

ORAU TEAM Dose Reconstruction Project for NIOSH

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ACRONYMS AND ABBREVIATIONS

AMAD activity median aerodynamic diameter

Bq becquerel

dpm disintegrations per minute

EEOICPA Energy Employees Occupational Illness Compensation Program Act of 2000

g gram

hr hour

KPA kinetic phosphorescence analysis

L liter

MDA minimum detectable amount MDC minimum detectable concentration

mg milligram mL milliliter mo month

nCi nanocurie

NCRP National Council on Radiation Protection and Measurements

NIOSH National Institute for Occupational Safety and Health

pCi picocurie

PGDP Paducah Gaseous Diffusion Plant

Ppb parts per billion ppm parts per million

RU Recycled Uranium

TRU transuranic

U.S.C. United States Code
UF₆ uranium hexafluoride
UO₃ uranium trioxide

μCi microcurie μg microgram μm micrometer

5.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety (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 facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384I(5) and (12)]. EEOICPA defines a DOE facility as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program)" [42 U.S.C. § 7384I(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ..." [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work.

As noted above, the statute includes a definition of a DOE facility that excludes "buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program" [42 U.S.C. § 7384l(12)]. While this definition contains an exclusion with respect to the Naval Nuclear Propulsion Program, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled "Exposure in the Performance of Duty"] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external dosimetry monitoring results are considered valid for use in dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction. NIOSH, however, does not consider the following exposures to be occupationally derived:

- Radiation from naturally occurring radon present in conventional structures
- Radiation from diagnostic X-rays received in the treatment of work-related injuries

¹ The U.S. Department of Labor is ultimately responsible under the EEOICPA for determining the POC.

5.1.1 Purpose

This TBD provides technical data and other key information which will serve as the technical basis for evaluating internal occupational radiation dose for EEOICPA claimants who were employed at the Paducah Gaseous Diffusion Plant.

5.1.2 <u>Scope</u>

This document discusses the radionuclides potentially encountered by Paducah Gaseous Diffusion Plant (PGDP) employees during the Plant's operational history. The PGDP mission was to enrich uranium in the form of uranium hexafluoride (UF $_6$) from roughly 0.7% 235 U (natural enrichment) to around 3% 235 U for use in domestic and foreign power reactors (DOE 2000, p. 8). Enrichment operations began in 1952 in the first four process buildings, C-331, C-333, C-310, and C-315. From 1953 until 1977, UF $_6$ feed material was produced from uranium trioxide (UO $_3$) at the plant. From 1953 to 1964, and again from 1968 to 1977, UF $_6$ was produced from the recycled uranium (RU) produced from spent reactor fuel. In May 1977, the feed plants ceased operation and all feed to the enrichment process was in the form of UF $_6$ obtained from outside sources. Other chemical compounds of uranium were present throughout the Plant's history including UO $_2$ F $_2$, UF $_4$, and UO $_3$.

ORAUT (2004a) contains detailed information on the history of PGDP and the feed conversion and enrichment process.

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

5.2 SOURCE TERM

The radionuclides potentially encountered by PGDP employees consist primarily of the isotopes of uranium, ²³⁸U, ²³⁴U, and ²³⁵U. The progeny of dosimetric interest for these radionuclides includes ²³⁰Th and ^{234m}Pa (BJC 1999, p. 8). While a range of ²³⁵U enrichment values (from natural to 3%) was encountered in feed and product material, the dose reconstructor should assume a nominal enrichment value of 2% ²³⁵U for all feed or product materials [1].

The presence of transuranic (TRU) and fission product isotopes has been known since the early days of operation (ORAUT 2004a). These radionuclides were present in small amounts in the feed material produced from RU, and include ²³⁷Np, ²³⁹Pu (PACE and University of Utah 2000), and ²⁴¹Am (Hightower et al. 2000). Technetium-99 is the fission product of concern from a dosimetry standpoint. During the conversion of the feed material from UO₃ to UF₆, the elemental species of the TRU and fission products react differently from a chemical standpoint. For example, ⁹⁹Tc tends to form a very volatile fluoride; readily introduced into the enrichment cascade, it essentially follows the UF6 to the surge and/or product station. On the other hand, both ²⁴¹Am and ²³⁹Pu form essentially nonvolatile fluorides. Most of these isotopes will remain in the feed conversion byproducts (i.e., ash) or in the feed cylinder heels, a primary source of TRU exposure from this process (Hightower et al. 2000). Some ²⁴¹Am will appear in the cascade components as a result of the decay of ²⁴¹Pu. While ²³⁷Np forms a volatile fluoride, it rapidly oxidizes and plates on the metal cylinder walls (PACE and University of Utah 2000, p. 26), so the main hazard from this isotope is related to cylinder cleaning operations and cascade component maintenance. PGDP conducted operations to recover 99Tc (April 1960 through June 1963) and ²³⁷Np (November 1958 through March 1962), which presents an increased potential to exposure from these isotopes (PACE and University of Utah 2000).

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Table 5-1 lists information on the isotopic species associated with PGDP operations and facilities and their dates of operation (when available).

Table 5-1. Radiological source term for PGDP processes and facilities.

Building	Process	Dates ^a	Radionuclides of concern ^b	Absorption type ^c
C-360	Product shipping and transfer	Dates	U-234, -235, -236, -238	F
0-300	Troduct shipping and transier		Np-237	M
			Pu-238, -239, -240, -242	M, S
			Tc-99	F
C-400	Converter maintenance	4/53-Present	U-234, -235, -236, -238	F
C-400		4/55-F1656111	Np-237	M
	Converter salvage	0/54 0/04		
	CIP/CUP Phase 1	9/54–6/61	Pu-238, -239, -240, -242	M, S
0.400/	CIP/CUP Phase 2	3/73–9/81	99-Tc	<u> </u>
C-400/	²³⁷ Np recovery	11/58–3/62	U-234, -235, -236, -238	F
C-710	⁹⁹ Tc recovery	4/60–6/63	Np-237	M
			Pu-238, -239, -240, -242	M, S
0.100		4070 4004	99-Tc	<u>F</u>
C-409	Converter refurbish	1973–1981	U-234, -235, -236, -238	F
			Np-237	М
			Pu-238, -239, -240, -242	M, S
			Tc-99	F
C-410	Green salt production	1953–1956	U-234, -235, -236, -238	F, M, S
	UF ₆ production	1953–Present	Np-237	M
	⁹⁹ Tc / ²³⁷ Np recovery (MgF ₂₎	9/61–6/63	Pu-238, -239, -240, -242	M, S
			99-Tc	F
			Th-230	S
			Am-241	M
C-420	Green Salt Production	1956-1977	U-234, -235, -236, -238	F, M, S
			Np-237	M
			Pu-238, -239, -240, -242	M, S
			Tc-99	F [']
			Th-230	S
			Am-241	M
C-331	Cascade ops/maintenance	9/52-Present	U-234, -235, -236, -238	F
C-333	Cascade ops/maintenance	9/52-Present	Np-237	M
C-335	Cascade ops/maintenance	4/54–Present	Pu-238, -239, -240, -242	M, S
C-337	Cascade ops/maintenance	7/54–Present	Tc-99	F.
C-310	Surge and product	9/52-Present	U-234, 235, 236, 238	F
0-310	⁹⁹ Tc / ²³⁷ Np recovery (MgF ₂)	1/63–6/63	Np-237	M
	107 Typ recovery (ivigi 2)	1/03-0/03	Pu-238, -239, -240, -242	M, S
			Tc-99	F
			Th-230	S S
C 24E	Curae and weets	0/F2 Dragant		5 F
C-315	Surge and waste	9/52-Present	U-234, -235, -236, -238	S S
0.040	Matal avaduation	40/57 40/00	Th-230	
C-340	Metal production	12/57–12/62	U-234, -235, -236, -238	M, S
		1/68–10/77	Np-237	M
			Pu-238, -239, -240, -242	M, S
0.700			Th-230	<u> </u>
C-720	Maintenance		U-234, -235, -236, -238	F, M, S
			Np-237	M
			Pu-238, -239, -240, -242	M, S
			Tc-99	F
			Th-230	S
			Am-241	M

Building	Process	Dates ^a	Radionuclides of concern ^b	Absorption type ^c
C-746	Waste metal smelting		U-234, -235, -236, -238 Np-237 Pu-238, -239, -240, -242 Th-230	M, S M M, S S
C-749	Uranium metal burial		U-234, -235, -236, -238 Np-237 Pu-238, -239, -240, -242 Th-230	M, S M M, S S

a. All dates are from DOE (2000) except those for C-409 and the Cascade Ops/Maintenance buildings, which are from PACE and University of Utah (2000) and ORAUT (2004a), respectively.

5.2.1 <u>Isotopic Concentrations</u>

At PGDP, monitoring for intakes of uranium, *in vivo* or *in vitro*, often resulted in reports of elemental uranium concentration in urine or the mass of elemental uranium in organs or the whole body. However, the internal dose assessment process requires the use of isotopic concentrations as input. As stated above, the chemical properties of the TRU and fission product contaminants resulted in changes in their relative concentrations in different parts of the process when compared to the original feed material. This section provides bounding estimates for radionuclide concentrations. The concentrations relative to the mass of total uranium and the activity of total uranium are shown in Tables 5-2 and 5-2a, respectively. A summary discussion on the information presented in the tables and its derivation is provided below. Details of units conversion and data sources can be found in Barton (2006).

Table 5-2. Bounding isotopic concentrations for PGDP operations (pCi/g U).^a

	1	2	3	4	5	6
Radionuclide	Pulverizer, ash handling, green salt C-410 pre-1983 ^b	Converter salvage line pre-1983 ^b	Converter salvage line post-1983 ^c	Tc/Np recovery operations, C-400 ^d	Balance of plant pre-1983 ^d	Balance of plant post-1983 ^c
Np-237	3.55E+04	1.61E+06	3.81E+04	1.76E+07	1.67E+03	3.67E+00
Pu-239/240	9.00E+05	3.24E+04	7.09E+04	4.42E+06	4.11E+01	1.95E+00
Pu-238	1.94E+05	7.01E+03	1.53E+04	9.54E+05	8.89E+00	4.21E-01
Pu-242	4.46E+01	1.61E+00	3.52E+00	2.19E+02	2.04E-03	9.67E-05
Pu-241	3.51E+07	1.27E+06	2.76E+06	1.72E+08	1.61E+03	3.38E-01
Am-241	1.56E+05	5.62E+03	1.85E+03	7.65E+05	7.13E+00	4.21E+00
Th-230	1.18E+06	1.67E+05	3.64E+05	2.27E+07	2.11E+02	1.00E+01
Tc-99	1.20E+05	1.20E+05	1.20E+05	3.81E+07	1.20E+05	1.20E+05
U-234	6.81E+05	6.81E+05	6.81E+05	6.81E+05	6.81E+05	6.81E+05
U-235	4.27E+04	4.27E+04	4.27E+04	4.27E+04	4.27E+04	4.27E+04
U-238	3.27E+05	3.27E+05	3.27E+05	3.27E+05	3.27E+05	3.27E+05
U-236	9.00E+02	9.00E+02	9.00E+02	9.00E+02	9.00E+02	9.00E+02

Based on uranium isotopic distribution for 2% enriched U (BJC 2000, Table 1-5). Assuming enriched as opposed to natural or depleted uranium provides bounding values for TRU intake estimates.

Pre-1983 Operations

Column 1, Pulverizer, Ash, Green Salt C-410; Column2, Converter Salvage

The values presented in Columns 1 and 2 are taken from PACE and University of Utah (2000) Table 7.9. The maximum air concentration data was used to provide bounding values for TRU

o. From Hill and Strom (1993, Table 16.2).

c. From BJC (1999, Table 1.7).

b. PACE and University of Utah (2000).

c. Hightower et. al. (2000).

d. BJC (2000).

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Table 5-2a. Bounding isotopic activity concentrations for PGDP operations (pCi/pCi U).

	1 1	2	2	<u>΄</u> Δ	5	6
Radionuclide	Pulverizer, ash handling, green salt C-410 pre-1983 ^b	Converter salvage line pre-1983 ^b	Converter salvage line post-1983°	Tc/Np recovery operations,	Balance of plant pre-1983 ^d	Balance of plant post-1983 ^c
Np-237	3.38E-02	1.53E+00	3.63E-02	1.67E+01	1.59E-03	3.49E-06
Pu-239/240	8.56E-01	3.08E-02	6.75E-02	4.21E+00	3.91E-05	1.86E-06
Pu-238	1.85E-01	6.67E-03	1.46E-02	9.08E-01	8.46E-06	4.01E-07
Pu-242	4.24E-05	1.53E-06	3.35E-06	2.08E-04	1.94E-09	9.20E-11
Pu-241	3.34E+01	1.21E+00	2.63E+00	1.64E+02	1.53E-03	3.22E-07
Am-241	1.48E-01	5.35E-03	1.76E-03	7.28E-01	6.79E-06	4.01E-06
Th-230	1.12E+00	1.59E-01	3.46E-01	2.16E+01	2.01E-04	9.52E-06
Tc-99	1.14E-01	1.14E-01	1.14E-01	3.63E+01	1.14E-01	1.14E-01
U-234a	4.89E-01	4.89E-01	4.89E-01	4.89E-01	4.89E-01	4.89E-01
U-235a	2.17E-02	2.17E-02	2.17E-02	2.17E-02	2.17E-02	2.17E-02
U-238a	4.89E-01	4.89E-01	4.89E-01	4.89E-01	4.89E-01	4.89E-01
U-236a	6.56E-04	6.56E-04	6.56E-04	6.56E-04	6.56E-04	6.56E-04

a. Based on uranium isotopic distribution for natural U (BJC 2000, Table 1-5). Assuming natural as opposed to enriched uranium provides bounding values for uranium intake estimates.

concentrations. The data in Table 7.9 is in units of uCi/cc which were converted to the desired units of pCi (TRU)/g (U) or pCi (TRU)/pCi(U). Details of this process can be found in Barton (2006).

The maximum plutonium activity in air is reported in PACE and the University of Utah (2000) as being comprised of ²³⁹Pu. However, it is more consistent with all references (BJC [2000], Hightower et. al. [2000]) to assume that the faction of the alpha activity attributed to ²³⁹Pu be considered as total alphaemitting isotopes plus ²⁴¹Am. A nominal isotopic distribution for TRU in fuel-grade plutonium provided in ORAUT (2005) was used to calculate the ratios of the various isotopes from the value provided in the reference.

Column 1 values are applicable to feed material production operations in C-410/C-420. Column 2 values are applicable to converter maintenance/salvage and cylinder cleaning operations in C-400.

Column 4, Tc/Np Recovery Operations; Column 5, Balance of Plant

The values in Column 4 and Column 5 are taken from BJC (2000), Table 2.4.1. This reference presents a detailed analysis of the mass balance of ²³⁹Pu, and ²³⁷Np using individual activity analysis. Table 2.4.1 presents the maximum concentrations (in ppb) of the ²³⁹Pu and ²³⁷Np for the various processes encountered at PGDP. The remaining plutonium isotopes and ²⁴¹Am concentrations are determined using the isotopic rations for fuel grade plutonium found it ORAUT (2005) and as described above.

The values in Column 4 are applicable to all Tc/Np recovery operations in C-400, **including those** that may have taken place after 1983.

The values for Column 5 consist of the upper 95% bound of the average Table 2.41 values (minus those areas/processes already accounted for in Columns 1 and 2). They are applicable to pre-1983 activities which include Cascade Operations (C-333, C-337, C-410), Production/Handling UF $_4$ (C-340), Connecting/Disconnecting UF $_6$ tails Cylinders (C-315, C-340), Connecting/Disconnecting Product Cylinders (C-310), Changing/Cleaning MgF $_2$ Traps (C-410, C-310), and U Metal Production (C-340) (BJC (2000), Table 2.4.1).

b. PACE and University of Utah (2000).

c. Hightower et. al. (2000).

d. BJC (2000).

Post-1983 Operations

Column 3, Converter Salvage; Column 6, Balance of Plant

Converter Salvage (Column 3) and converter salvage operations (Column 6) are based on Hightower et. al. (2000). These values were included in the TBD to account for the fact that an insignificantly small percentage (.008% of the total) of RU feed was introduced to the cascade after 1977 (BJC 2000, Appendix C). In addition, a two-phase (1954 to 1961 and 1973 to 1981) upgrade program effectively replaced the major components of the cascade [2]. These facts would imply that, over time, TRU and fission product contaminants would be greatly reduced from the process, with the exception of those entrained in the feed and tail cylinder heels. Only cylinder maintenance/washing operations would have a potential for exposure to these isotopes. Hightower et. al. (2000) contains analytical data in ppb for ²³⁷Np, ²³⁹Pu, and ²⁴¹Am in depleted uranium cylinders and feed material. The values in Table 3, the bounding concentrations for UF₆ feed, were used to represent Converter Salvage Operations (Column 3). As in the case for the Pre-1983 operations, these include converter maintenance/salvage and cylinder cleaning in C-400. The values for the remaining plutonium isotopes were determined as using the ratios in ORAUT (2005), as described above.

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For Balance of Plant Operations (Column 6) ²³⁹Pu, ²³⁷Np, and ²⁴¹Am values specified in Hightower et. al. (2000) Table 2 were used. These data represent the concentrations of dispersed contamination in DUF₆ cylinders and, considering the negligible quantity of RU introduced to the plant during this period, is believed to bound the actual ²³⁹Pu and ²³⁷Np levels since these isotopes were primarily concentrated in the pulverizer ash or plated within feed cylinder walls.

Column 6 is applicable to post-1983 activities which include Cascade Operations (C-333, C-337, C-410), Production/Handling UF $_4$ (C-340), Connecting/Disconnecting UF $_6$ tails Cylinders (C-315, C-340), Connecting/Disconnecting Product Cylinders (C-310), Changing/Cleaning MgF $_2$ Traps (C-410, C-310), and U Metal Production (C-340) (BJC (2000), Table 2.4.1).

Thorium 230 (²³⁰Th)

Pace and University of Utah (2000) list ²³⁰Th, a decay product of uranium as a potential source of concern and provide estimates for the activity concentration in Buildings C-400 and C-410 (Columns 1 and 2 of Table 5-2 and 5-2a). BJC (2000) and Hightower et. al. (2000) offers no data on ²³⁰Th. The ratio of Np-237 to Pu-239 data from each BJC (2000) Table 2.4.1 and Hightower et. al. (2000) was compared to the same ratio determined for the PACE "mixes". The mix for which the two ratios compared most closely was used to determine the ²³⁰Th concentration. A detailed discussion of this technique is provided in Barton (2006).

Technetium 99 (99Tc)

The ⁹⁹Tc concentration for all operations except ⁹⁹Tc recovery is assumed to remain constant throughout the process and is based on the reported 7-ppb maximum for the feed material (Smith 1984). The ⁹⁹Tc concentration for recovery operations is from BJC (2000).

Note: If the work location of a claimant is unknown, use Columns 2 or 3 in Table 5-2 (or Table 5-2a), as appropriate to the times of employment [3].

When determining potential intakes of TRU/99Tc by applying the isotopic ratios presented in Table 5-2 to urinalysis data, dose reconstructors should consider that PACE and University of Utah (2000) noted instances where the bioassay data in the electronic database are not consistent with original documents. Verify urinalysis results obtained from the electronic database against the written records.

5.3 IN VITRO MEASUREMENT METHODS

From the start of Plant operations in 1952, samples of urine from workers involved in enrichment operations were analyzed for uranium.² Over time, other workers were included in the monitoring program. In addition, special sampling occurred in response to incidents or issues (i.e., assessments of the radiological impacts of TRU elements and technetium); however, those analytical methods were typically performed off the PGDP site, at ORNL during early periods and later at analytical services contractor locations. Table 5-3 lists sampling frequencies and volumes for workers assigned to specific buildings during various periods. In addition, the table provides default values if the specific location of a claimant is not available [4].

Table 5-3. In vitro measurement frequencies. a, b

Period	Facility	Frequency	Volume
1953 to present	C-310	Every 4 weeks	Spot
1953 to present	C-315	Every 4 weeks	Spot
1953 to present	C-340	Every 4 weeks	Spot
1953 to present	C-400	Every 4 weeks	Spot
1953 to present	C-410	Every 4 weeks	Spot
1953 to present	Remainder	Yearly	Spot
1959 to present	All	Within 0.5 hr of incident	Spot
1960	C-410	Every 2 mo	Spot
1960	C-340	Every 3 mo	Spot
1960	C-331, C-333, C-335, C-337	Every 4 mo	Spot
1960	C-410	Every 6 mo	Spot
1991 to present	All	Every 4 weeks	24-hr collection or simulated 24-
			hr collection
Default	N.A.	Every 4 weeks	Spot

a. Sources: PACE and University of Utah (2000, Section 7.4).

At PGDP, routine urine samples were typically single voids collected during the middle of the week. Each could have been measured for specific gravity, pH, sugar, and albumin levels, as well as for uranium content. During the early years, total uranium concentrations were measured using a calibrated fluorimeter with a detection level of 0.005 mg/L (PACE and University of Utah 2000, p. 40).

In later years, total uranium content was assessed by kinetic phosphorescence analysis (KPA). Neither methodology included isotopic determinations (BJC 1999).

5.3.1 Measurement Types and Detection Levels

Table 5-4 lists the *in vitro* measurement types and detection levels during various periods. For samples analyzed at the site, the detection level for total uranium in urine was reported as 0.005 mg/L (PACE and University of Utah 2000, Section 4.2.1). If detection levels for specific methods were not found in the available references, levels specified as typical in ICRP (1989) were used [6].

In addition, if it is not clear from the monitoring records how or where a particular claimant's sample was analyzed, assume that they were analyzed in-house (i.e., at PGDP) and use the MDC from that

b. There is evidence that in 1960 the measurement frequency for Building C-410 and C-340 workers could have been either every 4 weeks or every 2 mo (C-410) or 4 mo (C-340). However, there is no referenceable indication whether there was a special class of worker to which the 2-mo frequency applied. The overlapping values have been left in the table.

² Fecal sampling was occasionally performed for special studies. However, a program for routine or diagnostic monitoring of fecal samples was never implemented [5].

Table 5-4. In vitro measurement types and detection levels for various periods.^a

Period	Measurement type	Radionuclide	MDCa	Comments
1952-1998	PGDP fluorimetry ^b	Total uranium	5 μg/L	
1985-1989	ORNL ^d	U-234, -235, and -238	0.03 pCi/sample	
1989-present	ORNL	U-234, -235, and -238	0.01 pCi/sample	
1989-present	Contractor ^e	U-234, -235, and -238	0.3 pCi/L	
1999-Present	KPA ^c	Total uranium	0.06µg/sample	DR should use the cite MDA for all urinalysis results by KPA, regardless of analysis date
1999-present	ORNL	Natural uranium	0.06 mg/sample	
1969-1985	PGDP gross beta ^c	Tc-99	10 dpm/ml	10 dpm/mL
1985-1989	ORNL	Isotopic plutonium	0.02 pCi/sample	
1989-present	ORNL	Isotopic plutonium	0.01 pCi/sample	
1985-1989	ORNL beta counting	Tc-99	18.1 pCi/sample	
1989-present	ORNL	Tc-99	90.9 pCi/sample	
1999-present	ORNL	Th-228, -230, and -232	0.014 pCi/sample	
1985 to 1989	ORNL	Np-237	0.04 pCi/sample	
1989-present	ORNL	Np-237	0.01 pCi/sample	
1985-1989	ORNL	Pu-238, -239, and -240	0.02 pCi/sample	
1989-present	ORNL	Pu-238, -239, and -240	0.01 pCi/sample	
1985-1989	ORNL	Am-241	0.02 pCi/sample	
1989-present	ORNL	Am-241	0.01 pCi/sample	

a. MDC = minimum detectable concentration.

measurement type to assess missed dose. Finally, if a record contains a notation of "less than X micrograms/L" or "< x pCi/sample", that value should reflect the MDC for that sample [7].

5.3.2 Reporting Formats and Codes

A variety of codes occurs on various urine bioassay records for PGDP. Table 5-5 summarizes those known at the date of this report, along with their interpretations.

5.4 IN VIVO MEASUREMENT METHODS

Whole-body counting and other *in vivo* methods were implemented beginning in the early 1950s. Until the present (2003), this measurement method was used primarily in response to incidents, or for assessing the magnitude of insoluble material intakes. Even when routine whole-body counting was instituted for certain PGDP employees in the late 1960s, the counting frequency was sporadic and seldom greater than once per year (PACE and University of Utah 2000).

5.4.1 <u>Measurement Types and Detection Levels</u>

At PGDP, whole-body counting was performed using a mobile counter provided by the Y-12 Plant (sometimes referred to as the MMES Counter) and at other facilities. Table 5-6 lists general information about the detection capabilities of this counting system for various periods.

5.4.2 Reporting Formats and Codes

A variety of codes and reporting formats appear in the *in vivo* bioassay records. Table 5-7 lists the known codes with their interpretations.

b. PACE and University of Utah (2000, p. 40) documents use of fluorimetry through 1982. No definitive reference for the specific method used for Total Uranium could be found in available documentation from 1983 -1998. It is favorable to the claimant to assume fluorimetry (as opposed to KPA) during this period.

c. BJC (1999), Table 3.9.

d. ORAUT (2004b) [8].

e. ICRP (1989).

Table 5-5. In vitro record codes. a,b,c,d

	Measurement			
Form identifier	type	Column identifier	Code	Interpretation
WCP-455	Urine bioassay	Reason for Visit	33, 35	Industrial health recheck
WCP-455	Urine bioassay	Reason for Visit	OB, 39, 35-1	Recall sample requested following elevated sample
WCP-455	Urine bioassay	Reason for Visit	MM recall	Monday morning recall sample; requested after days off work
WCP-455	Urine bioassay	Reason for Visit	32, 33, Term	Termination samples
WCP-455	Urine bioassay	Reason for Visit	30, 22	Rehire
WCP-455	Urine bioassay	Reason for Visit	37, 18, Per.	Periodic physical; confirmatory samples were collected during routine physicals; this typically did not pertain to those on routine monitoring program.
WCP-455	Urine bioassay	Reason for Visit	26	Preemployment
WCP-455	Urine bioassay	Reason for Visit	005, 07, 60, Exposure, Special, Release	Samples collected following exposure or potential exposure in uranium release or spill
WCP-455	Urine bioassay	Bottle No.	,	Permanent sample number
PGDP_HISTORICAL_URINE	Urine bioassay	Results		There is no distinction between positive results and detection limits.
PGDP_HISTORICAL_URINE	Urine bioassay	Sample Type	Physical	Routine physicals included collection of bioassay sample (random sampling program); this typically did not pertain to those on routine monitoring program.
WCP-455	Urine bioassay	Reason for Visit	I.H.R. or IHR	Industrial health recheck (associated with routine physicals)
WCP-455	Urine bioassay	Top of Card	"A"	Refers to shift worked ("A" = day shift)
WCP-455	Urine bioassay	Top of Card	"B"	Refers to shift worked ("B" = evening shift)
WCP-455	Urine bioassay	Top of Card	"O"	Refers to shift worked ("O" = midnight shift)
WCP-455	Urine bioassay	F, HG and OTHER	"B" (128)	Indicates shift and hours worked
PGDP_ANALIS_URINE	Urine bioassay	Results		Results are given in µg/L.
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	4	dpm/L
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	1	dpm/ml
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	2	dpm/day
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	3	dpm/sample
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	5	μg/ml
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	6	Bq/L
5EA HPINT - Bioassay Results Report	Urine bioassay	Units	7	Bq/day
5EA HPINT - Bioassay Results Report	Urine bioassay	Reason	3	Routine
5EA HPINT - Bioassay Results Report	Fecal analysis	Units	3	dpm/sample
5EA HPINT - Bioassay Results Report	Fecal analysis	Units	7	Bq/day
5EA HPINT - Bioassay Results Report	Breath analysis	Units	2	dpm/day
5EA HPINT - Bioassay Results Report	Breath analysis	Units	3	dpm/sample
5EA HPINT - Bioassay Results Report	Breath analysis	Units	4	dpm/L
5EA HPINT - Bioassay Results Report	Breath analysis	Units	6	Bq/L
5EA HPINT - Bioassay Results Report	Breath analysis	Units	7	Bg/day

Form identifier	Measurement type	Column identifier	Code	Interpretation
WCP-885	Urine bioassay	Schedule	A	Day sample is to be taken (1=Monday; 2=Tuesday; 3=Wednesday; 4=Thursday)
WCP-885	Urine bioassay	Schedule	В	Type analysis (1=Uranium; 2=Fluoride; 3=Mercury)
WCP-885	Urine bioassay	Location	Shifts	A, B, O and D
			Frequency	Uranium #1 = 1 a month; Fluoride #2 = 1 a year; Mercury #3 = Blank
Permanent log (sample collection log)	Urine bioassay	Number	Permanent sample number	Numbers are consecutive from top to bottom of page and continue from one page to next.
IBM Report Cards	Urine bioassay		Α	Name
IBM Report Cards	Urine bioassay		В	Badge
IBM Report Cards	Urine bioassay		С	Date (date shown on log)
IBM Report Cards	Urine bioassay		D	Code for this service
IBM Report Cards	Urine bioassay		Е	Analysis results in boxes labeled Uranium, Fluorides, and Mercury
UCN-5242, "Sample Analysis (Medical)."	Urine bioassay	Same as for NCP- 455	Same as for NCP-455	This form number was in use from 1969 to 1970.

- Sources: Maisler (2003); Eckerman and Ward (1992); Author unknown (no date 1). Form WCP-885 is referred to as "NCP-885" in Eckerman and Ward (1992); however, they appear to identify the same form.
- Around June 1956, form WCP-455 was modified with additional columns added. Individual bioassay records reviewed indicates the exact date the newly expanded form was used. This varies a little from person to person, but was in the middle of 1956 for all records reviewed.
- The bioassay records we have starting in 1977 (exact dates vary from person to person) are not copies of results recorded on bioassay cards; they are database printouts (e.g., "PGDP_Historical_Urine"). The uranium results are in units of µg/L with results recorded to the nearest integer, i.e., 0, 1, 2, etc.

Table 5-6. *In vivo* measurement types and detection levels for various periods.

				MDA		Action level
		Measurement		(units of	Action level	for work
Period	Equipment	type	Radionuclide	record)	for recount	restriction
1958	ORNL ^b	Lung	Pu-238	80 nCi	Not specified	Not specified
1960–1967	Y-12 ^c	Whole body	Np-237	2.7 nCi	Not specified	Not specified
1968–1980	Y-12 mobile counter ^d	Whole body	U-235	0.083 mg	Not specified	Not specified
1968–1980	Y-12 mobile counter ^d	Whole body	U-238	4 mg	Not specified	Not specified
1968–1980	Y-12 mobile counter ^d	Whole body	Np-237	17,000 pCi	Not specified	Not specified
1965–1991	Y-12 mobile counter ^d	Lung	Total uranium	4 mg	4 mg	27 mg
1965–1991	Y-12 mobile counter ^d	Lung	Enriched uranium (2% ²³⁵ U)	0.1 mg	0.1 mg	0.24 mg
1965–1991	Y-12 mobile counter ^d	Lung	Depleted uranium	4 mg	4 mg	37 mg
1965–1991	Y-12 mobile counter ^e	Lung	Np-237	200 pCi	1,700 pCi	17,000 pCi
1991–1995	Helgeson counter ^d	Lung	Total uranium	2 – 4 mg	2 – 4 mg	27 mg
1991–1995	Helgeson counter ^d	Lung	Enriched	0.04 - 0.07	0.04 - 0.07	0.24 mg
			uranium	mg	mg	
>1995	No counting perform	ied				

a. MDA = minimum detectable activity or amount. The MDAs shown for uranium, while given in units of mass, are presumed to have been based on measurement of Th-234 activities along with an assumed isotopic ratio. It is presumed that the results for enriched uranium are based on measurement of U-235 activities and an assumption of enrichment. However, these presumptions cannot be confirmed so cautious use of these MDA values is recommended.

- b. ORAUT (2004b).
- c. ORAUT (2006).
- d. ORAUT (2004c) [10].
- e. Hill and Strom (1993).

5.4.3 <u>Instructions for Addressing Possible Interferences and Uncertainties</u>

On occasion, *in vivo* measurement results included ¹³⁷Cs. However, those PGDP workers could have had body burdens of ¹³⁷Cs from nonoccupational sources (e.g., fallout and consumption of specific foodstuffs). There is no evidence of occupational intakes of ¹³⁷Cs at PGDP, so no dose of record should be associated with these measurement results, if any [9].

5.4.4 Assessment of Intake for Monitored Employees

In general, available urine results should be considered the primary method of dose reconstruction. *In vivo* measurements, especially in the earlier years of operation, were not used for routine monitoring purposes. However, those results might be useful for verifying assessments of dose based on urine bioassay results, in determining likely absorption types, or in providing upper and lower limits to the range of possible doses [11].

5.5 SIGNIFICANT INCIDENTS WITH INTERNAL DOSE POTENTIAL

During operations at PGDP, a number of incidents occurred that increased the potential for intakes of radioactive materials. If a claimant (or employee) recalls involvement in one or more of those

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Table 5-7. In vivo record codes.a

	Measurement	Column		
Form identifier	type	identifier	Code	Interpretation
In Vivo Radiation	Whole-body	Surface	Check mark,	Surface contamination on the subject was
Monitoring Report	count	contamination	yes	checked. If measurable activity was found, it
				was so noted on card.
In Vivo Radiation	Whole-body	Analysis		These align with Output - Analysis Sequence
Monitoring Report	count	Sequence		Results listed at bottom of card; they are not
				relevant to dose reconstruction process.
In Vivo Radiation	Whole-body	Output-Analysis	A. Enriched	The maximum U-235 enrichment was 2%
Monitoring Report	count	Sequence Results	Uranium	until 2000, after which it was 5%.
In Vivo Radiation	Whole-body	Output-Analysis	J. NLO	Refers to special spectrum region of interest
Monitoring Report	count	Sequence Results	Uranium	for National Lead of Ohio, early operator of
				Fernald facility.
In Vivo Radiation	Whole-body	Hand-written	No Np noted	Presence or absence of Np-237 was
Monitoring Report	count	notes		qualitatively evaluated.
In Vivo Radiation	Whole-body	Hand-written	BFD	Initials of whole-body counter operator
Monitoring Report	count	notes		(individual who filled out the card)
(with boxes for data				
entry)				
5EA HPINT-Bioassay	In vivo records	Units	M	μCi
Results Report				
5EA HPINT-Bioassay	In vivo records	Units	N	nCi
Results Report				
5EA HPINT-Bioassay	In vivo records	Units	Р	pCi
Results Report				
5EA HPINT-Bioassay	In vivo records	Units	D	dpm
Results Report				
5EA HPINT-Bioassay	In vivo records	Units	В	Bq
Results Report				
5EA HPINT-Bioassay	In vivo records	Units	U	μg
Results Report		Mand (4000). Authorium		

a. Sources: Maisler (2003); Eckerman and Ward (1992); Author unknown (no date 1).

incidents, dose reconstructors can use the information in Table 5-8 to identify the dates, location, and/or source term for an incident-specific dose assessment.

Table 5-8. A history of significant incidents and events.^a

Incident date	Incident description	Facility	Other information
1952–1990	Exposure to UF ₄ , UO, and process dust during guard patrolling	All buildings	
1952-1980	Exposure to uranium metal		
July 1953	First use of reactor tails		
November 1956	Fire	C-310	
1957–1977	Green salt, black oxides on floors and other surfaces	C-340	
December 1962	Fire	C-337	
March 1962	Explosion and fire	C-340	One fatality
April 1968	Worker overexposure	Unknown	Two workers overexposed
January 1978	Fire	C-315	
1958 to 1962	Cascade improvement program	C-331, C-333, C-335, and C-337	
1974 to 1982	Cascade improvement program	C-331, C-333, C-335, and C-337	
	Neptunium production	C-400	
1980 to 1982	Exposure to UF ₄ and uranium dust during drum crushing	C-746	

a. Source: PACE and University of Utah (2000).

5.6 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in the preceding text, bracketed callouts have been inserted to indicate information, conclusions, and recommendations to assist in the process of worker dose reconstruction. These callouts are listed in this section with information that identifies the source and justification for each item. Conventional references are provided in the next section that link data, quotations, and other information to documents available for review on the Oak Ridge Associated Universities (ORAU) Team servers.

- [1] Barton, Clark B. ORAU Team. Sr. Health Physicist. May 2006. The actual enrichment of uranium found in the process areas of PGDP is unknown because the facility produced product with a range of enrichments over the years. A nominal value of 2% was assumed because it is the approximate midpoint to the range, and provides a conservative (i.e., favorable to the claimant) result for the calculated values listed in Table 5-2 in comparison to natural uranium, which is assumed in the reference material (PACE and the University of Utah 2000; BJC 2000).
- [2] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006. The references PACE and the University of Utah (2000) and DOE (2000) include discussions stating that the CIP/CUP processes that ended in 1981 involved a cell-by-cell removal and replacement of the compressors and converters, process piping, and support system components. The year 1983 is an arbitrary time assumed to allow additional time to finish converter salvage line work, refurbishment of parts, etc.
- [3] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006.
 Column 2 data, based on PACE and the University of Utah (2000) values for the period before 1983, and Column 3 data, based on Hightower et al. (2000) values for the period after 1983, present the higher values for activities at the site that are not specific to one job or area.
- [4] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. February 2007. Extensive research during previous document development on this site showed that the majority of the bioassay results revealed a monthly (every 4 weeks) period for bioassay.
- [5] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. February 2007. Extensive research during previous document development on this site did not reveal any evidence of the implementation of fecal sampling.
- [6] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006.

 The nominal MDA values in ICRP (1989) are greater than those typically achievable by a laboratory using industry-accepted methods and techniques. Missed dose estimates will be greater with the use of the ICRP (1989) MDA value.
- [7] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006.
 The in-house MDC values are higher than offsite laboratory results, and any level indicated that is higher must be used. This is to ensure the use of the number most favorable to the claimant.
- [8] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006. Values for *in vitro* MDAs were often not available in references specific to PGDP. Because it is known that urine samples were sent to ORNL for analysis during the periods listed in Table 5-4, it was assumed that the values reported in ORAUT (2004b) would apply.

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- [9] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006. Extensive document review found no indication that ¹³⁷Cs had ever been considered an isotope of concern at Paducah beyond the occasional elevated individual due to dietary consumption and the increases due to global testing/incidents.
- [10] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006. Values for *in vivo* MDAs were often not available in references specific to PGDP. Because it is known that the Y-12 Mobile Counter was used for both the Paducah and Portsmouth sites for analysis during the periods listed in Table 5-4, it was assumed that the values reported in ORAUT (2004c) would apply.
- [11] Mantooth, Daniel S. ORAU Team. Sr. Health Physicist. May 2006. NIOSH (2002, Section 5.1) states: "Bioassay measurements are generally the most reliable data available for assessing internal exposures." In consideration of the limited *in vivo* data, those uses might be limited.

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GLOSSARY

absorption type

Categories for materials according to their speed of absorption in the body, which replaced the earlier inhalation clearance classes. Defined by the International Commission on Radiological Protection, the absorption types are F for fast absorption (formerly inhalation class D), M for moderate absorption (formerly inhalation class W), and S for slow absorption (formerly inhalation class Y). Also called solubility type.

activity median aerodynamic diameter (AMAD)

Diameter of a unit density sphere with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol. In relation to health physics, normally assumed to be 5 micrometers.

bioassay

Determination of kinds, quantities, or concentrations, and in some cases locations of radioactive material in a living body, whether by direct measurement (*in vivo* measurement) or by analysis and evaluation of materials excreted or removed from the body (*in vitro* measurement).

body burden

Amount of radioactive material in an individual's body at a particular point in time.

dose

In general, the effects of ionizing radiation in terms of the specific amount of energy absorbed per unit of mass. Effective and equivalent doses are in units of rem or sievert; other types of dose are in units of roentgens, rads, reps, or grays. Various terms narrow the type of dose, and some are additive:

- Absorbed dose is the amount of energy deposited in a substance by ionizing radiation.
- Collective dose is the sum of the doses to a specific population.
- Committed dose is the dose over time (usually 50 years for workers) to a specific organ or tissue from an intake of radioactive material.
- Cumulative dose is the sum of all doses to the same portion of the body or to the whole body over time.
- Deep dose is the dose at a 1-centimeter depth in tissue (1,000 milligrams per square centimeter).
- Effective dose is the sum of the equivalent doses in the principal tissues and organs of the body, each weighted by a tissue weighting factor that accounts for the probabilities of fatal and nonfatal cancers according to severity and the average length of life lost due to an induced cancer. It indicates the biological effect of the radiation exposure in that tissue.
- Equivalent dose or dose equivalent is the absorbed dose in a tissue or organ multiplied by a weighting factor for the particular type of radiation.
- Organ dose is the dose to a specific organ.

- Penetrating dose is that from higher energy photon (gamma and X-ray) radiation and neutron radiation that penetrates the outer layers of the skin. Nonpenetrating dose is that from beta and lower energy photon radiation.
- Personal dose equivalent is the dose equivalent in soft tissue below a specified point on the body at a specified depth.
- Shallow dose is the dose at a 0.07-centimeter depth in tissue (7 milligrams per square centimeter).
- Skin dose is the dose to the skin.
- Whole-body dose is the dose to the entire body excluding the contents of the gastrointestinal tract, urinary bladder, and gall bladder.

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dose equivalent (H, DE)

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

exposure

(1) In general, the act of being exposed to ionizing radiation. See acute exposure and chronic exposure. (2) Measure of the ionization produced by X- and gamma-ray photons in air in units of roentgens. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air.

insoluble material

A term loosely used to describe the relative degree of solubility of a material in body fluids. Recognizing that no material is absolutely insoluble, the terms low solubility or poorly soluble are preferable.

intake

Radioactive material taken into the body by inhalation, absorption through the skin, injection, ingestion, or through wounds. Depending on the radionuclide involved, intakes are in units of mass, activity, or potential alpha energy.

internal dose or exposure

Dose received from radioactive material in the body.

internal dose assessment

Estimation of an intake of radioactive material and the consequent internal radiation dose based on measurements in the work environment and/or bioassay.

in vitro

Of or relating to a process that takes place under artificial conditions or outside a living organism (e.g., in the laboratory). From Latin meaning in alass.

in vivo

Of or relating to a process that takes place in a living organism. From Latin meaning in life.

lung solubility type

See absorption type.

minimum detectable amount (MDA)

Lowest amount of radioactive activity or substance amount detectable by a specific instrument or process. Smallest amount or activity of a radionuclide in a sample or organ that yields a result above the detection level with a specific probability of a Type II (false negative) error while accepting an specific probability of a Type I (false positive) error.

minimum detectable concentration (MDC)

Lowest concentration of a material in a substance (e.g., urine) detectable by a specific instrument or process. Minimum detectable activity (or amount) in units of concentration.

minimum reporting level

Level below which an analytical dose is not recorded in the worker's dose record, usually based on a site-specific policy decision. The recording level is not necessarily the same as the minimum detectable amount or activity for that measurement. Also called less-than value, minimum reportable dose, minimum recordable or recording dose, recording level, and reporting level.

monitoring (personnel)

Periodic or continuous determination of the presence or amount of ionizing radiation or radioactive contamination in air, surface water, groundwater, soil, sediment, equipment surfaces, or personnel (for example, bioassay or alpha scans). In relation to personnel, monitoring includes internal and external dosimetry including interpretation of the measurements.

occupational dose

Internal and external ionizing radiation dose from exposure during employment. Occupational dose does not include that from background radiation or medical diagnostics, research, or treatment.

radiation

Subatomic particles and electromagnetic rays (photons) that travel from one point to another, some of which can pass through or partly through solid materials including the human body. Radiation, as used in this document, does not include nonionizing radiation, such as radio- or microwaves, or visible, infrared, or ultraviolet light.

reactor tails

Recycled uranium (typically UO₃) from reactor operations (typically Savannah River and Hanford) that contains traces of transuranic isotopes not removed during chemical processing (e.g., reduction-oxidation).

recording level

See minimum reporting level.

rem

Traditional unit of radiation dose equivalent that indicates the biological damage caused by radiation equivalent to that caused by 1 rad of high-penetration X-rays multiplied by a quality factor. The average American receives 360 millirem a year from background radiation. The

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sievert is the International System unit; 1 rem equals 0.01 sievert. The word derives from roentgen equivalent in man; rem is also the plural.

routine monitoring

Monitoring carried out at regular intervals during normal operations.

sievert (Sv)

International System unit for dose equivalent, which indicates the biological damage caused by radiation. The unit is the radiation value in gray (equal to 1 joule per kilogram) multiplied by a weighting factor for the type of radiation and a weighting factor for the tissue; 1 Sv equals 100 rem.

special monitoring

Monitoring in addition to the routine monitoring program carried out for special purposes such as estimating the amount of radionuclide deposited in a person after a known or suspected accidental intake or after a known or suspected environmental release.

spot sample

In relation to bioassay, usually a single void of urine.