

<p><b>ORAU Team</b>  <b>NIOSH Dose Reconstruction Project</b></p> <p>Technical Basis Document for the Hanford Site -- Occupational Internal Dose</p>	<p>Document Number:  ORAUT-TKBS-0006-5  Effective Date: 10/15/2003  Revision No.: 00  Controlled Copy No.: _____  Page 1 of 46</p>
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**TABLE OF CONTENTS**

<u>Section</u>	<u>Page</u>
Record of Issue/Revisions .....	3
Record of Issue/Revisions .....	3
Acronyms and Abbreviations .....	4
5.1 Occupational Internal Dose .....	5
5.2 In Vitro Minimum Detectable Activities, Analytical Methods, and Reporting Protocols .....	10
5.2.1 Plutonium .....	11
5.2.2 Americium .....	16
5.2.3 Curium .....	18
5.2.4 Tritium .....	18
5.2.5 Uranium .....	20
5.2.6 Fission Product Analysis .....	24
5.2.7 Strontium .....	25
5.2.8 Promethium .....	26
5.2.9 Polonium .....	27
5.2.10 Neptunium .....	28
5.2.11 Other Limited-Exposure Radionuclides .....	28
5.3 In Vivo Minimum Detectable Activities, Analytical Methods, and Reporting Protocols .....	29
5.3.1 Whole Body Counters .....	29
5.3.2 Chest Counters .....	33
5.3.3 Thyroid Counters .....	35
5.3.4 Head Counters and Other Counts .....	36
5.3.5 General Notes about Items in the Database .....	36

5.4	Mixtures .....	37
5.4.1	Reactor workers:.....	38
5.4.2	Separations Plants.....	38
5.4.3	Waste Management Facilities (tank farms, evaporators, transfer lines).....	40
5.5	Interferences, Uncertainties .....	40
5.5.1	Contamination of Samples .....	40
5.5.2	Uncertainties.....	40
5.6	Workers with No Confirmed Intakes.....	42
5.6.1	Special Consideration for Plutonium, Americium, and Thorium.....	42
5.6.2	Worst Case Chronic Intakes .....	42
5.7	Unmonitored Workers .....	43
	References.....	45
	Glossary.....	46

### LIST OF TABLES

<u>Table</u>	<u>Page</u>	
5.1-1	Air sample data.....	5
5.1-2	Codes and radionuclides associated with bioassay at Hanford.....	7
5.2.1-1	Routine plutonium urinalysis detection levels.....	12
5.2.1-2	MDAs for nonroutine Pu excreta analyses .....	13
5.2.1-3	Activity composition of Hanford reference weapons-grade plutonium mixture.....	14
5.2.1-4	Activity composition of Hanford reference fuel-grade plutonium mixture .....	15
5.2.1-5	Activity composition of Hanford reference commercial power fuel-grade plutonium mixture .....	15
5.2.2-1	Routine <sup>241</sup> Am urinalysis detection levels .....	17
5.2.2-2	MDAs for nonroutine <sup>241</sup> Am excreta analyses .....	17
5.2.3-1	Routine Cm urinalysis detection levels.....	18
5.2.3-2	MDAs for nonroutine Cm excreta analyses.....	18
5.2.4-1	Routine tritium urinalysis detection levels .....	19
5.2.5-1	Radiological characteristics of Hanford uranium mixtures .....	20
5.2.5-2	Impurities in recycled uranium at Hanford.....	21
5.2.4-3	Inhalation class for Hanford uranium compounds .....	21
5.2.5-4	Routine uranium urinalysis detection levels .....	23
5.2.6-1	Routine fission product urinalysis detection levels .....	24
5.2.7-1	Routine <sup>90</sup> Sr urinalysis detection levels.....	26
5.2.8-1	Routine <sup>147</sup> Pm urinalysis detection levels.....	27
5.2.9-1	Routine <sup>210</sup> Po urinalysis detection levels .....	28
5.2.9-2	MDAs for nonroutine <sup>210</sup> Po excreta analyses .....	28
5.3.1-1	Routine whole body counting detection levels.....	31
5.3.1-2	Mean body burdens of <sup>137</sup> Cs from fallout in the United States.....	33
5.3.2-1	Routine chest counting detection levels .....	34

### RECORD OF ISSUE/REVISIONS

<b>ISSUE AUTHORIZATION DATE</b>	<b>EFFECTIVE DATE</b>	<b>REV. NO.</b>	<b>DESCRIPTION</b>
Draft	10/02/2003	00-A	New document to establish TBD for occupational internal dose – section 5. Initiated by Edward D. Scalsky.
10/15/2003	10/15/2003	00	First approved issue. Initiated by Edward D. Scalsky.

## ACRONYMS AND ABBREVIATIONS

CEDE	Committed Effective Dose Equivalent
CF	Commercial Fuel
DU	Depleted Uranium
GeLi	Lithium drifted Germanium (detector)
GI	Gastro Intestinal
GOK	God Only Knows
HIE	Hanford Internal Exposure (database)
HPGe	High Purity Germanium (detector)
ICRP	International Commission on Radiological Protection
INEEL	Idaho National Environmental Engineering Laboratory
KPA	Kinetic Phosphorescence Analysis
LEPD	Low Energy Photon Detector (also computer code to indicate use of the LEPD)
MDA	Minimum Detectable Activity or, for elemental uranium, Minimum Detectable Amount
MPBB	Maximum Permissible Body Burden
MPC	Maximum Permissible Concentration
NCRP	National Council on Radiation Protection and Measurements
NU	Natural Uranium
ORE	Occupational Radiological Exposure (database)
PNL	Pacific Northwest Laboratory
PNNL	Pacific Northwest National Laboratory
RDA	Reliably Detectable Activity
REX	Radiological Exposure (database)
RU	Recycled Uranium
TPU	Total Propagated Uncertainty
TRU	Transuranic
TTA	Thenoyl trifluoroacetone
UST	United States Testing Company

## 5.1 OCCUPATIONAL INTERNAL DOSE

When the first reactor was started on the Hanford site, there were no programs to monitor an employee for internal dose, with the exception of measuring particles in the air. The site was operating three reactors, a fuel manufacturing facility and four processing plants from 1943 to 1946 before a bioassay program was in place. The responsibility of personnel monitoring was with the Medical Department. For this time frame, air sampling data, environmental data and incident data will likely be the only information available to use for recreating personnel exposure. Information about air sample results was highlighted in monthly reports of the Health Instrument Department. These were very brief summaries, mostly highlighting problems; often no values were listed, indicating air concentrations were below concern. Air sample data from the reactors were almost never listed in these reports; the radiation protection emphasis at the reactors seemed to be external dose and effluents in the water. The records show that high air concentrations at the other facilities prompted use of respiratory protection. Table 5.1-1 summarizes the air sample data found so far.

Table 5.1-1. Air sample data.

Year	Facility	Max concentration ( $\mu\text{Ci}/\text{cm}^3$ )	Most sampler concentrations ( $\mu\text{Ci}/\text{cm}^3$ )
1943	Building 305 Test Reactor		
1944	B Reactor		
	D Reactor		
	T Plant		
1945	T,U,B Canyon Bldgs	8E-12 Pu	<8E-13 Pu
	T,U,B Concentrator Bldgs (224T,U,B)	2E-11 Pu	<2E-12 Pu
	D Reactor		
	B Reactor		
	F Reactor		
	231-Z	One at 8E-11 Pu, very temporary and area immediately placed on mask; most highs were about 8E-12 Pu or less.	<8E-13 Pu
	300 Area Labs	6E-12 Pu	<8E-13 Pu
1946 <sup>a</sup>	Metal Fab. Bldgs.	1E-9 Unat	<2E-10 Unat
	T,U,B Canyon Bldgs.	4E-12 Pu <sup>b</sup>	
	T,U,B Concentrator Bldgs.		
	D Reactor		
	B Reactor		
	F Reactor		
	231-Z	4E-11 Pu	8E-13 Pu
	300 Area Metal Fab. Bldgs.	2E-9 Unat	<1E-10 Unat
	3706 Bldg.	6E-12 Pu, 5E-10 Unat	<2E-12 Pu
	200 W Laundry	1E-11 Pu	4E-12 Pu

a. Based on monthly reports for July and September through December only.

b. Excluding one incident in B canyon involving only two workers, for which special urine samples were obtained.

Air sample data were not routinely reported in the Health Instrumentation Section monthly reports for the reactors. There were three exceptions in 1946: 1) a high air sample of  $3 \times 10^{-9} \mu\text{Ci}/\text{cm}^3$  beta activity when a gasket around a thimble blew, 2)  $6.5 \times 10^{-9} \mu\text{Ci}/\text{cm}^3$  beta activity at 100 D – listed as the highest value for the year, 3)  $6 \times 10^{-8} \mu\text{Ci}/\text{cm}^3$  beta activity for a task at 100 F but workers were wearing respirators.

Considerable (hundreds per month) thyroid scans were being done for workers in the separation (canyon) buildings during this time. Concern was for  $^{131}\text{I}$  uptake for workers who entered the canyons, such as crane operators. No information was reported as to instruments used, MDAs, or results. The 1946 annual summary report stated that nothing significant was detected in any of the thyroid scans, without stating what was considered significant. However, the tolerance level for  $^{131}\text{I}$  in air had been established in October 1945 as  $1 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$  (Cantril 1945) based on a permissible equilibrium amount in the thyroid of 2  $\mu\text{Ci}$ . Based on other statements in the monthly reports, it is reasonable to assume that scans showing thyroid burdens over 2  $\mu\text{Ci}$  would have been considered significant.

The Health Instrument Section Monthly reports do mention contamination spreads in the reactor buildings, 231-Z, concentrator buildings, and uranium metal fabrication shops during these years, so intakes undoubtedly were occurring. Large intakes of plutonium would have been detectable in later years when bioassay was available; but some level of chronic intake during this period is a reasonable assumption. Chronic intakes of uranium in the metal fabrication shops should also be assumed. The tolerance air concentration for uranium machining was  $1.5 \times 10^{-4} \mu\text{g}/\text{cm}^3$ , which converts to  $1.1 \times 10^{-10} \mu\text{Ci}/\text{cm}^3$ . Most air sample data for the "Metal Fabrication Buildings" were simply listed as less than that level.

Because there is little information concerning intakes in the years prior to implementation of routine bioassay programs, the following default assumptions should be made unless there is better information in the worker's file.

**Reactor workers, 1944-49:** Assume intakes of 200 nCi per year each of  $^{46}\text{Sc}$ ,  $^{51}\text{Cr}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$ . (See Section 5.4.1 for absorption types.) (These values differ from assumed values for later years, Section 5.4.1, reflecting improved radiation protection programs over the years.)

**Separations plants, 231-Z, 1944-46:** Assume intakes of plutonium alpha (6%Pu mix, see Table 5.2.1-3) absorption class M at  $8 \times 10^{-13} \mu\text{Ci}/\text{cm}^3$  for 7 hr per day, 5 days per week. Assuming a breathing rate of  $1.5 \text{ m}^3/\text{hr}$ , this translates to 6 pCi per day chronic intake. This chronic intake would apply from either the first day of work for the worker or the start up of the plant (December 1944 for T Plant and April 1945 for B Plant). Assume absorption type M. Also assume chronic intake of  $^{131}\text{I}$  of  $1.3 \times 10^{+6}$  pCi per day (type F, 5  $\mu\text{m}$  AMAD), which will produce equilibrium thyroid burdens at just under the 2- $\mu\text{Ci}$  tolerance level. [When IMBA can handle vapors, the better intake value is  $7.5 \times 10^{+5}$  pCi per day assuming a type F SR-1 vapor (ICRP 1997, Table A.6.20).] The iodine intake, excluding environmental radioiodine, would only apply to workers that entered the canyons or perhaps the main stack sampling buildings, but it's unlikely the dose reconstructor will be able to differentiate these workers from general workers at the separations plants.

**300 Area uranium fabrication buildings (313, 314), 1944-47:** Assume chronic intakes of 1500 pCi per day natural uranium (based on assumed average air concentrations of  $2 \times 10^{-10} \mu\text{Ci}/\text{cm}^3$  and other parameters described above). (See Section 5.2.5 for the isotopic composition.). Assume absorption type S.

**Laundry 1944-46:** Assume chronic intakes of plutonium alpha (6% Pu mix, absorption class M) of 34 pCi per day based on an air concentration of  $2 \times 10^{-12} \mu\text{Ci}/\text{cm}^3$ , 8 hr per day, and other parameters described above. This intake assumes exposure to the soiled laundry for the entire day. If interviews indicate that part of the time was spent at the washing station or handling the cleaned laundry, the intake may be reduced by the ratio of hours spent handling clean laundry/8.

According to the history compiled by R. H. Wilson (1987), one of the priority tasks for a special studies group formed in 1944 was to determine a way to measure plutonium in the body. Limits on the

amount of plutonium in the body were set as early as 1944, and, after experimentation with various methods, routine urine sampling and analysis for plutonium was initiated in 1946. Urinalysis for uranium seems to have started in 1946 also and was well established by 1948. Urinalysis for fission products started in this time frame as well, although the Wilson document indicates that separation from <sup>40</sup>K was not always successful prior to 1949. Since then, monitoring for numerous radionuclides has occurred at Hanford because of the complex scope of work over the years, the many research projects, special “campaigns,” etc. Additionally, numerous techniques have been used because of improvements in techniques. The major sources of intakes have been plutonium, <sup>241</sup>Am either as an ingrown contaminant in the plutonium or as a separated waste product, uranium, fission products, activation products, and tritium. But the records as a whole list a wide spectrum of radionuclides that were monitored and an even longer list of codes used to identify either the radionuclides, groups of radionuclides, specific measurement techniques, or combinations of radionuclides and techniques. Many of the radionuclides apply to a small set of workers on a research project or to workers whose tasks “might” have exposed them to lots of different sources, for instance, radiation monitoring technicians.

Table 5.1-2 provides a fairly exhaustive list of codes for analyses that can be encountered in the bioassay or internal dosimetry records for Hanford workers. Some of the codes were used for scheduling bioassay but not for reporting results of the bioassay. For instance, IPA is a code for performing plutonium and americium separation chemistry and alpha spectrometry on an excreta sample, but the results would normally be reported separately for <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Am. However, if the sample was not obtained or the results could not be reported due to analysis problems, the record will just show the IPA code with a reason for not obtaining a result. Other codes refer to a type of in vivo count or a special type of sample analysis. For instance, LEPD is the code for performing an x-ray/gamma-ray analysis on an excreta sample using the low-energy photon detector (a thin window germanium detector); however, if anything was detected, the actual radionuclide was reported. The code GOK (God only knows) shows on in vivo count hardcopy records during the 1960s and 70s. This refers to net counts per minute from an undetermined source in a low-energy region of the spectrum from NaI-based whole body counters.

Table 5.1-2. Codes and radionuclides associated with bioassay at Hanford.

Code	Description	Comment
AAAA1	Americium	Probably Am-241
AAAA2	Americium	Probably Am-241
AAAA3	Americium	Probably Am-241
AAAA4	Americium	Probably Am-241
AAAA5	Americium	Probably Am-241
AAAA6	Americium	Probably Am-241
AAAA7	Americium	Probably Am-241
AC225	Actinium 225	
ACS	Actinium 227, thorium 227	Scheduling code
AC227	Actinium 227	
AC228	Actinium 228	
AG110	Silver 110	
AM241	Americium 241	
AM242	Americium 242	
AM243	Americium 243	
BA140	Barium 140	
BETA	Beta	
BI213	Bismuth 213	
BI214	Bismuth 214	
BK249	Berkelium 249	

Code	Description	Comment
BR 82	Bromine 82	
C 14	Carbon 14	
CE141	Cerium 141	
CE143	Cerium 143	
CE144	Cerium 144	
CF249	Californium 249	
CM242	Curium 242	
CM244	Curium 244	
CO 58	Cobalt 58	
CO 60	Cobalt 60	
CR 51	Chromium 51	
CS134	Cesium 134	
CS137	Cesium 137	
EU152	Europium 152	
EU154	Europium 154	
EU155	Europium 155	
EU156	Europium 156	
EV155	?	Probably a typographical error for Eu-155 that got left in the database
EV156	?	Probably a typographical error for Eu-156 that got left in the database
FE 59	Iron 59	
FP	Fission products	
GA	Gross alpha	
GB	Gross beta	
GELI	Gamma-GeLi detector	Excreta scheduling code for a gamma scan with a germanium detector
GOK	God only knows	See text
GS	Gamma NaI detector	Excreta scheduling code for a gamma scan with a NaI detector
H 3	Tritium	
I 125	Iodine 125	
I 129	Iodine 129	
I 131	Iodine 131	
I 133	Iodine 133	
IAM	Isotopic americium	Excreta scheduling code for americium separation and alpha spectrometry
ICA	?	Probably scheduling code for americium and curium via alpha spectrometry
ICM	Cm isotopic	Excreta scheduling code for curium isotopes via alpha spectrometry
IEU	Eu isotopic	Excreta scheduling code for europium separation and isotopic analysis
IPA	Isotopic Pu and Am241	Excreta scheduling code
IPIU	Isotopic Pu, isotopic U	Excreta scheduling code
IPS	Isotopic Pu and Sr	Excreta scheduling code
IPSA	Isotopic Pu, Sr tot & Am241	Excreta scheduling code; Sr tot means radiostrontium by gross beta
IPSR	Seq Pu isotopic Sr-total	Excreta scheduling code for isotopes of Pu and radiostronium
IPU	Isotopic plutonium	Excreta scheduling code
IPUB	Plutonium isotopic, Pu241	Excreta scheduling code; Pu-241 separate anal. by beta counting
IPUBA	Plutonium isotopic, Pu241, Am241	Excreta scheduling code
IPUL	Low level isotopic Pu	Pu-238 and Pu-239 using a 10,000 minute count

Code	Description	Comment
IRA	Radium isotopic	Excreta scheduling code
IR192	Iridium 192	Excreta scheduling code
ISCP	Sequential Sr 90 Ce Pm	Excreta scheduling code
ISPEC	Gamma spectroscopy	Excreta scheduling code
ISR	Sr isotopic	Excreta scheduling code
ITH	Thorium isotopic	Excreta scheduling code
ITPAC	Seq isotopic Pu, Cm & Am241	Excreta scheduling code
IU	U isotopic	Excreta scheduling code
IUPU	Isotopic Plutonium/U-natural	Excreta scheduling code
K 40	Potassium	
LA140	Lanthanum 140	
LEPD	Low energy photon detector	Excreta scheduling code for low-energy photon scan
MFP	Mixed fission products	
MN 54	Manganese 54	
MO 99	Molybdenum 99	
NA 22	Sodium 22	
NA 24	Sodium 24	
NAI	Gamma NaI detector	Excreta scheduling code
NB 95	Niobium 95	
NP237	Neptunium 237	
NP239	Neptunium 239	
PB210	Lead 210	
PB212	Lead 212	
PM147	Promethium 147	
PO210	Polonium 210	
PR144	Praseodymium 144	
PU	Plutonium alpha	Total alpha from Pu isotopes after separation
PUMIX	Plutonium alpha	Total alpha from Pu isotopes and Am-241
PU238	Plutonium 238	
PU239	Plutonium 239	When pertaining to excreta samples, it's actually Pu-239+240
PU240	Plutonium 240	
PU241	Plutonium 241	
PU242	Plutonium 242	
QUS	U	Quick Uranium Soluble; excreta scheduling code for elemental U
QUS 1	U	Same as QUS
QUS 2	U	Same as QUS
RA224	Radium 224	
RA225	Radium 225	
RA226	Radium 226	
RA228	Radium 228	
RH106	Rhodium 106	
RND	Radon daughters	
RU103	Ruthenium 103	
RU106	Ruthenium 106	
S 35	Sulfur 35	
SB124	Antimony 124	
SB125	Antimony 125	
SCP	Sequential Sr-total Ce Pm	Excreta scheduling code
SM153	Samarium 153	
SR	Strontium	Total radiostrontium by beta counting
SR 89	Strontium 89	
SR 90	Strontium 90	When pertaining to excreta samples, Sr-90 by yttrium ingrowth

Code	Description	Comment
TAC	Total actinides	
TC 99	Technetium 99	
TH227	Thorium 227	
TH228	Thorium 228	
TH230	Thorium 230	
TH232	Thorium 232	
TH234	Thorium 234	
TL208	Thallium 208	
U	Elemental uranium	
URAN	Elemental uranium	
U DEP	Depleted uranium	
U NAT	Natural uranium	
U 233	Uranium 233	See uranium discussion in text
U 234	Uranium 234	Actually U-234 + 233, but usually U-234
U 235	Uranium 235	
U 236	Uranium 236	
U 238	Uranium 238	
UMIX	Uranium mix	Total uranium, used for intakes not bioassay
UMS	U 235 U 236 U 238 U 234	
US	U	
XX 0	Isotope will have no result	
ZN 65	Zinc 65	
ZR 95	Zirconium 95	

Other bioassay codes have been used to indicate the

- sample type,
- *in vivo* count body location,
- reason for the sample/count,
- type of kit and some details about the sampling protocol,
- laboratory used,
- laboratory turnaround time versus analytical sensitivity,
- units associated with the result, and
- reason for not obtaining a valid excreta result or *in vivo* count.

In addition there are codes pertaining to the nature of the intake, including

- reason for an intake assignment,
- source of intake (as in at Hanford or other site),
- nature of intake, and
- mode of intake.

Tables listing and explaining these codes are provided in Attachment D.

## 5.2 IN VITRO MINIMUM DETECTABLE ACTIVITIES, ANALYTICAL METHODS, AND REPORTING PROTOCOLS

Most urinalysis records have, at some time, been entered into the electronic database(s). However, for some of the earliest urinalysis records, cases have been discovered where not all records were included in the electronic database. For any case where urinalysis might have been obtained prior to 1974, the hardcopy file for the case should be thoroughly reviewed for urinalysis results that might be

missing in the electronic database. The Hanford Internal Exposure (HIE) database was implemented in 1974, followed by the Occupational Radiological Exposure (ORE) database in 1983, and the Radiological Exposure (REX) database in 1993. In principal the REX database has all the information from the previous databases, but as stated above there may be isolated situations where some data never got into a database or some data did not get transferred from one database to another.

There is another anomaly found in the results circa 1946-1950. There is a urinalysis record with no result and no volume. This might indicate that the sample was not turned in or the analysis failed; however, experience has shown that this convention was also used to indicate a result that was a non-detection. In many cases the actual laboratory urinalysis results card is available in the worker's file and would show if the analysis was performed but the results were below detection or not.

Home sampling began very early in the program (1946) and has continued throughout the history of Hanford. Home sampling was used to prevent contamination of samples in the workplace.

In vitro analyses were performed in house until the breakup of the main Hanford contractor (General Electric) occurred in 1965. At that time the DOE-Richland Office established a contract for in vitro analyses with the United States Testing Company, which built and operated a commercial low-level radiochemistry lab in north Richland until 1990. The responsibility for awarding and overseeing the contract was subsequently transferred to Battelle as operators of the Pacific Northwest Laboratory. Except for a period between 1990 to 1992, despite a series of competitive procurements, in vitro analyses have been performed in the same facility since 1965. However, due to buyouts and mergers, the name of the laboratory has changed in the following sequence: United States Testing, International Technology Analytical Services, Quanterra Environmental Services, and Severn Trent Laboratories (present).

Battelle defaulted the contract with United States Testing in June 1990, and subsequently routine samples were collected and frozen (Lyon 1991, Lyon 1992). Between September and November 1990 temporary contracts/agreements were established and samples were being analyzed at the following laboratories: Los Alamos National Lab (plutonium), TMA-Norcal (strontium), PNL-Analytical Chemistry Lab [325 Building] (tritium), and Westinghouse Hanford Company [222-S Building] (elemental uranium). In February 1991, IT Analytical Services commenced analyses for plutonium, americium, curium, and isotopic uranium. Los Alamos National Lab was replaced by Oak Ridge National Lab and Reynolds Electric and Engineering Company at the Nevada Test Site (plutonium) in April 1991. The contract with IT Analytical Services replaced the former contract with United States Testing, but the other labs continued to process samples until the backlog was worked off. So the work at the temporary labs was finishing up during late 1991 through early 1992 with the last results being received in March 1992.

### **5.2.1 Plutonium**

By far the most serious intakes at Hanford involved plutonium and <sup>241</sup>Am. Routine urinalyses for plutonium started in September 1946. The first plutonium bioassay analysis consisted of lanthanum fluoride precipitation and thenoyl trifluoroacetone (TTA) extraction and gross alpha counting. Electrodeposition on a stainless steel disk combined with nuclear track emulsion (autoradiography) started in December 1952. Detection levels for these and subsequent procedures are listed in Table 5.2.1-1. The definition of "detection level" no doubt changed over the years, but the levels in Table 5.2.1-1 fit reasonably with the concept of limit of detection or MDA. For example, the Wilson history states, "From statistical evaluations of data collected in 1953, the true detection limit with nuclear-track film was determined. These evaluations showed 0.05 dpm was achievable within reasonable confidence levels. Occasionally recovery, counting, etc., allowed detection levels to be as low as

0.028 dpm and for a short period, a level of 0.027 dpm was reached and used as the detection level. This practice [of recording lower detection levels] was discontinued and the more conservative 0.05 dpm was used routinely even though lower levels were possible part of the time.

Table 5.2.1-1. Routine plutonium urinalysis detection levels.

Period	MDA, dpm/ sample	Decision Level, dpm/sample	Measured Quantity
Prior to June 1949	0.66		total Pu alpha
6/1949 to 11/1952	0.33		"
12/1952 to 1/27/53	0.18		"
1/28/53 to 3/26/53	0.15		"
3/27/53 to 11/06/53	0.05		"
11/07/53 to 12/04/53	0.07		"
12/53 to 4/55	0.057		"
5/55 to 8/55	0.027		"
9/55 to 9/55	0.04		"
10/55 to 9/30/83	0.05 <sup>a</sup>	0.025 <sup>a</sup>	"
10/01/83 to 12/31/83	0.035		Each Pu-238, Pu-239
1/02/84 to 4/88	0.02		"
5/88 to 5/90	0.02	0.01	"
6/90 to 11/91	0.03	0.015	"
11/91 to 4/2000	0.02	0.01	"
5/2000 to 8/2001	0.02	$X_b + 2.05x TPU^b$	"
9/2001 to present	0.02	$2 x TPU^b$	"

- a. During part of this period, results that were less than the detection limit were reported as 0.025. But if net activity above background and above 0.025 was detected the actual amount was recorded.
- b.  $X_b$  is mean of blanks and TPU is total propagated uncertainty.

Prior to October 1983 the recorded value was the total alpha activity from plutonium so would have included  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{240}\text{Pu}$ . Any  $^{241}\text{Pu}$  or  $^{241}\text{Am}$  present in the urine would not have been accounted for by the recorded results.

The results may have been reported as Pu or  $^{239}\text{Pu}$ , but until October 1983, the result was really the total alpha activity from isotopes of plutonium. Results on plutonium urinalysis sheets were recorded in units of dpm/sample, but the same results were recorded in units of  $\mu\text{Ci}/\text{sample}$  in the electronic database. The units in the electronic database should have a unit code of 5, meaning  $\mu\text{Ci}/\text{sample}$ , but if the code is missing or unreadable, the units are still recognizable because the exponent is normally  $-7$  or  $-8$ . A value of  $1.1 \times 10^{-8}$  was recorded for results for which plutonium was not detected (one half of the nominal 0.05 dpm MDA). This method of recording was used through 1974. In 1975 the units were changed to dpm/sample (unit code 1) and 0.025 was recorded for results for which plutonium was not detected.

In October 1983 several changes were made. The lanthanum fluoride/TTA method was replaced by the use of anion exchange columns, alpha spectrometry analysis replaced autoradiography, and chemical yield was established for each sample separately by use of a  $^{242}\text{Pu}$  tracer. The results of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  have been reported separately since then. A 2,500 minute counting time has been used since 1984. A 10,000-minute count time was introduced for special situations in 1996 but its use was rare.

Starting in the mid 1990s the fecal procedure was enhanced to ensure improved oxidation of highly insoluble plutonium. Added steps included wet ashing with hydrogen peroxide and fusion with hydrogen fluoride. This procedure was tested with special high-fired plutonium oxide samples from INEEL and found to work very well.

Fecal samples were usually not analyzed in total (were aliquoted after muffling, dry ashing, and wet ashing); hence, more than one analysis result for a given sample was possible and will often be found in the database.

The MDAs listed from 1983 to present are nominal MDAs based on contractual requirements. Generally the lab performed slightly better than the contractual MDA, but the true MDA varied slightly over time and the contractual MDA was a reliable estimate. Reporting of errors, which was the total propagated uncertainty including uncertainty associated with the determination of chemical yield, counting efficiency determination, and systematic errors, began in 1981. The implementation of a distinction between an MDA (type I and type II errors) and a decision level (type I error) occurred in April 1989. Initially a fixed value of 0.01 dpm/sample was used for all results, being one half the nominal MDA. The decision level was allowed to become sample-specific based on the total propagated uncertainty in 2000, and an adjustment was made to the formula in 2001.

The MDAs listed in Table 5.2.1-1 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5.2.1-2.

Table 5.2.1-2. MDAs for nonroutine Pu excreta analyses.

Period	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sample	
	Emergency <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite
1/1965 to 10/1983 <sup>b</sup>	0.9-1.5	NA	0.1-0.15	0.5-0.7	NA
10/1983 to 1/1985 <sup>c</sup>	9	NA	0.2	0.5	NA
1/1985 to 6/1990	9	3	0.2	0.5	0.08
6/1990 to 2/1991 <sup>d</sup>	20	4	(c)	2	0.4
2/1991 to present	9	3	0.2	0.5	0.08

- At times the emergency category was called "rush" and the routine category was called "normal."
- MDAs varied according to sample size over the range shown; the lower value was generally applicable except for very large samples. MDAs for this period apply to total Pu alpha.
- MDAs from this time forward apply to Pu-238 and Pu-239 separately.
- Emergency and expedited processing of urine and fecal samples was available through PNNL's Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite labs but the MDA was not established, probably about 0.2-0.5 dpm/sample considering state-of-the-art of those labs.

Normally, fecal sampling was done in response to suspected intakes; however, routine fecal sampling was used for some high risk plutonium workers, mostly operators at PUREX and the Plutonium Finishing Plant, from 1986 through June 1989. The special study showed that, when considered as a group, the mean fecal excretion was statistically significantly different from controls. Enhanced air sampling, initiated in response to the study, showed frequent-intermittent releases of plutonium in the workplaces, at levels below the detectability of normal air sampling. When modeled as chronic intake, the intakes and doses were low (less than 10 mrem committed effective dose equivalent), and were documented in the workers' records. (Bihl, 1993; Lyon et al 1988; Lyon et al 1989) When encountered in the workers' records, these fecal samples should be interpreted as chronic intakes, not as acute intakes, occurring many days prior to the sample dates.

Except for a few standards in radiochemistry laboratories, plutonium at Hanford was comprised of a mix of radionuclides, namely <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu and <sup>241</sup>Pu. The activity of <sup>242</sup>Pu in plutonium mixtures at Hanford was too small to contribute significantly to dose. Hanford plutonium mixtures were categorized by their weight percent of <sup>240</sup>Pu. When the reactors were operated with the purpose of producing plutonium for weapons, the target mixture was about 6% <sup>240</sup>Pu, a mixture referred to as weapons grade. N Reactor was also operated to produce electrical power for a local public power

company. When operated to produce power, the mixture in the fuel rods when removed from the reactor was nominally 12% <sup>240</sup>Pu, a mixture referred to as fuel grade. At any given time, individual fuel rods would have mixtures differing from these, as would individual batches of rods starting at the front end of the fuel rod dissolution and plutonium extraction processes. However, when refined and blended, the target mixture was the weapons grade mixture. Tables 5.2.1-3 and 5.2.1-4 lists the relative activities of plutonium isotopes and <sup>241</sup>Am, which grows in from <sup>241</sup>Pu, for 6% <sup>240</sup>Pu and 12% <sup>240</sup>Pu mixtures (from Carbaugh 2003). In these tables “aging” refers to the time since the <sup>241</sup>Am was separated from the plutonium then starts to build in again from decay of <sup>241</sup>Pu.

The values in these tables can help determine the total intake of plutonium and <sup>241</sup>Am if there are limited data concerning the composition of the source of the intake. For instance, only in rare, large intakes was <sup>241</sup>Pu measured as part of the intake so the activity of that isotope is almost never available. <sup>241</sup>Am at time of intake was also often not determined directly. Since 1983, <sup>238</sup>Pu and <sup>239+240</sup>Pu were measured separately so the ratio of one to the other can be used to estimate the category of the plutonium mixture and, from the tables, to estimate the activities of <sup>241</sup>Pu and <sup>241</sup>Am. Prior to 1983, the measured quantity was total alpha from plutonium, which means the total of <sup>238</sup>Pu and <sup>239+240</sup>Pu. So unless <sup>241</sup>Am was measured or there is other information about the intake, there may be no way to tell from the bioassay how much <sup>241</sup>Pu and <sup>241</sup>Am were present at intake.

**Most plutonium mixtures handled at Hanford were nominally weapons grade, and if the <sup>239</sup>Pu to <sup>238</sup>Pu ratio implies weapons grade the ratios in Table 5.2.1-3 should be used. However, lacking any helpful information about the intake, an assumption of 10-year-old fuel grade plutonium mixture would be claimant-favorable and reasonable. For intakes since about 1996, 20-year-old fuel grade mixture could be assumed.**

Table 5.2.1-3. Activity composition of Hanford reference weapons-grade plutonium mixture.

Mixture designation:	Fresh	5-Year	10-Year	15-Year	20-Year	25-Year	30-Year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
<sup>238</sup> Pu	8.56E-03	8.23E-03	7.91E-03	7.60E-03	7.31E-03	7.03E-03	6.75E-03
<sup>239</sup> Pu	5.77E-02						
<sup>240</sup> Pu	1.36E-02						
<sup>241</sup> Pu	8.24E-01	6.48E-01	5.09E-01	4.00E-01	3.15E-01	2.48E-01	1.95E-01
<sup>242</sup> Pu	1.97E-06						
<sup>241</sup> Am	0	5.83E-03	1.04E-02	1.39E-02	1.66E-02	1.87E-02	2.03E-02
<sup>239+240</sup> Pu	7.13E-02	7.13E-02	7.13E-02	7.13E-02	7.12E-02	7.12E-02	7.12E-02
Pu-alpha	7.99E-02	7.95E-02	7.92E-02	7.89E-02	7.85E-02	7.83E-02	7.80E-02
Total alpha	7.99E-02	8.53E-02	8.96E-02	9.28E-02	9.52E-02	9.70E-02	9.83E-02
Activity Ratios							
<sup>239+240</sup> Pu: <sup>241</sup> Am	NA	12.2	6.87	5.13	4.28	3.80	3.50
<sup>239+240</sup> Pu: <sup>238</sup> Pu	8.33	8.67	9.01	9.38	9.74	10.1	10.5
<sup>241</sup> Pu: <sup>239+240</sup> Pu	11.6	9.09	7.15	5.62	4.42	3.48	2.73
Pu alpha: <sup>239+240</sup> Pu	1.12	1.20	1.26	1.30	1.34	1.36	1.38
Pu alpha: <sup>238</sup> Pu	9.33	9.66	10.0	10.4	10.7	11.1	11.6
Pu alpha: <sup>241</sup> Am	NA	14.6	8.63	6.67	5.72	5.18	4.84
<sup>241</sup> Pu: Pu alpha	10.3	8.15	6.43	5.07	4.01	3.17	2.50

a. Time since separation of <sup>241</sup>Am from the Pu mix.

There was at least one project in the 1970s involving irradiated fuel rods from commercial power reactors (Nuclear Waste Vitrification Project). The 324 and 325 Buildings in the 300 Area were involved. Commercial fuel rods have a much higher degree of “burnup,” and the ones at Hanford were characterized by much more <sup>241</sup>Pu and nominally 26% <sup>240</sup>Pu. Table 5.2.1-5 provides the activity characteristics of the commercial fuel used in the Nuclear Waste Vitrification Project. In addition, the Plutonium Finishing Plant sometimes recycled plutonium from other DOE sites. This material would

be rich in <sup>241</sup>Am. Plutonium from the West Valley commercial reprocessing site is also stored at Hanford. But unless the records concerning the specific intakes being investigated have evidence of these unusual mixtures, the default mixtures mentioned above should be used.

Table 5.2.1-4. Activity composition of Hanford reference fuel-grade plutonium mixture.

Mixture designation:	Fresh	5-Year	10-Year	15-Year	20-Year	25-Year	30-Year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
<sup>238</sup> Pu	1.71E-02	1.64E-02	1.58E-02	1.52E-02	1.46E-02	1.40E-02	1.35E-02
<sup>239</sup> Pu	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.25E-02
<sup>240</sup> Pu	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.71E-02	2.71E-02
<sup>241</sup> Pu	3.09E+00	2.43E+00	1.91E+00	1.50E+00	1.18E+00	9.29E-01	7.30E-01
<sup>242</sup> Pu	3.93E-06						
<sup>241</sup> Am	0	2.19E-02	3.89E-02	5.22E-02	6.24E-02	7.03E-02	7.63E-02
<sup>239+240</sup> Pu	7.98E-02	7.98E-02	7.98E-02	7.97E-02	7.97E-02	7.97E-02	7.97E-02
Pu-alpha	9.69E-02	9.62E-02	9.56E-02	9.49E-02	9.43E-02	9.37E-02	9.32E-02
Total alpha	9.69E-02	1.18E-01	1.35E-01	1.47E-01	1.57E-01	1.64E-01	1.69E-01
Activity ratios							
<sup>239+240</sup> Pu: <sup>241</sup> Am	NA	3.64	2.05	1.53	1.28	1.13	1.04
<sup>239+240</sup> Pu: <sup>238</sup> Pu	4.67	4.86	5.05	5.24	5.46	5.69	5.90
<sup>241</sup> Pu: <sup>239+240</sup> Pu	3.87	3.05	2.40	1.88	1.48	1.17	9.16
Pu alpha: <sup>239+240</sup> Pu	1.21	1.21	1.20	1.19	1.18	1.18	1.17
Pu alpha: <sup>238</sup> Pu	5.67	5.87	6.05	6.24	6.46	6.69	6.90
Pu alpha: <sup>241</sup> Am	NA	4.39	2.46	1.82	1.51	1.33	1.22
<sup>241</sup> Pu: Pu alpha	31.9	25.3	20.0	15.8	12.5	9.91	7.83

a. Time since separation of the <sup>241</sup>Am from the Pu mix.

Table 5.2.1-5. Activity composition of Hanford reference commercial power fuel-grade plutonium mixture.

Mixture designation:	Fresh	5-Year	10-Year	15-Year	20-Year	25-Year	30-Year
Years of aging <sup>a</sup> :	0	5	10	15	20	25	30
Specific activity in mixture (Ci/g)							
<sup>238</sup> Pu	1.71E-01	1.64E-01	1.58E-01	1.52E-01	1.46E-01	1.40E-01	1.35E-01
<sup>239</sup> Pu	3.41E-02						
<sup>240</sup> Pu	5.90E-02	5.89E-02	5.89E-02	5.89E-02	5.89E-02	5.88E-02	5.88E-02
<sup>241</sup> Pu	1.34E+01	1.05E+01	8.28E+00	6.51E+00	5.12E+00	4.03E+00	3.17E+00
<sup>242</sup> Pu	1.97E-04						
<sup>241</sup> Am	0	9.49E-02	1.69E-01	2.26E-01	2.79E-01	3.04E-01	3.31E-01
<sup>239+240</sup> Pu	9.31E-02	9.31E-02	9.30E-02	9.30E-02	9.29E-02	9.29E-02	9.29E-02
Pu-alpha	2.65E-01	2.58E-01	2.52E-01	2.45E-01	2.39E-01	2.34E-01	2.28E-01
Total alpha	2.65E-01	3.53E-01	4.20E-01	4.71E-01	5.10E-01	5.38E-01	5.59E-01
Activity ratios							
<sup>239+240</sup> Pu: <sup>241</sup> Am	NA	0.981	0.551	0.411	0.344	0.305	0.281
<sup>239+240</sup> Pu: <sup>238</sup> Pu	0.544	0.568	0.589	0.612	0.636	0.664	0.688
<sup>241</sup> Pu: <sup>239+240</sup> Pu	144	113	89.1	70.0	55.1	43.3	34.1
Pu alpha: <sup>239+240</sup> Pu	2.85	2.77	2.71	2.63	2.57	2.52	2.45
Pu alpha: <sup>238</sup> Pu	1.55	1.57	1.59	1.61	1.64	1.67	1.69
Pu alpha: <sup>241</sup> Am	NA	2.72	1.49	1.08	0.857	0.770	0.689
<sup>241</sup> Pu: Pu alpha	50.6	40.7	32.9	26.6	21.4	17.2	13.9

b. Time since separation of the Am 241 from the Pu mix.

If some of the plutonium bioassay was obtained prior to October 1983 and some after, the two data sets are not compatible. For a first approximation, for curve fitting in IMBA and POC determination in IREP, the Pu alpha data can be treated as <sup>239</sup>Pu and for the post 1983 data, the <sup>239</sup>Pu and <sup>238</sup>Pu values can be summed and treated as <sup>239</sup>Pu. However, the intakes of <sup>241</sup>Pu and <sup>241</sup>Am must be included in the dose determination for input into IREP. If the POC is marginally close to the 50%

criterion, then the total Pu alpha intake (as  $^{239}\text{Pu}$ ) should be split out into actual intakes of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  because the dose conversion factors are not the same.

Most plutonium at Hanford was in moderately soluble form, e.g. nitrates, which can be modeled as inhalation (absorption) type M. But many forms were possible over the years, especially metal and oxides. Even material, such as old contamination, that was originally in soluble form has a tendency to oxidize when left in contact with air, such as old contamination. Oxides, metal, and old contamination should be treated as inhalation type S.

If nothing is known about the chemical form of the plutonium, then either type M or S can be used. If there are sufficient bioassay data to determine the type by curve fitting, use the best fit; otherwise use the type that is most claimant favorable, i.e., that maximizes the dose to the organ of concern.  $^{241}\text{Am}$  that is a component of plutonium contamination should be modeled in the lung the same as the plutonium matrix in which it has ingrown. In other words the americium should be treated as absorption type S if the plutonium is type S.

### 5.2.2 Americium

Americium was usually a trace contaminant in plutonium mixtures as discussed in section 5.2.1. However, because americium was separated from plutonium at the reprocessing plants (e.g., T Plant and S Plant (REDOX) in the early years, PUREX from 1956) and at the Plutonium Reclamation Facility (a wing in the Plutonium Finishing Plant), waste tanks, transfer lines, and a whole operation in the Plutonium Finishing Plant had  $^{241}\text{Am}$  that was chemically separate from plutonium. This americium should be treated as americium (as opposed to trace americium atoms bound in a plutonium matrix). The ICRP recommended inhalation type for americium is M.

It has not been discovered yet when americium analyses first started. There is no mention of americium excreta analysis in the 1948 report by Jack Healy, "Bioassay at Hanford;" no mention in a 1954 memo, "Bioassay Annual Report," that lists numbers of urinalyses for plutonium, fission products, and uranium; no mention in a compilation of bioassay procedures, given a title of "Bioassay Procedures and Analysis (Old Bioassay Bible)," but no author or editor, dated April 10, 1961. The first documentation found so far is a memo to file from John J. Jech, Senior Development Engineer in the Personnel Dosimetry Services, dated September 1969, that states that per a telephone conversation with Matt Lardy at the U. S. Testing Company, the new detection limit for  $^{241}\text{Am}$  is 2.0 dpm/sample as of July 10, 1969. Matt Lardy's personal recollection provided some confirmation that this might have been the earliest date for americium urinalyses. The procedure was described as DDCP extraction to a planchet and gross alpha counting. A letter was found from Matt Lardy to Harold Larson, manager of Personnel Dosimetry Services, dated March 27, 1974, stating that the new limit for  $^{241}\text{Am}$  in urine is 0.1 pCi/sample at the 90% confidence limit. This limit was still listed in a statement of work with U.S. Testing in 1979 and again in 1982, although it was therein stated as 0.2 dpm/sample. In the laboratory statement of work for a new contract starting October 1983, the detection level was listed as 0.04 dpm per sample. This was achieved by use of an alpha/gamma coincidence counter.

Until October 1983 the gross alpha count could have included  $^{242}\text{Cm}$  or  $^{244}\text{Cm}$  if any were associated with the intake. Assuming that the results are  $^{241}\text{Am}$  is claimant favorable. However, sometime between October 1983 and October 1985, both the chemistry procedure and the counting technique were changed. The chemistry method was similar to that described in the HASL-300 manual and commonly referred to as the "RICH-RC-50-80" method. This method involved sequential precipitation with calcium oxalate and iron hydroxide, removal of plutonium using anion exchange, loading on another column with nitric acid and methanol, and elution of the americium with HCl and methanol. Electrodeposition and counting by alpha spectrometry were also implemented at this time. The MDA

in the 1985 statement of work was listed as 0.02 dpm/sample consistent with the change to alpha spectrometry, and it has stayed there to the present. Presently, Eichrom TRU column exchange is used for the separation of the americium for urine; however, the MDA is the same.

Table 5.2.2-1 summarizes what has been uncovered concerning <sup>241</sup>Am MDAs for routine urinalysis.

Table 5.2.2-1. Routine <sup>241</sup>Am urinalysis detection levels.

Period	MDA, dpm/sample	Decision level, dpm/sample
7/1969 to 2/1974	2.0	Anything detected
3/1974 to 10/1983	0.2	"
10/1983 to 9/1985	0.04	"
10/1985 to 05/1988	0.02	"
05/1988 to 06/1990	0.02	0.01
07/1990 to 10/1991	0.03	0.015
11/1991 to 4/2000	0.02	0.01
5/2000 to 8/2001	0.02	$X_b + 2.05 \times \text{TPU}^a$
9/2001 to present	0.02	$2 \times \text{TPU}$

a.  $X_b$  is the mean of the blanks and TPU is total propagated uncertainty.

The MDAs listed in Table 5.2.2-1 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5.2.2-2. These analyses may have been used because of suspected intakes of pure <sup>241</sup>Am (such as the famous explosion of an americium exchange column at the Plutonium Finishing Plant in 1976) or to determine the activity of <sup>241</sup>Am in a

Table 5.2.2-2. MDAs for nonroutine <sup>241</sup>Am excreta analyses.

Period	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sample	
	Emergency <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite
1/1967 to 2/1974	(b)	NA	(b)	(b)	NA
2/1974 to 1981	(c)	NA	4	(c)	NA
1982 to 9/1983	3.6-12 (3.6 most probable)(d)	NA	1.2-5.0 (1.2 most probable)(d)	0.7-1.0 (0.7 most probable)(d)	NA
10/1983 to 9/1985	200	NA	0.16	1.0	NA
10/1985 to 6/1989	20	6	0.1	1	0.08
7/1989 to 10/1991 <sup>e</sup>	20	4	NA	2	0.4
11/1991 to present	20	6	0.1	1	0.08

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Probably available but MDAs not found.

c. Emergency analyses were available on request, but the statement of work (based on 1978 SOW) did not specify the MDAs. It implied that an MDA about 10 times the routine (or priority for fecal) MDA was expected.

d. Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples.

e. Emergency and expedited processing of urine and fecal samples was available through PNNL's Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite labs but the MDA was not established, probably about 0.2-0.5 dpm/sample considering state-of-the-art of those labs.

plutonium mixture. There is evidence of a few intakes of pure <sup>241</sup>Am prior to 1969, involving usual circumstances such as using a supposedly sealed source that had ruptured. These intakes were analyzed by urinalysis so obviously a procedure existed at that time, although not part of the contract with U.S. Testing. On rare occasions for a serious intake, samples were analyzed for <sup>241</sup>Am using a

low-energy photon detector, prior to any chemistry. This technique came into existence in 1986 or 87. Its detection level was about 5 dpm/sample. Generally, the LEPD result was just used as a rapid indicator, and a more accurate result was obtained by wet chemistry/alpha spectrometry days later.

### 5.2.3 Curium

The curium isotopes of concern were 242 and 244, although sources of curium at Hanford were minor, usually calibration sources or as minor constituents in an actinide mixture. The curium and americium procedure were the same so the results would have been reported as curium only if so requested through the bioassay request system, until alpha spectrometry was initiated. After 1985, the chemistry is the same as americium, but <sup>241</sup>Am, <sup>242</sup>Cm and <sup>244</sup>Cm were reported separately if requested. The MDAs were not always identical with <sup>241</sup>Am, however. Routine urinalysis MDAs for curium are provided in Table 4.2.3-1 and non-routine excreta analyses are provided in Table 4.2.3-2.

Table 5.2.3-1. Routine Cm urinalysis detection levels.

Period	MDA, dpm/sample	Decision level, dpm/sample
7/1969 to 1981	Not specifically mentioned	
1982 to 9/1983	Listed for emergency processing only	
10/1983 to 4/1988	0.02	Anything detected
5/1988 to 6/1990	0.02	0.01
6/1990 to 10/1991	0.03	0.015
11/1991 to 4/2000	0.02	0.01
5/2000 to 8/2001	0.02	$X_b + 2.05 \times \text{TPU}^a$
9/2001 to present	0.02	2 x TPU

a.  $x_b$  is the mean of the blanks and TPU is total propagated uncertainty.

Table 5.2.3-2. MDAs for nonroutine Cm excreta analyses.

Period	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sample	
	Emergency <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite
Prior to 1982	(b)	NA	(b)	(b)	NA
1982 to 9/1983	10 <sup>c</sup>	NA	NA	0.5-1.0 (0.5 most probable) <sup>cd</sup>	NA
10/1983 to 9/1985	240	NA	0.8	10	NA
10/1985 to 6/1989	240	70	0.8	1	1.2
7/1989 to 10/1991 <sup>e</sup>	NA	NA	NA	NA	NA
11/1991 to present	240	70	0.8	1	1.2

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Probably available but MDAs not found.

c. Total alpha; would have included any americium present also.

Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples.

### 5.2.4 Tritium

The history of tritium urinalysis at Hanford is not well documented. Tritium urinalysis was not mentioned at all in the Wilson history of personnel dosimetry (Wilson 1987). The earliest report found to date on tritium urinalysis at Hanford dates to 1949 by Jack Healy, the leading internal dosimetrist at Hanford for many years (and apparently an instrumentation expert as well). That procedure was based on "production of acetylene from the active water, with subsequent measurement of the ionization caused by the tritium beta particle" (Healy 1949). No detection level was mentioned in that letter, but one was mentioned in an internal memo from Herbert M. Parker to A.B. Greninger, dated

January 1950, that referred to the acetylene method for urinalysis and provides a sensitivity of about 1.2  $\mu\text{Ci/L}$  in water (Parker 1950). However, that method apparently didn't work well because Healy stated in a 1951 letter to H.F. Schultz at Los Alamos that, "Your problem on the determination of tritium in the urine samples is one that we have been working on for the last two years, and have finally obtained what appears to be a decent method for routine use" (Healy 1951). The copy of the letter is of such poor quality that the method described is hard to follow, but it definitely was not liquid scintillation counting. A 1961 report entitled "The Estimation of Whole Body Dose from Tritium by Urine Analysis" indicated that liquid scintillation was used by that time, but again no detection level was given. Liquid scintillation counting was implemented for tritium bioassay at the Savannah River Site in 1958 and it is reasonable to expect that Hanford did so at about the same time. In the previously mentioned interview with Matt Lardy, Mr. Lardy stated that liquid scintillation counting of a 1-ml aliquot of raw urine has been used since U.S. Testing was awarded the bioassay contract in 1965.

Tritium intakes were accounted for as part of external dose until about 1986 or 87, when they were entered in the dose database as an internal dose.

Basically tritium was not a major source of radionuclide exposure for large numbers of workers at Hanford. A 1967 report states, "Battelle-Northwest and its predecessor at Hanford, the General Electric Company, have been involved in activities with tritium since about 1950, initially as a manufactured product for weapons applications and later as a by-product of heavy water reactor operations. Our most recent experience is from operation of the Plutonium Recycle Test Reactor (PRTR)" (McConnon, 1967). There was also some work on a tritium target program in the 1990s in the 300 Area and tritium light sources in the 1980s (involving just a few people), and there has been low-level use of tritium as a tracer in various biology experiments. Tritium exposure was assumed to be chronic during the exposure period, unless a very large acute intake was known to occur.

Tritium was referred to as P-10 in the 1950s. The main source of tritium in the 1950s was 108-B, also called the P-10 Plant, which started in August 1949.

Very little data on MDAs has been discovered. A 1964 letter to the PRTR Radiation Monitoring personnel (McConnor, 1964) states that a tritium bioassay result exceeding 5  $\mu\text{Ci/L}$  will reported to the Radiation Monitoring Office the day after the samples are picked up, indicating a level of concern probably well above the MDA. One P-10 Personnel Sample Analysis card, with entries in 1952, shows several values below 5  $\mu\text{Ci/L}$  with the smallest value being 2.5  $\mu\text{Ci/L}$ . None of the values are listed as less-thans. The 1965 statement of work with US Testing shows an MDA of 1  $\mu\text{Ci/L}$  (which is consistent with the MDA at Savannah River Site throughout the 1950s). Table 5.2.4-1 provides MDAs for routine tritium urinalysis as best has been compiled to date. From 1978 to present the MDAs were obtained from statements of work with the bioassay laboratory; the MDAs and time periods prior to that are guesses.

Table 5.2.4-1. Routine tritium urinalysis detection levels.

Period	MDA
1949 through 1960 <sup>a</sup>	$\approx 5 \mu\text{Ci/L}$
1961 through 1981	1 $\mu\text{Ci/L}$
1982 through 10/1991	10 dpm/ml
11/1991 to present	20 dpm/ml

a. Dates and MDA are best guesses. The change in 1961 was based on earliest reference to liquid scintillation counting.

### 5.2.5 Uranium

Uranium exposure at Hanford involved principally three physical forms: depleted (DU), natural (NU), and slightly enriched that was also called recycled uranium (RU). Small numbers of researchers may have experimented with more enriched uranium at different times, e.g. metallurgy on commercial grade fuel, but such exposure would have been to small groups for limited periods. Table 5.2.5-1 provides the default uranium mixtures (Carbaugh 2003). Generally, personnel working in the production facilities (e.g., fuel fabrication, the reactors, fuel dissolution and plutonium processing, waste management) were exposed to natural uranium during operation of the early reactors (through about 1958) and recycled uranium starting in 1957 (fuel fabrication shops) or 1958. Recycle uranium also had impurities build up and track with the uranium over time. Impurities can be approached in two ways, representative levels based on averages of several measurements at different times and upper limits based on tolerance specifications (e.g., not to exceed). Both of these approaches are given in Table 5.2.5-2.

Table 5.2.5-1. Radiological characteristics of Hanford uranium mixtures.

<b>Uranium mixture</b>				
<b>Weight percentage<sup>a, b</sup></b>	<b>Natural (NU)</b>	<b>Depleted (DU)</b>	<b>Recycled (RU)</b>	<b>Commercial fuel (CF)</b>
<sup>234</sup> U	0.0057	0.0005	0.0082	0.0300
<sup>235</sup> U	0.7204	0.2500	0.9700	2.9600
<sup>236</sup> U	Negligible	Negligible	0.0680	Negligible
<sup>238</sup> U	99.2739	99.7500	98.9500	97.0100
<b>Specific constituent activity in mixture (uCi/g, nCi/mg, or pCi/ug)<sup>c</sup></b>				
<sup>234</sup> U	0.3563	0.0313	0.5125	1.8750
<sup>235</sup> U	0.0156	0.0054	0.0210	0.0639
<sup>236</sup> U	Negligible	Negligible	0.0440	Negligible
<sup>238</sup> U	0.3336	0.3352	0.3325	0.3260
Total	0.7054	0.3718	0.9099	2.2649
<b>Specific constituent activity in mixture (dpm/ug)<sup>c</sup></b>				
<sup>234</sup> U	0.7909	0.0694	1.1378	4.1625
<sup>235</sup> U	0.0345	0.0120	0.0465	0.1419
<sup>236</sup> U	Negligible	Negligible	0.0977	Negligible
<sup>238</sup> U	0.7405	0.7441	0.7381	0.7236
Total	1.5659	0.8254	2.0200	5.0281
<b>Constituent fraction of total uranium activity in mixture</b>				
<sup>234</sup> U	0.5051	0.0840	0.5632	0.8279
<sup>235</sup> U	0.0221	0.0145	0.0230	0.0282
<sup>236</sup> U	Negligible	Negligible	0.0484	Negligible
<sup>238</sup> U	0.4729	0.9014	0.3654	0.1439
Total	1.0000	1.0000	1.0000	1.0000

- NU, DU, and CF data from Rich et al. 1988.
- RU data based on average of data presented by Sula, Carbaugh, and Bihl 1991.
- Can be used to represent specific alpha activity in the mixture as well.

Table 5.2.5-2. Impurities in recycled uranium at Hanford.

Constituent	Maximum allowed <sup>a</sup>	Observed range <sup>b</sup>	Reference level <sup>c</sup>
Plutonium	10 ppb U	<1 - 2 ppb U	0.4 nCi Pu-alpha/gU
Neptunium	Not established	0.04 - 0.16 ppm U	0.4 nCi <sup>237</sup> Np/g U
Thorium	750 ppm U	8 - 10 ppm U	5 pCi <sup>232</sup> Th/g U
<sup>99</sup> Tc	Not established	3 - 4 ppm U	0.2 uCi <sup>99</sup> Tc/g U
<sup>103,106</sup> Ru	<20 uCi/lb U	<6 uCi/lb U	40 nCi <sup>106</sup> Ru/g U
<sup>95</sup> ZrNb	<10 uCi/lb U	<4 uCi/lb U	20 nCi <sup>95</sup> ZrNb/g U
Other gamma emitters	<2 uCi/lb U	0.09 - 0.75 uCi/lb U	Negligible

a. From UO<sub>3</sub> Plant operating specifications, OSD-U-185-0001 (Thompson 1986).

b. From analysis of uranium lots 88-1, 88-2, 88-3 that were processed in 1988, and lots 93-01, 93-02, 93-03, 93-04, and 93-05, processed in 1993.

A reference level is chosen for determining bioassay monitoring needs and for use as an initial assumption in evaluating intakes. The use of the reference levels is expected to result in a slight overestimate of dose compared to levels actually observed in 1988.

The rigorous radiation protection barriers and procedures designed to prevent intakes of plutonium and fission products were not, in general, applied to work with uranium. Hence, exposure to uranium in the major uranium facilities was considered chronic exposure until 1992.

Uranium compounds at Hanford ranged from very soluble uranyl nitrate and soluble UO<sub>3</sub> to relatively insoluble UO<sub>2</sub> and U<sub>3</sub>O<sub>8</sub>. Dissolution tests in simulated lung fluid were conducted on samples from the major uranium handling facilities. Results are shown in Table 5.2.5-3. Because the relationship between the old lung fluid studies and the ICRP 66 absorption types is not established, Table 5.2.5-3 also shows claimant-favorable recommended absorption types for intakes from the listed facilities, which should be used unless person-specific data are available. These absorption type assumptions should be applied to the impurities as well. <sup>239</sup>Pu can be assumed for the plutonium alpha impurity.

Table 5.2.4-3. Inhalation class for Hanford uranium compounds.

ICRP 30 inhalation class from lung fluid studies	Compound and location	Recommended ICRP 66 lung absorption type
80% D 20% W	Hanford UO <sub>3</sub> Plant smear sample dissolution study in 1984 <sup>a</sup> , (UO <sub>3</sub> powder)	b
10% D 90% Y	Hanford 303-M Building air sample dissolution study <sup>c</sup> (300 Area Uranium Fuel Production Facilities)	b
29% D 71% Y	Hanford 333 Building air sample dissolution study <sup>c</sup> (300 Area Uranium Fuel Production Facilities)	b
20% D 80% Y	Hanford 306-W Building Machine Shop air sample dissolution study <sup>c</sup>	b
	Uranyl nitrate at PUREX or UO <sub>3</sub> Plant	F
	UCl <sub>4</sub> or U carbonate (assumed form after discharge to the soil)	M <sup>d</sup>

a. Sula, Bihl, and Carbaugh (1989).

b. Because the conversions from the solubility studies to the ICRP absorption types are not exact, the dose reconstructor may use the same percentages for D to F, W to M, etc. or may just use the predominant form to maximize dose to the organ of concern; for instance, the 303-M Building uranium might be considered 10% F, 90% S or all type S.

c. Letter Report to Monte J. Sula from Darrell R. Fisher, January 20, 1986.

d. Cooke and Holt 1974.

A note about sampling of UO<sub>3</sub> Plant workers: Because chemical toxicity was the principal concern for uranium exposures at UO<sub>3</sub> Plant, one sampling scheme used was to obtain both a Friday evening sample and Monday morning sample. The period of this sampling scheme was not established, other than in the 1970s and maybe earlier. This scheme was changed to a Monday-morning-only sampling circa early 1980s. Change over should be clear in the records.

The Friday/Monday sampling scheme was also used in 1962-63 for 313 and 314 Building workers.

- c. The Wilson history states that the uranium urinalysis program prior to 1948 was not reliable. The fluorometric method, which fused uranium from raw urine with sodium fluoride and measured the fluorescence when the compound was exposed to ultraviolet light, was implemented sometime during the first half of 1948 (Healy 1948, Wilson 1987). This method was used for elemental uranium analyses, with various refinements over the years including some upfront chemistry on the raw urine, until about 1991, when it was replaced by kinetic phosphorescence analysis (KPA) (Lardy 2003). [Note: Mr. Lardy said about 1990 but other evidence indicates late 1991.] A 1970 letter describes two procedures: one with wet-ashing with nitric acid and hydrogen peroxide, then acidification and counting of a 100  $\mu\text{L}$  aliquot with a detection level of 0.5  $\mu\text{g/L}$ ; another with extraction (after wet-ashing) with methyl isobutyl ketone and ammonium hydroxide. The detection limit for the latter was listed as 0.05  $\mu\text{g/L}$  but the recoveries were about the same for both methods so the latter must have used a 10 times larger aliquot. Based on requirements in later statements of work, it is assumed that the first method was used for routine analyses. A third method was also listed; this was a radiometric procedure using the same separation chemistry as the second procedure, but the sample "is measured by a gas flow proportional counter or a ZnS(Ag) scintillation counter." (Lardy 1970) The detection limit was given as 0.5 dpm/sample. A 1989 description of the chemistry was wet-ashing with HCl and extraction with hexone. A 100 ml aliquot was used, but the results were reported as per total sample. The chemistry for the KPA involves a 50-ml aliquot that is wet-ashed with acid, passed through an ion exchange column, then eluted with weak acid. Results are reported as per total sample.

When alpha spectrometry was introduced in 1983, two uranium urinalyses procedures were offered: the elemental procedure discussed above and the alpha spectrometric procedure to provide isotopic results. Generally, the elemental procedure was used for workers exposed to natural or slightly enriched forms of uranium, and the isotopic procedure was used for depleted or more than slightly enriched forms of uranium. Generally, personnel working in the production facilities were monitored by the elemental analysis, whereas Pacific Northwest Laboratory workers were monitored by the isotopic analysis because of the wide scope of research projects that occurred over the years.

Alpha spectrometry cannot differentiate between  $^{233}\text{U}$  and  $^{234}\text{U}$ . Prior to 1994, the results for this region of the alpha spectrum were reported as  $^{233}\text{U}$ ; they were reported as  $^{234}\text{U}$  from 1994 to present unless it was specifically determined that the worker was exposed to  $^{233}\text{U}$ . Work with  $^{233}\text{U}$  did occur at Hanford, but was rare after the early 1970s, long before alpha spectrometry came into use for bioassay. So unless specifically mentioned in an intake investigation report, assume  $^{233}\text{U}$  results since 1983 are actually  $^{234}\text{U}$ .

$^{233}\text{U}$  was handled at 231Z Building in the mid 1960s as a special project, maybe extending into the early 1970s. This project involved thorium campaigns at PUREX, separation of the  $^{233}\text{U}$ , and shipment to 231Z Building. No details about this work have been uncovered as yet, such as isotopic purity. Because of the time frame, bioassay must have been for elemental uranium, at least until about 1970. If so, because of the high specific activity of  $^{233}\text{U}$ , the bioassay MDA would have been only about 90,000 dpm/L. Hopefully, something better was done, and the worker's record might show that, but as yet the specific bioassay used for the  $^{233}\text{U}$  project has not been discovered.

Table 5.2.5-4 summarizes the routine urinalysis detection levels and Table 5.2.5-5 summarizes nonroutine detection levels.

Table 5.2.5-4. Routine uranium urinalysis detection levels.

Period	Elemental		Isotopic	
	MDA, µg/L	Decision level, µg/L	MDA, dpm/sample	Decision level, dpm/sample
Prior through 1948	Not specifically mentioned		NA	NA
1948 through 1949	10	Anything detected	NA	NA
1950 through 1969(a)	4	(b)	NA	NA
1970 through 1974	0.5		0.5	
1974 through 1981	0.4			
1982 through 9/1983	0.05 – 0.25 (0.1 most probable)(c)			
10/1983 through 12/1983	0.03	0.5 (d)	0.035	
1/1984 through 8/1985	0.03		0.02	
9/1985 through 6/1990	0.03/0.5(e)	0.2(f)	0.02	
6/1990 through 10/1991	0.2/0.5(e)	0.2(f)	0.03	0.15/0.015(g)
11/1991 through present	0.06/0.5(e)	0.2(f)	0.02	0.15/0.10(g)

- Estimated time period based on 1954 and 1970 letters.
- Values were reported well below the 4 µg/L value so either the MDA was thought to be lower than that value or a decision level of 2 µg/L was being applied.
- MDAs were based on sample size, but 0.1 µg/L applied to most sample sizes.
- Values below this were recorded but not followed up as occupational intakes.
- The larger value is the MDA for a special (rapid) analysis for UO<sub>3</sub> Plant workers based on potential chemical toxicity. The need for this special analysis ceased in 1994 after the last processing in the UO<sub>3</sub> Plant.
- Based on upper level for natural background excretion. See text for discussion.
- First value applied to <sup>234</sup>U and <sup>238</sup>U; second value applied to <sup>235</sup>U based on natural background in urine. In 2002 the <sup>235</sup>U decision level was lowered to 0.007 dpm.

Starting about 1995, mass spectrometry has been used as an investigational tool to discriminate between natural background uranium and recycled uranium through measurement of <sup>236</sup>U. The presence of <sup>236</sup>U confirms an occupational intake of recycled uranium; the detection limit for <sup>236</sup>U is such that urinary excretion of uranium greater than 0.2 µg/L (see discussion of natural background excretion below) from an intake of recycled uranium should have a detectable amount of <sup>236</sup>U.

Natural uranium from nonoccupational intakes (primarily food and water) is excreted in urine at levels above the analytical MDAs for either the elemental uranium analysis or the alpha spectrometry analysis. The <sup>234</sup>U to <sup>238</sup>U ratio can be used to distinguish depleted uranium from natural uranium, but, considering uncertainties in analytical results, that ratio can not be used to distinguish recycled uranium. Three studies were conducted, in 1985, 1990, and 1995, to establish the range of natural background excretion in unexposed persons living near the Hanford site. The third study purposely looked for possible geographic and seasonal differences in the background. All studies found natural excretion to be lognormally distributed. Although the 50 percentiles and slopes of the excretion curves were different in the studies, each study found 0.2 µg/d to be about 99 to 99.9 percentile, although the 1995 study had one result that greatly exceeded the 0.2 µg/d value. (Carbaugh 2003) Hence, 0.2 µg/d was established in 1985 and continues to be used at present as the environmental decision level for exposures to natural or recycled uranium. Only urinary excretions values greater than 0.2 µg/d, which converts to 0.15 dpm/d for <sup>234</sup>U and <sup>238</sup>U and 0.007 dpm/d for <sup>235</sup>U, are considered indicative of a potential occupational source. Nevertheless, the one result in the 1995 study and many worker-specific investigations of urinary results exceeding 0.2 µg/d have shown that results well above the environmental screen level do occur from natural sources. Some of these were shown to be due to a specific home water well; others occurred from workers on city water from wells (but apparently not all wells).

It is reasonable to use the urinary excretion values of 0.2 µg/d for elemental analyses, 0.15 dpm/d for <sup>234</sup>U and <sup>238</sup>U and essentially anything detected for <sup>235</sup>U, to distinguish between natural background and potential occupational exposure for natural and recycled uranium, unless the worker's file shows <sup>236</sup>U results or other studies that show the excretion was from natural sources. These environmental decision levels should apply to the entire history of Hanford. Prior to 1985, there will undoubtedly be excretion values exceeding the environmental screening levels that were nevertheless due to natural sources, but it's unlikely there will be data available to prove it.

Background excretion of uranium in feces probably varies over an even larger range than urinary excretion; however, a definitive study for the Hanford area has not been conducted. Fecal samples were rarely obtained for potential uranium intakes; when they were, the investigation report should discuss how the results were interpreted.

### 5.2.6 Fission Product Analysis

Fission product urinalysis was the method used to monitor for intakes of fission products until whole body counting was implemented in 1960. Routine fission product urinalyses started in January 1947, but ferrous hydroxide precipitation was used on the supernatant from the plutonium lanthanum fluoride procedure, and the results were erratic with occasional breakthrough of <sup>40</sup>K. So data prior to 1948 should be considered unreliable and should be ignored (see guidance in 5.1 instead). The procedure initiated in 1948 was to add Sr carrier to the aluminum oxide solution for the plutonium procedure, then precipitate La hydroxide. This procedure was shown to extract the rare earths and strontium with yields ranging from 90% for Ce to 23% for Sr. The dried planchet was counted for beta activity with an approximate detection level of 30 dpm. (Healy 1948, Wilson 1987) The same procedure was in use in 1954 with the addition of a Ce carrier. It was also listed in the compilation of procedures referred to as the "Old Bioassay Bible" in 1961, but that same compilation had a separate procedure for <sup>90</sup>Sr in urine. A memo in the Old Bioassay Bible discusses the start of use of a gas-flow, beta proportional counter in November 1958 which resulted in increased counting efficiency. The new detection limit was stated as 1.4 x 10<sup>-5</sup> µCi/sample, based on the counting efficiency of <sup>90</sup>Sr. "Gross fission products" are also mentioned in the 1970 letter from Matt Lardy at US Testing with a brief description that seems to imply the same procedure was still available, although probably not used much. The detection level was given as 5 dpm/sample based on the beta counting efficiency for <sup>90</sup>Sr. Table 5.2.6-1 summarizes the detection levels for the fission product urinalysis as best has been uncovered.

Table 5.2.6-1. Routine fission product urinalysis detection levels.

Period	MDA
1948 to 2/1956	30 dpm/sample
3/1956 to 10/1958	70 dpm/sample <sup>a</sup>
11/1958 to 1960s	31 dpm/sample
1970 <sup>b</sup>	5 dpm/sample

- a. Recorded as 3.1 or 3.17 E-5 µCi/ sample; not clear if this was intended to be the MDA or just a reporting level.
- b. Listed in the bioassay contract but probably not used; replaced by whole body counting and <sup>90</sup>Sr urinalyses.

It's a challenge to interpret the fission product urinalysis in a way that is meaningful as representative of all the possible fission products and activation products that a worker might theoretically have been exposed to. The procedure separated and counted radionuclides of alkaline earths and rare earths,

such as strontium, yttrium, barium, lanthanum, cerium, europium, and promethium. It did not account for radionuclides of ruthenium, cesium, zinc, cobalt, manganese, niobium, or zirconium. The abundances of all the fission products, relative to each other, varied (considerably) as a function of the time from when the reactor fuel was removed from the core and allowed to cool to when the contamination was inhaled or ingested. See section 5.4 for a discussion about interpreting fission product mixtures.

After whole body counting came into routine use, regular use of the fission product urinalysis continued for many workers at facilities such as B Plant and Semi-Works where intakes of pure  $^{90}\text{Sr}$  were possible. So it was apparently being used as a  $^{90}\text{Sr}$  bioassay. The records show fission product analysis being used this way until early 1964. The same workers show actual  $^{90}\text{Sr}$  analysis results starting in 1965, probably starting with the new contract with US Testing.

### 5.2.7 Strontium

Records of  $^{90}\text{Sr}$  urinalyses, both routines and specials, begin showing up in the database in 1965. However, the compilation of procedures called the Old Bioassay Bible, 1961, had a procedure specific for strontium in urine and fecal salts that included counting total strontium and then allowing for  $^{90}\text{Y}$  ingrowth, yttrium separation, and counting of  $^{90}\text{Y}$  to account for  $^{90}\text{Sr}$  separate from gross strontium beta, if desired. This procedure was also mentioned in a memo, dated July 1963, documenting discussions between the Analytical Laboratories and Internal Dosimetry clarifying logistics of handling these samples and reporting  $^{90}\text{Sr}$  results. There are handwritten notes on this memo indicating that the detection level is about 20 dpm. Nevertheless, the database records show fission product urinalyses being used into 1964 and  $^{90}\text{Sr}$  urinalyses apparently starting in 1965. The value of  $1.67 \times 10^{-5} \mu\text{Ci/L}$  (37 dpm/L) is frequently entered in the database during 1965 and 1966 and seems to be the reporting level. This is consistent with a draft of the first contract with UST (the official one has not been found), dated August 1964, that listed a detection limit for  $^{90}\text{Sr}$  as 25 pCi/1.5L which converts to 56 dpm/1.5L or 37 dpm/L. The 1970 Lardy to Corley letter states that the detection limit is 1 pCi/L (2.2 dpm/L) (at 90% confidence), and describes the procedure as precipitation as the oxalate, then nitrate, removal of yttrium and barium, then reprecipitation as the carbonate and gross beta counting on gas flow proportional counters. A 1974 letter discussing terms of the statement of work with US Testing shows an "analytical limit" (defined as  $\pm 25\%$ ) at 50 dpm/sample and a reporting level of 2 dpm/sample. These values show again in the 1978 statement of work except the analytical limit is defined as  $\pm 100\%$ . A 1979 letter from Bob Robinson (PNL Internal Dosimetry) to R.B. Swoboda (US Testing bioassay supervisor) requests changes for  $^{90}\text{Sr}$  urinalyses so that the analytical limit ( $\pm 100\%$ ) be lowered from 50 dpm/sample to 5 dpm/sample, the reporting level be increased from 2 to 5 dpm/sample, and that an emergency analysis capability be added with an analytical limit of 10 dpm/sample and reporting level of 5 dpm/sample. In 1982 the detection limit was listed as 2.5 dpm/sample for  $^{90}\text{Sr}$  and 5 dpm/sample for  $^{89}\text{Sr}$ . But in the new contract starting October 1983 the detection limit was listed as 2.0 dpm/sample, and it stayed at that value until 1992 when it was raised to 10 dpm/sample. However, the procedure stayed the same throughout this period and the true MDA probably held at about 2 dpm/sample.

The results of the  $^{90}\text{Sr}$  procedure usually were reported as  $^{90}\text{Sr}$  although sometimes a value for  $^{89}\text{Sr}$  was also reported. Sometime in the 1980s a shortcut was added to the procedure that allowed skipping the  $^{90}\text{Y}$  ingrowth portion of the procedure if the first beta count was less than 1 dpm. When this happens the result is reported as Sr total or SRTOT, but the result may be interpreted as  $^{90}\text{Sr}$ . These results were below the required detection level anyway. Table 5.2.7-1 summarizes the routine urinalysis detection levels for  $^{90}\text{Sr}$  procedure.

Table 5.2.7-1. Routine <sup>90</sup>Sr urinalysis detection levels.

Period	MDA or MDC
Prior to 1965	May have been available but MDA not known
1965 to 1969	37 dpm/L
1970 to 1974	2.2 dpm/L
1975 to 3/1979	50 dpm/sample <sup>a</sup>
4/1979 to 1981	5 dpm/sample
1982 to 9/1983	2.5 dpm/sample
10/1983 to 6/1990	2 dpm/sample
9/1990 to 11/1991	30 dpm/sample
11/1991 to present	10 dpm/sample <sup>b</sup>

- a. Based on an unusual definition of “analytical limit” and probably conservative on the high side. Results <2 dpm were reported as 2 dpm; results > 2 dpm were reported as measured.
- b. Decision level was 5 dpm/sample. Prior to that time the MDA was also used as the decision level.

All strontium results at Hanford should be considered absorption type F. It is claimant-favorable to assume that <sup>90</sup>Sr and total radiostrontium results are <sup>90</sup>Sr even though <sup>89</sup>Sr may be present. Because the <sup>90</sup>Sr urinalyses method coincided in time with whole body counts, which would signal intakes of other fission products, <sup>90</sup>Sr urinalysis results should represent only strontium intakes (i.e., not be used as an indicator for other fission products unless they were detected in whole body counts). The exception would be <sup>147</sup>Pm, which probably tracked with the strontium through the various processes. See 5.4 for discussion of mixtures.

### 5.2.8 Promethium

Hanford was involved in the manufacture of heat sources using <sup>147</sup>Pm. The time period seems to start in 1966 and continue into the early 1970s (Howell and King 1968). The high activity work (kilocuries) took place in the 325 Building, but some exposure apparently occurred as early as 1962 or 1963 in the 222-S Chemistry Laboratory and as late as 1971 in the 308 Fuels Laboratory. Also animal studies were conducted with <sup>147</sup>Pm as part of research to develop a human biokinetic model for the behavior of promethium in the body. A small human volunteers study using <sup>143</sup>Pm was conducted in 1967 or 1968 (Palmer et al 1969).

The work on the heat sources involved converting promethium/cerium nitrates into Pm<sub>2</sub>O<sub>3</sub> by separation chemistry then calcining. There was also one mention of cold-pressed, sintered Pm<sub>2</sub>O<sub>3</sub> for heart implants. According to ICRP 68, the nitrate form should be considered absorption type M and the oxide form absorption type S.

In the 1960s, <sup>147</sup>Pm sample results were reported as, for urine - μCi/L, for feces – μCi/kg, which is different than most radionuclides, which were reported as per sample. From 1974 forward, the results appear to be reported as per sample. Table 5.2.8-1 lists the <sup>147</sup>Pm minimum detection levels at various times.

Fecal samples were analyzed for <sup>147</sup>Pm for some of the potential intake events in the late 1960s. The MDA or at least the lowest reporting level appears to be 1.67x 10<sup>-5</sup> μCi/kg. An MDA for fecal samples does not appear in laboratory statements of work during the 1970s; but reappears in the 1980s: 28-110 dpm/sample in 1982 depending on sample size (roughly 400 dpm/kg); 220 dpm/sample in 1983 – 1990s.

Table 5.2.8-1. Routine <sup>147</sup>Pm urinalysis detection levels.

Period	MDA or MCA
Prior to 1965	May have been available but MDA not known
1965 to 1969	37 dpm/L (1.67E-5 μCi/L)
1970 to 1973	22 dpm/L (1.0E-5 μCi/L)
1974 to 1979	50 dpm/sample <sup>a</sup>
1980 to 1981	20 dpm/sample
1982 to 9/1983	5 dpm/sample
10/1983 to 6/1990	4.0 dpm/sample
11/1991 to present	30 dpm/sample

a. Based on an unusual definition of “analytical limit” and probably conservative on the high side. Results <25 dpm were reported as 25 dpm; results > 25 dpm were reported as measured.

Only one description of the procedure was found, and that same procedure showed up in documents dated 1970, 1974 and 1977. Promethium and rare earths were precipitated as the fluoride. Interferences such as zirconium, scandium and IV actinides were removed by extraction by TTA in xylene, first at pH <1, then at pH about 4. The final sample was counted by liquid scintillation. Remaining rare earths were distinguished from <sup>147</sup>Pm by proper setting of the counting window on liquid scintillation spectrometer.

### 5.2.9 Polonium

Considerable activity toward initiating a bioassay procedure and establishing a biokinetic model for <sup>210</sup>Po was found in the files circa 1968 through the mid 1970s. There is an indication of work with pure <sup>210</sup>Po in the 308 Building in 1968 and again in 1975. Whether the work in the 308 Building was continuous through that period or just in those two years was not determined. Inference can be made that there was work somewhat prior to 1968 based on a handwritten note documenting a telephone conversation in November 1967 in which it was stated that the <sup>210</sup>Po starts in the process in the soluble form but is converted to the insoluble form. However, U.S. Testing was asked to develop a bioassay procedure in March 1968 and did so shortly thereafter, so apparently concern for possible intakes became important in early 1968. There also was work with <sup>210</sup>Po in the 325 Building that started in June 1972 and was slated “to run for 2-3 years.”

The procedure developed for <sup>210</sup>Po by U.S. Testing in March 1968 was as follows. For urine, gold, mercury, platinum, and tellurium were removed by reduction in hydrazine in an HCl solution. Iron was removed by reduction with ascorbic acid. The polonium was then removed from solution by deposition on silver film by heating at 95 degrees C for 2 hours. The silver film was counted by alpha proportional counting. Fecal samples were first wet-ashed in concentrated nitric acid and peroxide then treated the same as urine samples. Sometime between 1968 and 1974, the silver foil was replaced by copper foil and alpha spectrometry counting had replaced proportional counting. Detection limits for routine urinalysis are shown in Table 5.2.9-1 and for nonroutine excreta bioassay in Table 5.2.9-2.

Because <sup>210</sup>Po is a natural radionuclide from the <sup>238</sup>U decay chain, <sup>210</sup>Po exists naturally in urine and feces. Nothing was found in the records indicating that a study on natural excretion levels for persons living around Hanford had been conducted. ICRP 23 (1975) indicates that excretion levels differ between smokers and nonsmokers, and provides the following estimated excretion values: urine, smokers: 0.065 pCi/d, nonsmokers: 0.011 pCi/d; feces, smokers: 3.3 pCi/d, nonsmokers: 3.2 pCi/d.

Table 5.2.9-1. Routine <sup>210</sup>Po urinalysis detection levels.

Period	MDA
3/1968 to 1973	5.4E-7 $\mu$ Ci/L
1974 to 1979	1 dpm/sample <sup>a</sup>
1980 to 9/1983	0.1 dpm/sample <sup>a</sup>
10/1983 to present	No longer listed in the contract except for expedited or emergency samples. Not likely used.

a. Based on an unusual definition of “analytical limit” and probably conservative on the high side. Reporting level listed as 0.5 dpm/sample.

Table 5.2.9-2. MDAs for nonroutine <sup>210</sup>Po excreta analyses.

Period	Fecal samples, MDA, dpm/sample			Urine samples, MDA, dpm/sample	
	Emergency <sup>a</sup>	Expedite	Priority	Emergency <sup>a</sup>	Expedite
3/1968 to 1973	NA	NA	5.4E-7 $\mu$ Ci/kg	NA	NA
1974 to 9/1983	NA	NA	(b)	NA	NA
10/1983 to 9/1985	340	NA	NA	0.8	NA
10/1985 to 6/1989	340	100	NA	0.8	0.1

a. At times the emergency category was called “rush” and the routine category was called “normal.”

b. Probably available but not listed in the contract.

These values were based on only 7 subjects, however, and even so, the fecal excretion ranged from 1.7 to 6.4 pCi/d.

If there are person-specific baseline values for urine or fecal excretion of <sup>210</sup>Po, those should be used to subtract from later results. If not, then the ICRP 23 values above should be used; if smoking status is not known, use the values for nonsmokers.

### 5.2.10 Neptunium

At PUREX from 1958 through 1972 <sup>237</sup>Np was removed from the dissolved fuel, purified, and packaged for shipment offsite. It was downloaded from an ion exchange column and packaged in liquid form, but the chemical form has not been discovered yet. Although mostly <sup>237</sup>Np by mass, the small mass of <sup>238</sup>Pu in the product produced most of the radioactivity. Plutonium bioassay was considered sufficient to monitor for intakes.

### 5.2.11 Other Limited-Exposure Radionuclides

Hanford has always been a center for research, first as part of Hanford Works, then (1965 to present) as part of Pacific Northwest Laboratory. As such, small scale (in terms of either the number of persons or activity of the source) use of various radionuclides not addressed above has occurred throughout the history of Hanford. The following discussion, addressing <sup>14</sup>C, <sup>232</sup>Th, radon, <sup>90</sup>Y, <sup>227</sup>Th, <sup>227</sup>Ac, and <sup>32</sup>P, is not likely comprehensive.

**Carbon-14** exposure occurred at the 3731 Building in the mid 1950s when irradiated graphite samples were brought from the operating reactors to the 3731 Building for destructive testing. No information has been uncovered yet as to what bioassay if any was done. <sup>14</sup>C was also used as a tracer in biological experiments. One documented study was conducted in the late 1990s in the Life Science Laboratory-II Building, involving a total of about 4 Ci of <sup>14</sup>C. Urinalyses were obtained on about 20 researchers. The MDA was 10 dpm/ml. Baseline samples were obtained from each worker because natural excretion levels had not been established. ICRP 68 and 71 assign <sup>14</sup>C in organic compounds to class SR2, which has not been modeled in IMBA yet. If a claimant appears to have

been exposed to organic  $^{14}\text{C}$ , special consultation with the IMBA programmers may be necessary to determine an acceptable method to determine the dose.

PUREX ran thorium campaigns in the 1960s and early 1970s. In terms of grams or curies, the thorium campaigns were small compared to the normal separation of plutonium.  $^{232}\text{Th}$  was irradiated to produce  $^{233}\text{Th}$ , which decays to  $^{233}\text{U}$ . Although called a thorium campaign, it was the  $^{233}\text{U}$  that was separated at PUREX and transported to 231-Z for experiments. Hence, the most likely source of intake was the  $^{233}\text{U}$  during the loading out and transportation. Thorium exposure was more likely at the 3732 Building where the powdered thorium fuel targets were fabricated, which apparently contaminated the building with thorium "fines." Some work was also done with  $^{232}\text{Th}$  slurries in the 3720 Building in the mid 1990s. The plan was to collect baseline urine samples on the few workers involved, then collect special bioassay samples if air samples exceeded a cumulative exposure of 40 DAC-hrs. The urinalysis MDA was stated to be 0.1 dpm/sample.

There was a **radon** generator used for animal studies in the 108F Building and was later moved to LSLII. Monitoring was probably just by air sampling; but no information has been discovered yet. There should have been only a few researchers potentially exposed.

Some unusual radionuclides were isolated in the 325 Building for nuclear medicine studies in the mid to late 1990s. One of these projects isolated  $^{90}\text{Y}$  from  $^{90}\text{Sr}$  and packaged and shipped the 90Y to various users around the world. Only a few workers were involved. The work was monitored by air samplers and no loss of control of the material occurred so no bioassay was obtained. The material was in an insoluble form so that chest counting would have been the only possible bioassay because of the 64-hr physical half-life; however, the need to perform chest counting never arose.

Another project involved "milking"  $^{227}\text{Th}$  from  $^{227}\text{Ac}$  on an ion exchange column. A bioassay procedure was developed specifically for this project under the assumption that the project was going to continue for several years; however, the project ceased after only a few milkings. Only a couple of researchers were involved. The bioassay procedure had a stated MDA of 0.1 dpm/sample for  $^{227}\text{Th}$ .

**Phosphorous-32** was used for biological tracer studies, and according to one retired researcher, "pipetting was done by mouth in the old days." Such exposure would be limited to a few researchers and would have to be established through the claimant interview or by some indication of  $^{32}\text{P}$  bioassay samples in the worker's record. More information might be uncovered if such a case is encountered.

### 5.3 IN VIVO MINIMUM DETECTABLE ACTIVITIES, ANALYTICAL METHODS, AND REPORTING PROTOCOLS

In vivo counting equipment and techniques were developed in the late 1950s and have been in routine use for measuring x-ray and gamma-ray-emitting radionuclides since 1960. (Unless otherwise noted, the in vivo information below came from Wilson 1987 and Lynch 2001).

#### 5.3.1 Whole Body Counters

The first whole body counter started counting workers in mid 1959 and became a routine method in 1960. It consisted of a single NaI crystal (9.375-in. diameter and 4-in. thick) housed in a counting room with 10-in. thick pre World War II steel plate on all six sides, and graded shielding on the inner surfaces (lead, cadmium, copper) (Wilson 1987, Roesch et al 1960). The counting geometry was a chair configured to simulate a one-meter arc. The original count time was 20 minutes which was reduced to 10 minutes in October 1962. A second, same-sized NaI detector was added in 1963 (Brady 1964). According to personal recollection of H.E. Palmer, the two-detector system improved

the detection capabilities somewhat. However, the MDAs quoted in a report in the fall of 1964 were the same MDAs listed in Mr. Palmer's Laboratory Record Notebook in 1960, so apparently the difference between the systems was not great enough to warrant republishing the MDAs. So the MDAs shown in Table 5.3.1-1 are the only MDAs found for the 1960s and 70s, and apparently were meant to apply generally to the various whole body counters in operation during this period.

Shortly after the chair counter in the "Iron Room" became operational, an entirely new design called the shadow shield counter was developed. The shadow shield consisted of a bed shielded on the bottom and sides by lead. The bed moved under a large NaI crystal (11.5-in. diameter by 4-in thick) that was also shielded by lead except for the downward-looking face that looked directly onto the body as it passed under the crystal. The shadow shield detector was mounted in a mobile trailer and moved to areas located nearer the worksites on the Hanford site. The mobile trailer also had a thyroid detector and a wound counter. The mobile, shadow shield detector became operational in 1963 (Brady 1964). The mobile counter was described as having comparable sensitivity to the "larger, conventional whole body counters installed in massive iron rooms. There is, however, some decreased sensitivity in the lower energy region below about 300 keV, due to increased contribution to the background from scattered radiation." (Swanberg 1963).

A report listing the radionuclides detected in workers at the whole body counter facility in 1961 listed  $^{24}\text{Na}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{95}\text{Zn}$ ,  $^{95}\text{Nb}$ ,  $^{99}\text{Mo}$ ,  $^{99}\text{Tc}$  [presumably  $^{99}\text{Mo}$ ,  $^{103}\text{Ru}$ ,  $^{106}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$ , and  $^{144}\text{Ce}$  (Henle 1962). A similar report summarizing 1961-63 results added  $^{46}\text{Sc}$ ,  $^{51}\text{Cr}$ , and  $^{59}\text{Fe}$  to the list.

A shadow shield whole body detector was added at the whole body counting facility in 1977. This assembly had two 35% GeLi detectors and a 4-in. by 4-in. by 16-in NaI detector. It ceased operation in 1987 when the two new counting rooms were added. A listing of MDAs was found that applied to 1980. These are used to represent this shadow shield detector.

By 1978 there were four shadow shield whole body counters available for use: one at the Whole Body Counting Facility, two in mobile trailers, and one at the Emergency Decontamination Facility, the latter designated for use for large, acute intakes with potentially high levels of external contamination.

A "standup" counter was put in operation in 1985 and is still in operation today. It consists of five vertically-stacked NaI crystals in a small lead-shielded area. The worker stands in front of the detectors with the detectors to his/her back; the detector array is raised or lowered to best fit the height of the person being counted. There are four 9.375-in-diameter-by-4-in.-thick detectors and one 11-in.-diameter-by-4-in.-thick detector, the latter being located behind the thoracic region. Count time is 200 seconds.

In July 1989, a coaxial HPGe scanning array was developed and is still in operation today. For this system the person lies on a bed in a shielded room and the detector array moves under the bed. The configuration of this system, in terms of number and size of the detectors, has changed many times. It started as four 68% HPGe detectors; one of the detectors was replaced with a 120% detector in late 1995; in May 1997 the system was upgraded to include seven detectors including three 120% detectors. When a 4-detector array, the system was used only when a count on the "standup" counter had detectable activity of an occupationally-related radionuclide. However, it was considered the count of record. In 1997, because of its greater resolution and lower decision levels, it started being used for routine counts for workers exposed to mixtures of  $^{137}\text{Cs}$  and plutonium. The count time was usually 10 minutes; however, 20-minute count times are used as confirmation of an initial count with detectable activity. Consequently, the database will usually show a 10-minute count and a 20-minute count on the same day or a few days later if the first count had detectable activity (excluding  $^{40}\text{K}$  or medical radionuclides).

Table 5.3.1-1. Routine whole body counting detection levels.<sup>a</sup>

Period	Nuclide	MDA (nCi)	Reporting level (nCi)
1960 -1976 <sup>b</sup>	Na-22	1.0	10
	Na-24	0.3	0.3
	Cr-51	50	50
	Fe-59	2.0	10
	Co-60	0.4	10
	I-131	0.5 <sup>c</sup>	10
	Cs-137	0.5	0.5
1977-1984 <sup>d</sup>	Na-22	1.0	10
	Na-24	0.5	0.5
	Cr-51	15	15
	Mn-54	2.0	10
	Fe-59	4.0	10
	Co-60	2.0	10
	Zn-65	3.0	3
	Zr/Nb-95	2.0	10
	Ag-110m	2.0	10
	Ru-106	12	12
	Sb-125	3.0	10
	I-131	4.0 <sup>c</sup>	10
	Cs-137	2	2
	Ce-144	100	100
1985-86	Na-22	1.5	1.5
	Mn-54	3	3
	Fe-59	6	6
	Co-60	3	5
	Zr-95	3	3
	Ru-106	12	12
	Eu-154	4.5	4.5
1987	Co-60	3	3
	Cs-137	3	3
	No changes for other radionuclides. Anything detected is reported.		
1992	New formalism for decision level calculation; "limit" in electronic database changed from MDA to decision level.		
1993	Actual values, regardless of amount, reported for Co-60 and Cs-137, including negative numbers.		
1995-10/1999 <sup>e</sup>	Co-60	4	Every result
	Cs-137	4	Every result
	I-131	5	Every result
	Mn-54	3	Every result
	Na-22	2	Every result
	Na-24	1	Every result
	Pr-144(Ce-144)	230	Every result
	Other radionuclides		Anything detected
10/1999 to present <sup>e,f</sup>	Co-60	1.25	Every result
	Cs-137	1.3	Every result
	Eu-154	3.75	Every result
	Other radionuclides		Anything detected

- Nominal MDAs based on whatever phantom was available at the time period, the routine count time, and the least sensitive of various whole body counters in operation at the time. Listing of an MDA for a given radionuclide does not necessarily mean that that radionuclide was frequently encountered. If smaller MDAs are listed in the database for a given count, use them.
- Based on 95% confidence of detection.
- See also discussion on thyroid detectors.
- Based on 99% confidence of detection.
- Least sensitive of many options throughout the period. Much better sensitivities were available using the HPGe system.
- Physical configurations stayed essentially the same but ABACOS software introduced changes to methodology for determining MDAs and decision levels.

The first mobile counter stopped being used at various onsite locations in the early 1980s. A new trailer was obtained in 1989 and reconfigured with a new, standup counter consisting of five 4-in. by 16-in. by 4-in. thick NaI detectors plus one 4-in. by 8-in. by 4-in. thick detector. The trailer was parked in the 200 East Area and operated remotely starting in 1991. The sensitivity of the detector was comparable to the standup counter at the Whole Body Counter Facility. The use of this facility was infrequent and it was discontinued in August 1995.

From 1960 to 1983, four radionuclides were reported routinely:  $^{24}\text{Na}$ ,  $^{40}\text{K}$ ,  $^{65}\text{Zn}$ ,  $^{137}\text{Cs}$ . For dose reconstruction, only the  $^{137}\text{Cs}$  is of interest because  $^{40}\text{K}$  is strictly a natural source, and the  $^{65}\text{Zn}$  and  $^{24}\text{Na}$  came from drinking water, the  $^{65}\text{Zn}$  from water in the cities surrounding Hanford and the  $^{24}\text{Na}$  from drinking water at the reactors. Net counts in a fifth region of the spectrum was also commonly calculated but not usually associated with a radionuclide. This was the low energy portion of the spectrum noted as the GOK region. The technique was to calculate the activity of the higher energy radionuclides  $^{24}\text{Na}$ ,  $^{40}\text{K}$ , etc., then subtract the Compton scatter contribution from those radionuclides and see if there were any counts left over in the low energy region. If there were sufficient counts left over, then they would have investigated further to see if an occupational radionuclide was the source, recognizing that the low energy region was also subject to increased electronic noise and general background scatter in the crystal. (GOK stands for God Only Knows.)

If the hardcopy form (In-Vivo Counter Results) shows the "traces of xxx invalidate routine calculation" statement, then some radionuclide other than the standard four was detected; often this was  $^{60}\text{Co}$ . The activity of that radionuclide may or may not be written on the form. Activities that exceeded 10 nCi or 1% of the MPBB were calculated and reported on a Whole Body Counter Evaluation form (Glenn 1968). See section 5.3.5 for instructions.

Most workers in the early days of whole body counting had detectable activities of  $^{137}\text{Cs}$ . Most of this was attributed to fallout. Some workers had even higher levels of  $^{137}\text{Cs}$  from consumption of wild game. A decision level used to establish the difference between occupational and nonoccupational sources of  $^{137}\text{Cs}$  intake has not been uncovered in the records, and may not have been developed so long as the  $^{137}\text{Cs}$  measurement didn't exceed 1% of a MPBB. The following guidance may be used however.

- The  $^{137}\text{Cs}$  intake should be considered occupational if the same whole body count detected other fission or activation products (excluding  $^{65}\text{Zn}$  or  $^{24}\text{Na}$ ). It should also be considered occupational if a fission product or radiostrontium urinalysis showed detectable activity and the sample was obtained within the period between the previous and next whole body count.
- If an investigation was done and the record clearly shows that the intake was due to a nonoccupational source, then the  $^{137}\text{Cs}$  may be disregarded.
- NCRP Report No. 94 (NCRP 1987) provides mean body burdens of  $^{137}\text{Cs}$  for the United States for the years most likely to produce interference with occupational whole body count results. Those values are listed in Table 5.3.1-2. If no other fission or activation products are linked to the intake (excluding  $^{65}\text{Zn}$  or  $^{24}\text{Na}$ ) and the  $^{137}\text{Cs}$  result is less than the values given in Table 5.3.1-2, the  $^{137}\text{Cs}$  result may be assumed to be due to fallout.

Table 5.3.1-2. Mean body burdens of  $^{137}\text{Cs}$  from fallout in the United States.<sup>a</sup>

Year	Body burden (nCi)	Year	Body burden (nCi)
1953	0.27	1966	9.7
1954	1.1	1967	5.6
1955	2.2	1968	3.5
1956	4.3	1969	2.7
1957	5.1	1970	2.7
1958	6.5	1971	2.7
1959	8.1	1972	2.7
1960	6.8	1973	2.7
1961	4.6	1974	1.6
1962	6.0	1975	1.1
1963	11	1976	1.6
1964	19	1977	1.1
1965	16		

From NCRP Report No. 94.

### 5.3.2 Chest Counters

In 1967 the original large NaI detector in the Iron Room started to also be used for chest counting, with emphasis on uranium workers. The detector was placed directly over and nearly in contact with the chest region with the worker in the supine position. Count time was 30 minutes. MDAs were determined to be 6.7 nCi for “U natural,” presumably based on  $^{234}\text{Th}$ , 0.15 nCi for  $^{235}\text{U}$ , and 0.33 nCi for  $^{241}\text{Am}$ . However, in the next year a new counting room was built, called the Lead Room, specifically for chest counting. It was outfitted with four 5-in.-diameter by 0.375-in-thick NaI detectors, located two in front and two in back of the subject. Count time was 30 minutes. A lung phantom with variable chest wall thickness was developed for calibration of the new system. MDAs were listed as 0.15 to 0.6 nCi for  $^{241}\text{Am}$ , 2.0 to 3.7 nCi for  $^{234}\text{Th}$  (assumed to be in equilibrium with  $^{238}\text{U}$ ), and 0.17 to 0.37 nCi for  $^{235}\text{U}$ , depending on a subject’s weight to height ratio. (Chest count MDAs are summarized in Table 5.3.2-1.) MDAs for direct measurement of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  using the 17 keV x-rays were calculated at times, but the values were extremely large relative to the Maximum Permissible Lung Burden so primary reliance was placed on measuring  $^{241}\text{Am}$  and applying a plutonium to americium ratio. The chest counter was also calibrated to measure bremsstrahlung radiation from  $^{90}\text{Sr}$  or  $^{147}\text{Pm}$ , although these counts were probably not routine counts. MDAs for those counts were listed as 25 – 40 nCi and 0.5 – 1.5  $\mu\text{Ci}$  for  $^{90}\text{Sr}$  and  $^{147}\text{Pm}$ , respectively. A second chest counting system became operational in 1978. A phoswich detector became available and was used occasionally for special chest counts but was never implemented on a routine basis.

A solid state germanium counting system using 3 planar HPGe detectors replaced the NaI detector in the Iron Room chair counter in 1983. The HPGe detectors provide better spectral resolution than the NaI detector, thus lower backgrounds in the region of interest and better discrimination against radon decay products and better detection of low-energy photon emitters in the presence of large activities of high-energy photon emitters (e.g.  $^{137}\text{Cs}$  or  $^{60}\text{Co}$ ). They also have a thin window on the end of the detector facing the chest for better transmission of low-energy photons. The detectors were positioned over the front of the chest (two over the right lung) with the subject in the supine position. Counting time was 2000 seconds. MDAs were quoted for “an average size person” as 0.1 nCi for  $^{241}\text{Am}$ , 0.5 nCi  $^{144}\text{Ce}$ , 0.7 nCi of  $^{234}\text{Th}$  ( $^{238}\text{U}$ ), 0.05 nCi  $^{235}\text{U}$  (Palmer and Rieksts 1984). These values were quoted as being the RDA or Reliably Detectable Activity, which was defined as 3 standard deviations of the background continuum plus was discernable by naked-eye inspection of the spectrum (Carbaugh et al 1988).

Table 5.3.2-1. Routine chest counting detection levels.

Period	Radionuclide	MDA (nCi)
1967	Am-241	0.33
	U-238 (Th-234)	6.7
	U-235	0.15
1968-1983	Am-241	0.15-0.6 <sup>a</sup>
	U-238	2.0-3.7 <sup>a</sup>
	U-235	0.17-0.37 <sup>a</sup>
	Sr-90	25-40 <sup>a</sup>
	Pm-147	0.5-0.15 <sup>a</sup>
1983-1986	Am-241	0.24 <sup>b</sup>
	U-238	1.1 <sup>b</sup>
	U-235	0.08 <sup>b</sup>
	Ce-144	0.78 <sup>b</sup>
	Eu-154	0.07
1987	Am-241	0.28 <sup>c</sup>
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
1988 -6/1989-	Am-241	0.18 <sup>c</sup>
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
7/1989 – 1991 <sup>d</sup>	Am-241	0.18 <sup>c</sup>
	U-238	1.2
	U-235	0.08
1992 - 5/1996 <sup>e</sup>	Am-241	0.18 <sup>c</sup>
	U-238	3
	U-235	0.2
6/1996 - 10/1999	Am-241	0.28 <sup>c</sup>
	U-238	1.6
	U-235	0.095
11/1999 – present	Am-241	0.25 <sup>c</sup>
	U-238	1.5
	U-235	0.090

- a. Range for different weight to height ratios, a chest-wall thickness adjustment for both front and back chestwalls. Use highest value for default to cover large persons.
- b. Assumed MDA = (RDA)(4.65/3). Am-241 adjusted for 95<sup>th</sup> percentile male chestwall (.2/.13)
- c. Adjusted for 95<sup>th</sup> percentile male chestwall.
- d. <sup>144</sup>Ce and <sup>154</sup>Eu no longer automatically reported for chest counts because now can be quantified in the Ge whole body counter.
- e. Applies to the 6-detector array. Better sensitivity was obtained by the 4-large-area-detector array in the Stainless Steel Room.

Special chest counts, as follow-up to high routine chest counts or upon special request, were twice the normal counting time so the MDAs were about 0.7 times lower.

The 3-detector system was soon (within about a year) upgraded to a 6-detector array, which allowed routine counting times to be reduced to 1000 seconds with nearly the same RDAs (Carbaugh et al 1988). A second HPGe-detector array became operational in July 1989 in a new shielded cell called the Stainless Steel Room because the inner (i.e., visible) lining of the graded shield was stainless steel. Although intended to be a 6-detector array, this counter had only 4 detectors at first because of operational problems with the detectors. Counting times were increased to 2000 seconds for the 6-detector array and 3000 seconds for the 4-detector array.

In September 1994 the chest counter in the Stainless Steel Room was converted to a 4-detector array using larger area detectors. The same change was implemented in the Iron Room in June 1996. This configuration continues to the present. The routine counting time was increased to 3000 seconds for the larger area arrays in November 1995; special counts and recounts were 3600 seconds.

Ultrasonic measurements of chestwall thickness for workers that had activity in the lung began in about 1979 and continues today. So decision levels for non-detected activities use a weight-to-height ratio to estimate chestwall thickness, whereas detected activity is corrected for chestwall thickness using ultrasound.

Individual-specific decision levels were reported to the database for each count, each radionuclide, starting in 1992.

For in vivo counting, the assumption was made that  $^{234}\text{Th}$  was in equilibrium with  $^{238}\text{U}$ . This was a reasonable assumption at Hanford. Certainly, uranium recently separated from dissolved fuel was not in equilibrium, and uranium being treated at the  $\text{UO}_3$  Plant may or may not have been in equilibrium depending on how long the material had taken to go through the separation process and be transported to the  $\text{UO}_3$  Plant. However, uranium in this part of the fuel cycle was very soluble and not important relative to chest counting. Chest counts were used to monitor for intakes of insoluble forms of uranium, which also were very old forms in terms of time since purification from decay progeny (e.g. machining on metal, uranium metallurgy studies).

### **5.3.3 Thyroid Counters**

Thyroid counting appears to have started on a limited basis for high risk workers at least as early as 1956. (See also first part of chapter 5 for discussion on thyroid counting in 1945 and 46.) A letter to file, dated June 1960, states, "At the present time routine thyroid monitoring is conducted on a limited basis in the Redox and PUREX facilities. Generally the pattern for coverage in the PUREX facility includes about four to five employees weekly picked from the sampling crews, crane operators, and a Radiation Monitor assigned to the stack area. At the Redox facility routine monitoring is accomplished on a weekly basis for the shift crane operators." (Wilson, 1960) The letter goes to discuss counts and other data obtained in 1959; however, there is no indication if those results were placed in workers' files. Radiation Monitoring data sheets from 1956 show that results below 10 nCi for  $^{131}\text{I}$  were recorded as "less than." The first mobile whole body counter also had a thyroid counter consisting of a 3-in. by 3-in. NaI detector (assumed to mean 3-in. diameter by 3-in thick) that was positioned right next to the neck. The MDA was given as 0.020 nCi for  $^{131}\text{I}$  for a 30-minute count. The exact same detector and MDA were included in a description of in vivo counting capabilities at the Whole Body Counting Facility in 1971 and again in 1985.

For counting  $^{125}\text{I}$  in the thyroid, a thin, 2-in.diameter NaI crystal with a beryllium window was used starting at least as far back as 1967. The thickness of the crystal has not been uncovered yet. The MDA was listed as 0.11 nCi for a 1-minute count or 0.07 nCi for a 10-minute count, but there was no

mention as to which count time was regularly used. There probably were not many workers exposed to  $^{125}\text{I}$  on a regular basis; however, there are indications of a contamination spread in 1978 involving several workers. The same counter is described for thyroid counting in 1982, except that the "reporting level" is given as 0.020 nCi; it's not known if this better sensitivity came from a longer count time, better positioning, or an improved crystal.

By 1985, thyroid counting for  $^{125}\text{I}$  was performed using two intrinsic germanium detectors, with an MDA of 0.005 nCi for a 2000-second count.

Thyroid counting for either of the iodine isotopes has been rare since 1987.

#### **5.3.4 Head Counters and Other Counts**

Miscellaneous counts have been performed over the years at Hanford, including wound counts, head counts, liver counts, lymph node counts, and various longitudinal scans with collimated detectors to pinpoint the location of external or internal contamination. Results of these will show in the database almost always listed as special counts associated with known intakes.

Since the mid 1980s, for intakes of plutonium or americium, head counts have been used to correct chest counts for activity in the bones of the chest region. Since the mid 1990s liver counts were added to the protocol for correcting chest counts to account for possible shine from the liver.

Routine head counting for  $^{90}\text{Sr}$  or  $^{147}\text{Pm}$  did occur for awhile in the 1970s. These were not very sensitive plus there is the question as to what a head count means relative to the activity in the total skeleton. Hopefully the same worker will have  $^{90}\text{Sr}$  urinalysis results. The latter should be given preference as to confirming or quantifying an intake.

#### **5.3.5 General Notes about Items in the Database**

All in vivo results appear to be given in nCi. "Limits" were MDAs, which were treated the same as decision levels until 1992. The decision level is listed under "limits" starting January 1992.

Sometimes a radionuclide is listed without a value or limit. This probably means a "trace" was found. More information may be available on the In Vivo Counter Results Form if it was sent to the worker's personal radiation exposure history file. If not, assume the result of the count is 100 nCi.

Prior to the advent of GeLi detectors, when a significant peak in a whole body count of a radionuclide not  $^{24}\text{Na}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ , or  $^{65}\text{Zn}$  occurred, the activity of the trace or "interfering" radionuclide may or may not have been quantified. Additionally, the activity of one or more of the regular four radionuclides may have been marked as invalid because of overlap with the interfering peak or because of impact of the interfering peak on the spectrum stripping calculations. For the small activities involved, there is no merit in trying to recalculate or estimate actual quantities. It is claimant favorable to use the activities of  $^{137}\text{Cs}$  as given plus include the activity of the interfering radionuclide as given as well. Use 100 nCi for the interfering radionuclide if not given directly.

Which radionuclides were routinely reported to the database changed over the years. From the beginning until 1983,  $^{24}\text{Na}$ ,  $^{40}\text{K}$ ,  $^{137}\text{Cs}$ , and  $^{65}\text{Zn}$  were the only routinely reported radionuclides, with only  $^{137}\text{Cs}$  being of interest to the dose reconstructor. In 1983, as part of the switch to the ORE database, only  $^{40}\text{K}$  and  $^{137}\text{Cs}$  results (or the MDAs) were routinely reported; in late 1987  $^{60}\text{Co}$  was added. In 1995, with the start-up of a new spectrum analysis software program (NEXEC), the standup counter's energy spectrum was divided into 12 regions and a radionuclide was assigned to

each region, including more naturally-occurring radionuclides such as  $^{214}\text{Bi}$  and  $^{208}\text{Tl}$ . During this time if a worker had a count using the coaxial HPGe whole body counter, up to 20 radionuclides may have been listed in the records. The listing of that many radionuclides was simply a bookkeeping approach, and had nothing to do with the sources of exposure. Because of the shutdown of the last reactor in 1986, radionuclides such as  $^{59}\text{Fe}$ ,  $^{24}\text{Na}$ ,  $^{22}\text{Na}$ ,  $^{144}\text{Ce/Pr}$ , and  $^{131}\text{I}$  had decayed away to negligible levels at Hanford (unless a researcher was using a small source for studies). The lack of the need to report all these radionuclides routinely, unless a peak was actually present, was recognized, and when NEXEC was replaced by Abacos (October 1999), the routinely reported list was reduced to  $^{40}\text{K}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{154}\text{Eu}$ . **Reporting of radionuclides at levels below the MDA or decision level should not be interpreted as implying exposure to those radionuclides.**

For chest counting, the database usually lists  $^{234}\text{Th}$  as the potentially measured radionuclide as an indicator of  $^{238}\text{U}$ . Until recently, routinely reported radionuclides for chest counting were  $^{241}\text{Am}$ ,  $^{234}\text{Th}$ , and  $^{235}\text{U}$  for anyone receiving a chest count. This does not imply exposure to both plutonium/americium mixtures and uranium. Very recently, workers have been scheduled for types of chest counts based on their exposure in the workplace so that for plutonium workers, for instance, only the  $^{241}\text{Am}$  results are determined and reported.

## 5.4 MIXTURES

Except in a few facilities in the weapons production cycle (such as B Plant/WESF after 1968,  $\text{UO}_3$  Plant), bioassay methods did not measure all the radionuclides in the intake mixture. Emphasis was on measuring exposure to radionuclides having the greatest impact relative to radiation protection standards (for instance, MPBB or CEDE), or radionuclides that were most common. Unmeasured radionuclides generally do not have a big impact on dose but might target different organs or might have a larger relative impact over times less than 50 years. Hence, this section attempts to estimate possible mixtures of radionuclides that might have been part of an intake that was indicated by a measured radionuclide. **In all cases, where actual bioassay data are available, those data should be used in preference to the following conservative mixtures.**

Plutonium isotopic mixtures and uranium isotopic mixtures are discussed in sections 5.2.1 and 5.2.5, respectively.

Fission and activation product mixtures up through 1987 (when N Reactor shut down) were much more complex and variable. The fission product urinalysis procedure measured beta activity from any radionuclides of strontium, yttrium, barium, lanthanum, cerium, europium, and promethium. It was calibrated for the  $^{90}\text{Sr}/^{90}\text{Y}$  betas so would have underestimated soft beta emitters, but the uncertainty associated with calibration is overwhelmed by the uncertainty in guessing what the mixture was truly composed of and what the abundances of unmeasured radionuclides were. The relative abundances of radionuclides in a potential intake of mixed fission and activation products varied according to location (reactors, fuel separation facilities, waste management facilities), type of fuel, enrichment of fuel, amount of burn-up, and cooling time (i.e., time since removal of the fuel from the reactor). Reactor operators were most likely exposed to activation products, but contamination from leaking fuel rods, especially in fuel storage pools, cannot be ruled out. Even the exposure to activation products was considerably uneven and depended on which reactor components a worker had recently worked on, especially for radionuclides such as  $^{51}\text{Cr}$  or  $^{46}\text{Sc}$ .

Due to complexity mentioned above, there is no straightforward way to determine the true makeup of an intake that resulted in a high fission product urinalysis result. However, many of the principal fission products target the same organs. For instance, of the top 20 fission products (by activity) produced in Hanford reactors from 1945-60, isotopes of Sr, Y, Nb, Zr, Ce, La, Sb, Sn, Pm, Pr, Te, Nd,

Eu are bone seekers; Y, Nb, Ce, Pm, Pr, Nd, Eu are also liver seekers; Zr, La, Sb, Sn also distribute a significant component to "all other tissues," Cs and Ru distribute evenly to all tissues except, because of small uptake from the gut, Ru delivers more dose to the GI tract organs; Ba principally affects the GI tract; and I concentrates in the thyroid.

#### 5.4.1 Reactor workers:

Reactor workers would have been principally exposed to activation products; however, as yet it has not been determined how workers were monitored for possible intakes of activation products until 1960, except by air sampling. Air samples were analyzed for total alpha and total beta so the mix of activation and fission products was not determined. The fission product urinalysis did not measure activation products. Because of once-through cooling water, effluents from the reactors show lots of short-lived radionuclides, but an interview of a health physicist who worked at the early reactors (Marvin Smith) revealed that workers were not allowed into the rear face and rear tubing area, where short-lived activation products were present, for 12 hours after shutdown to allow for decay. So, workers were most likely exposed to the usual mix of activation products:  $^{54}\text{Mn}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Fe}$ ,  $^{51}\text{Cr}$ . The list of radionuclides detected in the first couple years of whole body counting also showed  $^{46}\text{Sc}$ , and  $^{99\text{m}}\text{Tc}$  (indicator for  $^{99}\text{Mo}$ ). Another 100 Area health physicist (G. Yesberger) also said that the workers wore assault masks when working on contaminated parts of or equipment associated with the reactors. Both men said that airborne contamination was never an important consideration, dose-wise, relative to the high external exposure rates in those areas.

Based on what little information has been uncovered, if an intake of anything other than  $^{131}\text{I}$ ,  $^{24}\text{Na}$ , or  $^{65}\text{Zn}$  is indicated for a worker at the reactors, prior to 1960, the dose reconstructor should assume intakes of  $^{46}\text{Sc}$ ,  $^{51}\text{Cr}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ ,  $^{99}\text{Mo}$  occurred as well. The amounts of such intakes would not necessarily correlate well to any fission product radionuclides. Instead about 100,000 pCi (intake) of each would be a conservative estimate, which is roughly 1% of the MPBB of each.

$^{154}\text{Eu}$  and  $^{155}\text{Eu}$  were activation products of concern at N Reactor because of activation of samarium balls used for neutron flux control. These radionuclides would have been included in the fission product urinalysis or would have been detected in whole body counts. If one is detected, then assume the intake included an equal amount of the other.

#### 5.4.2 Separations Plants

The following guidelines may be used to determine intakes from fission product urinalysis results if no other information is available about the radionuclide composition of the contamination. (See Attachment D 3.2 for basis for guidelines).

(Assume the following absorption types: Ce type M, Y type M, Sr type F, Nb type M, Zr type M, Ru type F, Pm type M.)

##### 1944 - 1955

Bone: Calculate the intake assuming the fission product activity is  $^{141}\text{Ce}$ . Then add intakes of the following (in multiples of the  $^{141}\text{Ce}$  intake): 1.0  $^{91}\text{Y}$ , 0.5  $^{89}\text{Sr}$ , 1.5  $^{95}\text{Nb}$ , and 0.5  $^{103}\text{Ru}$ . Calculate the dose assuming that the cerium intake is  $^{144}\text{Ce}$ .

Liver: Calculate the intake assuming the fission product activity is  $^{141}\text{Ce}$ . Then add intakes of the following (in multiples of the  $^{141}\text{Ce}$  intake): 1.5  $^{95}\text{Nb}$ , 1.0  $^{91}\text{Y}$ , and 0.5  $^{103}\text{Ru}$ . Calculate the dose assuming that intake is  $^{144}\text{Ce}$ .

GI: Calculate the intake assuming the fission product activity is  $^{141}\text{Ce}$ . Add intakes (in multiples of the  $^{141}\text{Ce}$  intake): 1.5  $^{95}\text{Nb}$ , 1.0  $^{95}\text{Zr}$ , 1.0  $^{91}\text{Y}$ , and 0.5  $^{103}\text{Ru}$ .

Lung: Calculate the intake assuming the fission product activity is  $^{141}\text{Ce}$ . Add 1.5  $^{95}\text{Nb}$ , 1.0  $^{95}\text{Zr}$ , 1.0  $^{91}\text{Y}$ , 0.2  $^{147}\text{Pm}$ . Calculate the dose assuming that the intake is  $^{144}\text{Ce}$ .

All other organs: Calculate the intake assuming the fission product activity is  $^{141}\text{Ce}$ . Add intakes of 1.5  $^{95}\text{Nb}$ , 1.0  $^{95}\text{Zr}$ , 1.0  $^{91}\text{Y}$ , 0.2  $^{147}\text{Pm}$ . Calculate the dose assuming that the intake is  $^{144}\text{Ce}$ .

### **1956 - 1960**

Bone: Calculate the intake assuming the fission product activity is  $^{144}\text{Ce}$ . Add (in multiples of the  $^{144}\text{Ce}$  intake): 0.6  $^{91}\text{Y}$ , 0.4  $^{89}\text{Sr}$ , 0.8  $^{95}\text{Nb}$ , 0.1  $^{90}\text{Sr}$ , and 0.6  $^{106}\text{Ru}$ .

Liver: Calculate the intake assuming the fission product activity is  $^{144}\text{Ce}$ . Add: 0.8  $^{95}\text{Nb}$ , 0.6  $^{91}\text{Y}$ , and 0.6  $^{106}\text{Ru}$ .

GI: Calculate the intake assuming the fission product activity is  $^{144}\text{Ce}$ . Add: 0.8  $^{95}\text{Nb}$ , 0.7  $^{95}\text{Zr}$ , 0.6  $^{91}\text{Y}$ , and 0.6  $^{106}\text{Ru}$ .

Lung: Calculate the intake assuming the fission product activity is  $^{144}\text{Ce}$ . Add 0.8  $^{95}\text{Nb}$ , 0.7  $^{95}\text{Zr}$ , 0.6  $^{91}\text{Y}$ , 0.4  $^{147}\text{Pm}$ .

All other organs: Calculate the intake assuming the fission product activity is  $^{144}\text{Ce}$ . Add 0.8  $^{95}\text{Nb}$ , 0.7  $^{95}\text{Zr}$ , 0.6  $^{91}\text{Y}$ , 0.4  $^{147}\text{Pm}$ .

### **1961 - 1972**

Because whole body counting became routine, whole body counts can be used to determine intakes of fission or activation products. If any of the main gamma-emitting fission products were detected and  $^{90}\text{Sr}$  urinalysis was not obtained, determine the intake of the gamma-emitting radionuclide, then add (in multiples of the gamma-emitting radionuclide intake) 1.0  $^{90}\text{Sr}$ , 7.0  $^{89}\text{Sr}$ , 12  $^{91}\text{Y}$ , and 4.0  $^{147}\text{Pm}$ .

### **1973 - 1983**

During the period when PUREX was shutdown, 1973 through 1983, considerable facility upgrades and maintenance activities were conducted; hence, exposures to contamination continued, but the mixture would not have contained much activity from short half-life radionuclides. Ratios from 2-year-cooled N Reactor 6% fuel would be conservative with the adjustment that  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  would have built up in contamination over the lifetime of the plant more than  $^{144}\text{Ce}$  or  $^{106}\text{Ru}$ . Consequently it would be claimant favorable but reasonable to assume equal intakes of the major five radionuclides left in the mix, i.e., if an intake of any one of these was incurred, then assume an equal intake of the others:  $^{144}\text{Ce}$ ,  $^{137}\text{Cs}$ ,  $^{147}\text{Pm}$ ,  $^{106}\text{Ru}$ ,  $^{90}\text{Sr}$ .

### **1984-1989**

When PUREX restarted in November 1983, it mostly processed very long cooled fuel or blends of old fuel with some fuel cooled at least 180 days. Either way the short-lived beta-emitting contamination in the plant was not significant. If either of the cerium isotopes or ruthenium isotopes were detected, then because of the difference in MDAs,  $^{137}\text{Cs}$  should have also been detected. If not, it means that the cesium was reduced in the mixture due to processing activities and can be ignored. However, if  $^{90}\text{Sr}$  urinalysis was not obtained in approximately the same time period, then it is claimant-favorable to assume (relative to the cerium or ruthenium intake) 0.6  $^{90}\text{Sr}$  and 1.0  $^{147}\text{Pm}$ .

## **Post 1989**

Because N Reactor shut down in 1987, fission product contamination at PUREX would have been almost entirely  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{147}\text{Pm}$  in approximately equal activities.

### **5.4.3 Waste Management Facilities (tank farms, evaporators, transfer lines)**

By the time waste products reached tank farms and were further manipulated by the evaporators, B Plant, and settling in the tanks, ratios among fission products could be vastly different than in the fuel going into the separations process.  $^{144}\text{Ce}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  are usually associated with the supernatant and generally more available as contaminants. Plutonium,  $^{90}\text{Sr}$ ,  $^{147}\text{Pm}$  are associated with the sludge. Lacking any other information, assume an intake of one radionuclide of the first group (supernatant or general contamination) exists in a mixture of the following: equal activities of  $^{144}\text{Ce}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ , 0.1  $^{99}\text{Tc}$ , 0.1  $^{90}\text{Sr}$ , and 0.001  $^{241}\text{Am}$ . Assume an intake of one radionuclide of the second group (sludge) exists in a mixture of the following: equal activities of  $^{90}\text{Sr}$  and  $^{147}\text{Pm}$ , 0.1  $^{137}\text{Cs}$ , 0.001  $^{239}\text{Pu}$  and 0.001  $^{241}\text{Am}$ . (Carbaugh, 1995) For absorption types, use either M or S for cerium, F or S for ruthenium, F for strontium, cesium and technetium, M for americium, and M or S for plutonium and promethium, depending on whether the organ of concern is associated with the respiratory tract or GI tract (S) or all other organs (F or M).

## **5.5 INTERFERENCES, UNCERTAINTIES**

### **5.5.1 Contamination of Samples**

Home collection of excreta samples started very early in the bioassay program; hence, contamination of excreta samples can be assumed to be negligible. Laboratory contamination and mix-up of samples in the laboratory are a possibility, although laboratory Quality Control procedures and performance of test samples were designed to minimize this source of contamination.

It is likely that a contaminated sample will show up as an obvious outlier in the dataset for a given worker. If the dataset shows an unusually high urinalysis result for a radionuclide other than tritium or uranium, and if follow-up samples were collected that were not consistent with the high result, then the high result may be considered an outlier. However, if the result is not obviously an outlier, then it is claimant-favorable to assume the result is real. For plutonium, if a single high result exists in the database and later (even years later) bioassay results at lower MDAs exist that do not show detection, it is reasonable to assume the high result was an outlier.

For in vivo measurements, contamination can occur as external to the body or, in the case of chest counting, as external to the lung. If a follow-up in vivo count is obtained the same day or within a few days that shows a dramatic decrease in activity or no detectable activity, then external contamination can be assumed. Radon progeny and medical diagnostic or therapeutic procedures involving radionuclides can cause interferences to in vivo measurements, especially for NaI detectors. However, unless the count was invalidated or noted as being influenced by such interferences, the results should be used as recorded.

### **5.5.2 Uncertainties**

Uncertainties for the bioassay measurements were included in the database starting in late 1981 for excreta measurements. These are listed in the database under Error and represent total propagated uncertainty (one  $\sigma$ ) including counting uncertainty, yield uncertainty, and various other systematic uncertainties. These should be used when available. For excreta, uncertainty can also exist in the sample date. For routine samples an uncertainty of  $\pm$  two weeks can be assumed. This is because

one sample date is used for the month regardless of when the sample was actually obtained. For special samples an uncertainty of  $\pm$  two days is reasonable unless the sample is within 2-3 days of a known intake.

The time period the sample represents is also a source of uncertainty. Most urine samples at Hanford were 24-hour simulated samples (kit code 1), meaning the sample was collected over two evening-through-morning periods. Medley indicated that this sampling method produced only about half of a true 24-hour sample based on volume for a group of 9 workers over a 3-day period (Medley 1994); however, Hanford collection protocol was based on percent of day not volume so the true bias (when samples were collected according to procedure) was about 75% of a true 24-hour sample. If a worker has enough urine samples to establish the individual-specific excretion pattern, then a sample can be normalized to the individual's expected 24-hour excretion. Generally, the error associated with collection time period results from under collection of a 24-hour volume. It is claimant favorable to normalize a volume that is less than reference man or reference woman; however, volumes larger than reference man or reference woman should be considered 24-hour samples without normalizing.

For in vivo results, uncertainties were not reported until 1986 for detected radionuclides and 1993 for the default set of radionuclides. These were one sigma counting errors until 1995. Total propagated error was determined and submitted to the records since then. The propagated uncertainty includes counting uncertainty, calibration uncertainty, and a generic 5% positioning error (for both whole body and lung). The calibration uncertainty includes the uncertainty in source activity, counting error, decay correction and interpolation using the calibration curve. Uncertainty associated with reproducibly positioning a person to get the same result was studied at Hanford and found to be about 5%. All calibrations are made using phantoms, and there is considerable uncertainty associated with the representativeness of phantoms versus humans. Just recently a study was done for whole body counting at Hanford using a 95th percentile reference man phantom. There was a low bias of about 20% for the coaxial HPGe detector system for 662 and 1332 keV gamma rays. A similar value of uncertainty ( $\pm 20\%$ ) can reasonably be assumed for the other whole body detectors (1-meter arc, shadow shield, and standup counters).

Uncertainties associated with chest counting are reduced by use of different calibrations for different chest wall thicknesses and use of ultrasound to measure chest wall thickness. One study showed a one-sigma uncertainty of about 20% for americium and uranium values in chest counting, not including correction for interferences from bone and liver. Uncertainties would be much higher for an individual with activity in the bone and/or liver. The uncertainty in lung activity estimates affected by contributions from activity in the liver and skeleton would likely range from 100% or more for levels near or below the MDA to 50% or more for activity above the MDA. The uncertainty in the estimate of chest thickness using the height/weight correction was at least 50% for the front/back lung counter.

Based on the above discussion, the assumption provided in the *Internal Dose Reconstruction Implementation Guide* (NIOSH 2002b) is adequate and should be used, namely the standard deviation is 0.3 times the MDA or reporting level, except for chest counts for which 0.5 times the MDA should be used. For results greater than 3 times the MDA or reporting level, the standard deviation can be assumed to be 0.1 times the result, based on Currie's quantification level (Currie 1968). Actual tests for in vivo counts of phantoms show even smaller uncertainty, but 0.1 is good for broad applications. If actual standard deviations or other indications of error are reported with a bioassay measurement result, the reported value should be used.

For intake estimates during the early periods prior to implementation of routine bioassay programs, where intakes are based on scanty air concentration data, a geometric uncertainty of  $x/\div 2$  is reasonable.

## 5.6 WORKERS WITH NO CONFIRMED INTAKES

### 5.6.1 Special Consideration for Plutonium, Americium, and Thorium

Because plutonium, americium, and thorium stay in the body for a very long time, and because the urine excreta curve (activity per day excreted versus days after intake) has a small slope beyond the first year, an intake of plutonium or thorium that might have been missed in the 1950s or 60s because of poor detection capability, missed samples, or poor sampling after a potential intake, can still be confirmed or otherwise by urinalysis obtained years later. This is especially true for type S materials, but even applicable to type M. For instance, the urine excretion curve for type M plutonium, thorium, and americium all decrease only a factor of 3-4 from one year to 4000 days (~11 years) after the intake. So if an intake is suspected but was not confirmed, the dose reconstructor can use the more sensitive urinalysis data obtained much later to determine a worst case intake. The MDA applicable at the later time can be used or, if there are many samples all showing no detection, then 0.5 times the MDA can be used for the urine value.

### 5.6.2 Worst Case Chronic Intakes

Chronic intakes, or frequent, intermittent intakes that can be modeled as chronic, occurred for tritium and uranium. For other radionuclides, very low level, frequent, intermittent intakes may have occurred for the highest risk workers, such as operators and maintenance workers, at the reactors, separations plants, and Plutonium Finishing Plant, but the intakes were below detectability at the time. For radionuclides with long residence times in the body, chronic intakes lead to a slow build-up of activity in the body and a concomitant increase in urinary excretion. For workers with many bioassay results over a long time but no confirmed intakes, a maximum chronic intake can be determined by using the MDA of the last sample as the upper bound of excretion assuming chronic intake for the entire exposure period. The MDA not the decision level should be used for this calculation. The rate of the chronic intake (pCi/day) needed to reach the MDA level of excretion varies with the duration of the intakes. A lower rate of intake is needed to reach the MDA level excretion if the duration of intakes is 20 years as opposed to 2 years, for instance.

Attachment D, Section D.3 provides tables of chronic intakes used to reach MDA levels of urinary excretion for plutonium, americium, and type S uranium. The tables are based on a unit MDA (1 dpm/day). Adjustment of the actual chronic intake is linear with MDA, so if the true MDA is 0.02 dpm/day, the actual intake is 0.02 times the table value. For the period when the plutonium MDA was for total alpha, Table 5.2.1-3 can be used to determine the isotopic composition of the intake. For the period when the plutonium MDA was for  $^{239}\text{Pu}$  directly, Table 5.2.1-3 can also be used to determine the other components of the mixture. Guidance on adjusting for different enrichments of uranium is given in Table 5.2.5-1.

Attachment D Section D.3 also provides tables of chronic intakes used to reach MDA levels of urinary excretion or retained quantities in the whole body for radionuclides that have short-half lives or short retention in the body. For these radionuclides the daily urinary excretion or retained quantity in the whole body does not continue to increase throughout the exposure period; instead, equilibrium is reached quickly. For these radionuclides, if there were changes in the MDA throughout the period of employment, the calculation of daily intakes and cumulative intake must be made separately for each period. Or it is claimant-favorable and faster to just assume the highest MDA applies for the whole exposure period.

For whole body counts, it is unreasonable to assume that a worker was exposed to all the radionuclides potentially reportable simply because an MDA was determined; on the other hand, for

years Hanford only reported  $^{24}\text{Na}$ ,  $^{65}\text{Zn}$ ,  $^{40}\text{K}$ , and  $^{137}\text{Cs}$ , of which only  $^{137}\text{Cs}$  is of concern to dose reconstruction, while other fission and activation products were ignored if less than 1% of a MPBB. A recommended approach would be to use an indicator radionuclide to determine the intake, then add intakes of other radionuclides in the mixture, as discussed in Section 5.4. Since 1987 or 1988 the only whole body count radionuclides of potential exposure have been  $^{137}\text{Cs}$  and  $^{154}\text{Eu}$  (maybe  $^{60}\text{Co}$  into the early 1990s).

Using the mixtures in Section 5.4 does not work well when the worker's work location is unknown or was variable. In that case it is claimant-favorable to pick the fission or activation product that produces the highest dose to the organ of interest rather than using an indicator radionuclide. To assist with this determination, a spreadsheet Radionuclide Chooser that produces 50-year committed doses to various organs from chronic intakes producing MDA-level body burdens has been developed and is recommended (available from the MJW Task 5 manager).

## 5.7 UNMONITORED WORKERS

From the start Hanford has always had a strong radiation protection program and many innovations in health physics were developed at Hanford (for instance, the first shadow shield whole body counter). Developing a robust bioassay program for plutonium and uranium was a major focus right from the start; and at-risk workers were incorporated into the bioassay program as soon as possible. Air sampling programs, on the other hand, were used mostly to detect contamination spreads and to decide if an area had to be "on mask." Air sampling results that were well below mask level (MPC Table 1 values) were generally ignored. Construction jobs were monitored if the work occurred in a contaminated facility and, supposedly, all outside jobs with potential for encountering underground contamination. But construction workers were neither consistently placed on routine bioassay nor consistently scheduled for termination bioassay.

Due to rigorous workplace monitoring the probability that a worker received a large intake of radioactive material that was unmonitored and unnoticed is very remote (once bioassay programs were established). For instance, even among monitored workers, high routine bioassay results were rare except for tritium, uranium, and fission and activation products up through the 1970s. However, especially among construction workers, the probability of unmonitored, small intakes is larger. On the other hand, many workers had jobs that never required them to enter contaminated areas or to do so only rarely on tours or inspections.

Under certain conditions, airborne effluents from one facility became air intakes for other facilities. Also workers were exposed to diluted effluents when walking between buildings or parking lots or while driving on the site. So, workers in buildings who did not enter contaminated or airborne areas and construction workers almost anywhere could have incurred environmental level intakes.

One other applicable point is that up to 1992, workers with even a remote chance of being exposed to external radiation had a dosimeter, in most cases, this was a single-chip TLD, referred to as the Hanford basic dosimeter.

These considerations lead to the following reasonable, yet claimant-favorable, assumptions for unmonitored workers at Hanford.

- If a worker's record shows no bioassay and no evidence of ever having worn a dosimeter, the unrecorded internal dose should be based on environmental intake only.

- If a worker wore a dosimeter, then the unrecorded internal dose would be no greater than that for a worker who was monitored but had no bioassay results exceeding reporting levels.

In the latter case, use the guidance in section 5.6.2 in conjunction with period-specific MDAs levels and facility-specific radionuclides of concern to estimate an upper bound of possible unrecorded intakes. Claimant interview information may be helpful in determining which facilities the person worked at or near.

## REFERENCES

## GLOSSARY

### **active**

A term used in the early writings at Hanford, circa 1940s and 1950s, to mean radioactive. Example, "production of acetylene from the active water, with subsequent measurement of the ionization cause by the tritium beta particle."

### **aging**

In the context of reactor fuel and mixtures of plutonium isotopes, aging refers to the time since  $^{241}\text{Am}$  was separated from the plutonium mixture.

### **cooling**

In the context of reactor fuel, cooling refers to the time since the fuel was removed from the reactor core.

### **reliably detectable activity**

Three standard deviations of the spectral continuum plus has a peak discernable by the naked eye; used in in-vivo counting circa 1980s.

### **simulated**

In the context of urine sampling, means collection of urine from about one-half hour before retiring to bed, through the sleep period, and for about one-half hour after rising for 2 consecutive nights to simulate a 24-hour sample or 4 consecutive nights to simulate a 48-hour sample.