual identifier.
- Only use data with “urine” in Column G.
- Data set exclusions:
 - Exclude all data from claim [redacted] beginning in 1972 due to an incident.
 - Exclude data after 12/31/1973.
- Assume all results are plutonium gross alpha measurements.

- Assume records with units of “dpm/sample” or “dpm/s,” and no sample volume, have a sample volume of 1 L.
- Assume records with units of “dpm/l” have a sample volume of 1 L.
- Assume record with volume units of “ml” have a volume of mL. Convert this volume to L.
- The sample with “UniqueID#” = 876 has no volume units. Assign it volume units of mL and convert the volume to L.
- Assume any “Result” equal to 0 is censored at 0.03 dpm/L.
- Divide the “Result” by the volume and multiply by 1.4 L to convert “Result” to activity excreted per day.
- Perform the statistical analysis on an annual basis for 1967 through 1973 (except merge 1972 and 1973) in accordance with the TWOPOS method in the latest revision of ORAUT-RPRT-0053 (ORAUT 2014).

Attachment C, Exposure Model To Develop Plutonium Intakes Based on Airborne Concentration Data

This attachment provides the detailed method used to develop the plutonium intakes based on airborne concentration data.

Methods

Annual or semiannual average airborne concentrations in percent of the maximum permissible concentration (MPC) from plutonium facilities were converted to inhalation intake rates in units of dpm/day. A lognormal distribution was fit to the annual data using the regression on order statistic method (ORAUT 2014, 2018b). The data were analyzed as two periods with a single distribution in each period. The first period was 1976 through 1987, and it consists of 84 values. The second period is 1988 through 1993, and it consists of 51 values. The reason for analyzing the data as two separate periods is due to the trend in the results. From 1976 through 1987, the facility was shutdown. From 1988 through 1993, characterization, preparation for D&D, and D&D activities occurred. The preparatory work for D&D appears to coincide with the trend of lower airborne concentrations beginning in 1988 (NFS 1994, PDF p.7-8). D&D of the MOX facility began September 1990.

Airborne Concentration Data

Gross alpha airborne concentration data were obtained from the Site Research Database (SRDB) (NFS ca. 1979, ca. 1989, ca. 1999, ca. 2005). These data were assumed to be representative of W. R. Grace radiological worker exposure because the data were taken from the plutonium buildings at W. R. Grace. Although gross alpha airborne concentration existed at W. R. Grace through 2005, there were no data from plutonium buildings beyond 1993. Therefore, this exposure model ended in 1993.

These alpha airborne concentration results were compiled by the Project's data entry group into a spreadsheet that contains average airborne alpha radioactivity concentrations in units of % MPC for 1976 to 1993 by building, area, and process. These data were subjected to 100% review for transcription errors and corrected, as necessary. A second spreadsheet was developed from the previous one for summarizing and further review.

NFS (1988) identified specific areas in Buildings 110 and 234 as plutonium related. The plutonium work locations from these facilities were reviewed and verified. The air concentration results from these plutonium areas were then selected for development of an exposure model. The only exceptions to the plutonium data were the airborne concentration data associated with the 234A-Pu Wet Chem Cell and 234 Cell, which were excluded. The air concentration data outside the cell areas were used to assess plutonium intakes for unmonitored workers after the end of plutonium operations at the site.

Table C-1 was used to define which air concentration results were associated with plutonium.

Table C-1. Determination of plutonium vs. uranium areas.

Air Sample Identifier	Description/Comment	Include or Exclude Data
110A, B	Uranium areas	Exclude all results unless identified as plutonium
110C, D	Plutonium areas (see 110D-1 exception below)	Include unless identified as uranium
110D-1	Uranium Spectrographic Lab; NDA Lab results through 1st half of 1987	Exclude unless identified as plutonium
110C, D	Pu, ²³³ U Labs	Include
110 Pu areas	Any 110 air sample identified as Pu such as Pu Wet Chem Lab, Pu Spec Lab	Include
110 Pu Wet Chem Lab 110 Pu Spec Lab	Results for 1st half 1989 are not valid	Exclude 1st half 1989, otherwise include
110	Any 110 samples not identified as Pu	Exclude unless identified as plutonium
110 Avg	Element not specified, but location added in 1990 for D&D	Include
234A	Production area, several locations	Include except for "Cell"
234A Cell	Wet Cell or any identified as "Cell"	Exclude
234B- ²³³ U Process Area	This was a decommissioned ²³³ U area used for other projects; it has results reported through 1985	Exclude
234C	Various descriptions (Pu Dissolution Area, Fabrication Area)	Include
234 Building	Location first appeared in 1986 report, determined to be the same as the "234A Pu production area"	Include
234 All areas (1990-1993)	No uranium areas reported during decommissioning period	Include

Note: A couple locations appear to have had slight name changes, with the same result reported in subsequent reporting period. These are readily identifiable. Do not duplicate that data.

Analysis

Average results in % MPC were reported annually (1976) and semiannually (1977-1993) for each plutonium sampling location. The results were converted to intake rates in units of dpm/day according to the MPC the records indicate was being used for plutonium, which was 2×10^{-12} $\mu\text{Ci/mL}$, or 4.44 dpm/ m^3 .

A breathing rate of 1.2 m^3/hour , an 8-hour workday, and 250 days/year exposure were assumed for a total of 2,400 m^3/year .

Table C-2 provides a summary of the plutonium air concentration results.

Table C-2. Plutonium gross alpha air concentration results (1976 to 1993).

Year	Percent of MPC Average	Percent of MPC Maximum	Total Number of Airborne Concentration Results
1976	23.7	46.5	4
1977	10.3	25.0	8
1978	19.9	22.5	8
1979	15.2	21.3	8
1980	13.8	30.0	8
1981	12.2	16.9	8
1982	7.60	10.0	8
1983	8.07	10.8	8
1984	21.0	37.0	8
1985	11.7	17.8	8
1986	20.7	23.6	4
1987	7.99	10.8	4
1988	2.92	5.5	6
1989	3.00	3.3	3
1990	0.07	0.29	12
1991	0.16	0.67	12
1992	0.36	2.38	12
1993	0.44	1.08	6

Intake Modeling and Assessment

A lognormal distribution was fit to the data using the regression on order statistic method (ORAUT 2018b).

Table C-3 summarizes the 50th- and 95th-percentile plutonium intake rates that correspond to an intake that was calculated from the airborne concentration data exposure model. Ingestion intakes should also be assigned when airborne concentration data are used to assign inhalation intakes, as described in OCAS-TIB-009, *Estimation of Ingestion Intakes* (NIOSH 2004), and are included in the Table C-3.

Table C-3. 50th--percentile plutonium intake rates (dpm/d).

Start	End	Inhalation 50th Percentile	Ingestion 50th Percentile	GSD	Inhalation 95th Percentile	Ingestion 95th Percentile
01/01/1976	12/31/1987	3.503	0.701	1.743	8.741	1.748
01/01/1988	12/31/1993	0.040	0.008	7.616	1.132	0.226

The 1993 intakes should be extended from 1994 through 2011. Figures C-1 to C-2 below show the observed plutonium air concentration versus the standard normal quantiles. The y-axis is on a logarithmic scale, so data that fall on a straight line on this plot are lognormally distributed. The line in the plot is the best-fit lognormal model, where the slope of the line is the GSD and the y-intercept is the GM.

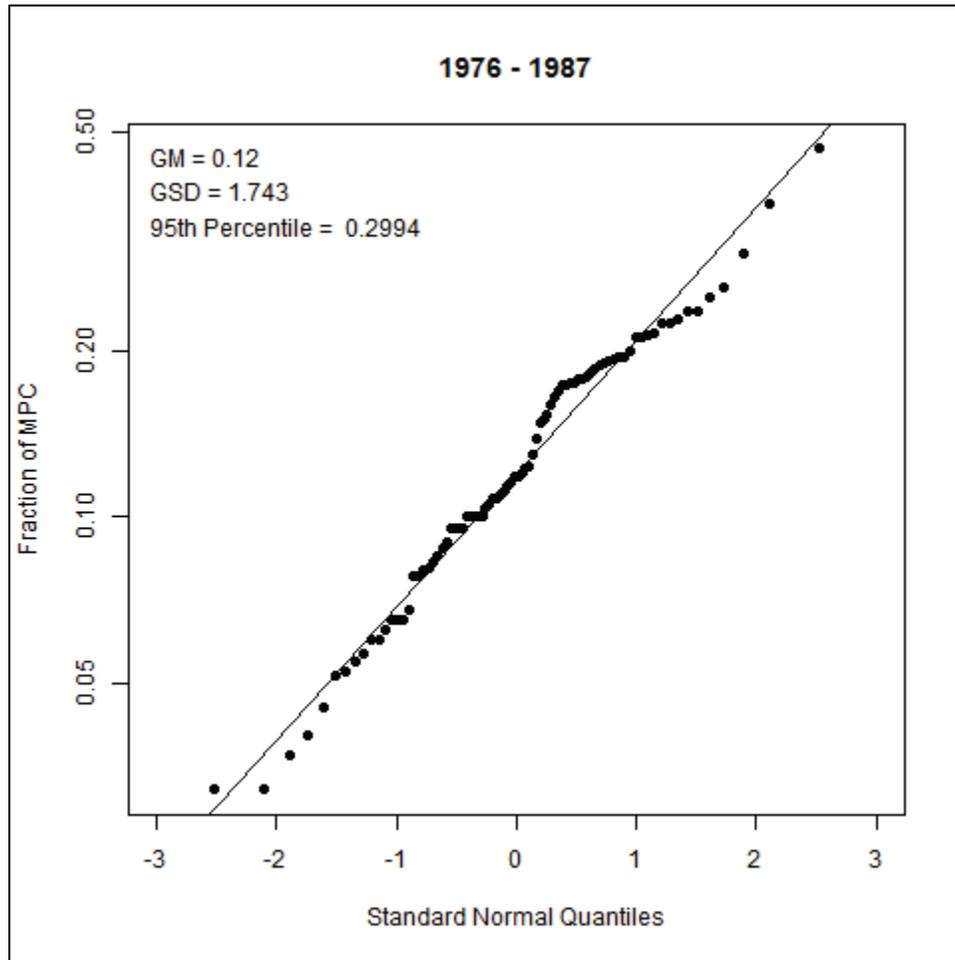


Figure C-1. Observed 1976-1987 fraction of plutonium MPC versus the standard normal quantiles (censored data do not appear on the plot)

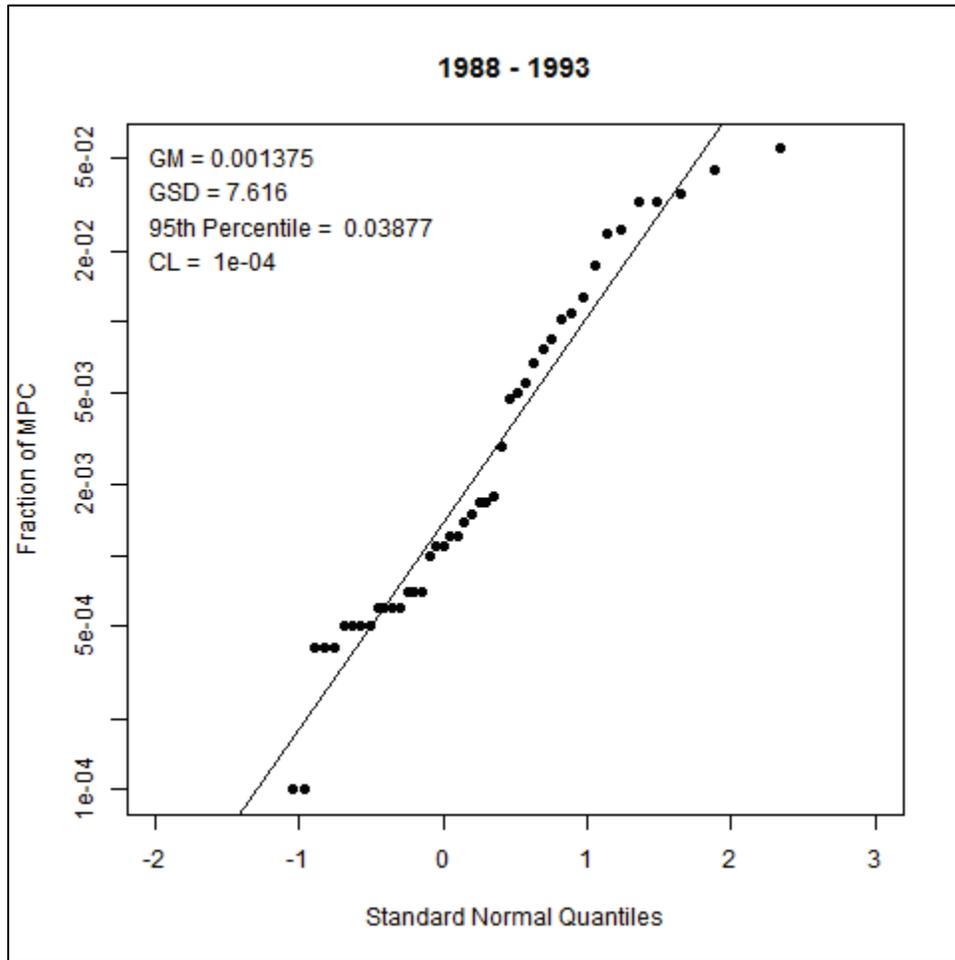


Figure C-2. Observed 1988-1993 fraction of plutonium MPC versus the standard normal quantiles (censored data do not appear on the plot)

Attachment D, Plutonium Bioassay Program Review

This attachment provides a summary of the plutonium bioassay program at W. R. Grace. Information was taken from records in the Site Research Database (SRDB) and from worker records in NOCTS. It should be noted that the bioassay data from NOCTS discussed in this review is not a complete accounting of all workers who were monitored for intakes of plutonium, but some conclusions about the bioassay program can be made from those records coupled with other information.

Table 1 shows key dates for plutonium work, including the production period, idle period, and several key dates for the D&D project. For purposes of assessing potential exposures, the plutonium work was divided into three time periods based on potential for exposure and the differences in bioassay requirements at the site. Each of the three periods are discussed below. Although nearly all personnel were monitored for intakes of uranium during these periods, only some workers were monitored for intakes of plutonium.

1965 through 1973—Plutonium Production Period

Plutonium fuel was produced starting sometime in 1965 and continuing through 1972. Some references indicate the production continued into 1973 before the production facilities were shut down (see Table 3). The presence of routine sampling for some workers extended into 1973, so production was assumed in 1973 whether it was actual production or work on closing the facility.

The bioassay data from claimant records indicates some workers were routinely monitored for plutonium intakes by urinalysis from April 1967 through the first half of 1973. The bioassay dataset from NOCTS used for the coworker analysis includes multiple workers with multiple sample data throughout that period. It is not known if all exposures were monitored during that period, so coworker intake data is provided for use in dose reconstructions.

Per the Table 3 timetable of plutonium projects, the coworker intake assessment covers all the production periods except the MOX fuel project for DuPont/SROO that used 16 kg of plutonium from 1965 through 1966. The coworker intake rates for the 1967 through 1968 period should be used for 1965 and 1966 because no other data are available for that period. There is also an existing Special Exposure Cohort class for that period that extends through 1970.

Workers with potential plutonium exposures should be assigned intakes based on individual bioassay data and/or coworker data (as provided in this paper) for the period from 1965 through 1973. Furthermore, because all the plutonium projects involved MOX fuel, all workers with potential exposure to plutonium should be assigned uranium intakes based on their uranium bioassay data. For the period after 1970, when residual uranium intakes would normally apply for exposure to weapons-related AEC uranium in other buildings, those intakes should not be applied because uranium bioassay should be used to assess intakes.

1974 through 1989—Post-Production Period

The intakes for 1976 through 1989 in Table 7 are based on the 95th percentile of the general area air sampling data from the monitored plutonium areas. Because there are no data available for 1974 and 1975, favorable intake rates were modeled assuming a lognormal depletion curve from the operational intakes in 1973 to the intakes based on air samples starting in 1976.

Based on trends in the air sampling data, calculated intakes from that sampling data were assessed as two periods, one from 1976 through 1987 and another from 1988 through 1993. The intakes from the 1976-1987 period were based on a concentration of 6.0×10^{-13} $\mu\text{Ci/mL}$, or 30% of the MPC of plutonium, while the intakes for the 1988-1993 period were based on a concentration of 7.8×10^{-14} $\mu\text{Ci/mL}$, or 3.9% of the MPC. The site was using an MPC of 2×10^{-12} $\mu\text{Ci/mL}$. A bounding intake for workers who have no plutonium bioassay, but might have entered the plutonium facilities, is based on continuous exposure at those levels for 2,000 hours per year. Based on the MPC, this results in an exposure of 600 MPC-hours per year for the 1974-1987 period and 78 MPC-hours per year for the 1988-1993 period.

From 1974 through 1989 the plutonium facility was shut down awaiting D&D. The NOCTS data indicate the routine plutonium bioassay program ended in 1973. There are no references or data to indicate a routine bioassay program for plutonium from 1974 through 1989. However, NOCTS has a few bioassay results for some workers in some of these years. Table D-1 summarizes the urinalysis data for plutonium in NOCTS from 1974 through 1989. It should be noted that some of the records for a particular worker include laboratory reports of plutonium bioassay for several other workers, but those numbers are not included in Table D-1 because the results for other workers were redacted and the implied number of additional monitored personnel cannot be determined with certainty.

There were a few known activities that occurred in 1974 through 1989. Records of entries in the plutonium facilities was found for the following activities:

- A routine air sampling program in multiple locations started in 1976.
- Some facility maintenance activities occurred.
- A limited D&D project was performed in 1984.
- Preparation work for the main D&D project was done in 1988 through 1989.

Available references and NOCTS data and information about the workers who were monitored for plutonium during these years were reviewed for pertinent information on plutonium work and bioassay during the 1974 through 1989 period.

No information was found on activities for the latter half of 1973 through 1975. One worker had a single positive bioassay result in 1974 (see Table D-1), but no information is available for the

Table D-1: Plutonium bioassay data in NOCTS 1974 through 1989.

Year	Number of Pu Bioassay Results	Notes
1974	1	One worker was monitored. Results were reported as [redacted].
1975-1977	0	No Pu bioassay results.
1978	6	[Redacted] workers were monitored. Four results were reported as 0 dpm/L. Two results were 0.03 dpm/L.
1979	0	No Pu bioassay results.
1980	7	Seven workers were monitored in July and August. All reported 0 dpm/L.
1981	2	Two workers were monitored. Results were reported as 0 dpm/L.
1982-1983	0	No Pu bioassay results.
1984	6	[Redacted] workers were monitored [redacted] through [redacted]. Four results were either 0 or less than the sample lower limit of detection. The other two samples were [redacted] and [redacted]. The [redacted] monitored in [redacted].
1985	2	[Redacted] monitored. Results were reported as 0 dpm/L.
1986-1987	0	No Pu bioassay results.
1988-1989	1+	One worker was monitored. Positive sample followed by bioassays.

Note: One worker had multiple bioassay results; however, those were results were not included in this table because that worker was on a long-term monitoring program and did not enter plutonium areas during the period.

reason for that monitoring. That worker's employment ended in [redacted], and no additional bioassay or information is available.

The air sampling records indicate a multilocation routine air sampling program throughout the site was established by 1976; it included locations in the plutonium facilities (see Appendix C for more information). These samples are believed to have been fixed-location routine samples rather than job-specific data. At a minimum, personnel would have made entries into the plutonium areas to set up and maintain the air sampling pumps and to make routine collections of the filters. The sample results are sufficient to estimate or bound intakes from those entries into the building or for entries for inspections. As noted in Attachment C, in some years air samples were collected in the Wet Cell area of Building 234, which was a separately contained cell requiring multiple protective clothing and protection. Although the Wet Cell data are not included in the analysis in Attachment C, the monitored areas outside of the Wet Cell were included.

No records were found of other specific maintenance activities other than records of a worker who was monitored several times in 1988 and 1989 after working [redacted]. Table D-1 also shows plutonium bioassay records in 1978, 1980, and 1981. Of the 15 total results in NOCTS for those years, 13 were reported as 0, and the other 2 were reported as 0.03 dpm/L, although the sample-specific detection limit is not provided. No information was found for a reason for those bioassay data, although intakes can be estimated from the data.

In addition to the samples and activities discussed above there are also monitoring results from 1984 and 1985, which were collected for a limited D&D project. NOCTS had six plutonium bioassay samples for [redacted] workers in 1984. Two of those are presumed positive, and [redacted] had a plutonium bioassay in early 1985. Per the record of the computer-assisted telephone interview (CATI) for one of the monitored workers (claim redacted), part of his duties was [redacted]. The CATI provided this information on work that was done in 1984-1985:

While performing decommissioning work in the Plutonium Facility, [redacted].

This information coincides with a reference concerning a limited D&D project to remove welding devices and X-ray equipment previously used on plutonium fuel rods. The NRC described the work (NRC 1991):

During 1983 and 1984, NFS began work to decommission the fabrication area of Building 234. This area contained welding devices and X-ray equipment used to fabricate and inspect plutonium fuel rods. Materials removed during this effort were packaged in drums and burial boxes and are currently being stored onsite.

The reference for this information was an NRC document on the status of the site in 1986 that was used to brief a Congressional subcommittee. The same report noted that the plutonium processing equipment and gloveboxes continued to remain in the building. Although the worker interview and NRC document have slightly different dates (one says the project started in 1983 the other says 1984), the descriptions of the work are similar. NOCTS had no plutonium bioassay data from January 1985 until 1988. For part of that period, May 1985 to April 1, 1986, the union was on strike (Congress 1986, PDF pp. 12-46).

Per Table D-1, the plutonium facility was characterized and systems upgraded in 1988 and 1989 to prepare for the D&D project, including cold testing of the equipment. One claimant in NOCTS was monitored in 1988 and 1989, but that was from cleanup work [redacted]. The routine air sample data show airborne levels were lower in these years than in the earlier years.

Starting in 1988 nearly all worker records in NOCTS have routine chest count results that provide detection limits for several radionuclides including ^{241}Am and ^{239}Pu . These were performed on a germanium detector system that became operational in September 1987 (Author unknown 1988). Because it appears all radiological workers had in vivo counts in this era, the presence of a chest count is not necessarily an indicator of exposure to plutonium. However, the results can be used to estimate a bounding or missed dose to workers exposed to plutonium.

1990 through 1993—Plutonium Facility D&D

Table D-1 shows cold testing of D&D equipment in the plutonium facility in 1990 before the September start of the actual D&D work. The project was completed in December 1993 with the exception of some subsurface areas inside the Wet Cell that remained under containment; final cleanup of that subsurface area was deferred.

An April 1989 addendum to the NFS NRC license contained requirements for D&D of the plutonium facilities that included bioassay requirements. In addition to the routine bioassay program, workers participating in the plutonium D&D project were required to have quarterly urine and/or fecal bioassay and annual in vivo examinations (NFS 1989b, PDF p. 170).

A review of NOCTS bioassay records indicate a routine bioassay program was initiated for several workers in January 1990, and these records indicate a quarterly frequency consistent with license modification. In addition, the records contain additional special samples for some claimants. In vivo chest counts are provided in this period as well. It appears routine urine quarterly plutonium bioassay ended after the D&D project ended in 1993, although annual in vivo examinations continued.