

ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I Dade Moeller I MJW Technical Services

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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
CFR	Code of Federal Regulations
cm	centimeter
d DOE DOL dpm	day U.S. Department of Energy U.S. Department of Labor disintegrations per minute
EEOICPA	Energy Employee Occupational Illness Compensation Program Act of 2000
ft	feet
g	gram
HPGe hr	hyper-pure, intrinsic germanium hour
ICRP in.	International Commission on Radiological Protection inch
keV	kiloelectron-volt, 1,000 electron-volts
L	liter
m MDA mL min mo MPBB MPOB mrem	meter minimum detectable activity milliliter minute month maximum permissible body burden maximum permissible organ burden millirem
Nal Nal-Csl Nal(Tl) Nal/Ge nCi NCRP NIOSH	sodium iodide sodium iodide-cesium iodide phoswich detector thallium drifted sodium iodide sodium iodide/germanium nanocurie National Council on Radiation Protection and Measurements National Institute for Occupational Safety and Health
ORAU ORNL	Oak Ridge Associated Universities Oak Ridge National Laboratory
pCi POC	picocurie probability of causation
SEC SBC	Special Exposure Cohort scanning bed counter

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SRDB Ref ID Site Research Database Reference Identification (number)

TBD	technical basis document
U.S.C.	United States Code
WBC wk	Whole-Body Counting (Facility) week
μCi μm	microcurie micrometer
§	section or sections

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5.1 INTRODUCTION

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

In this document the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384I(5) and (12)]. EEOICPA defines a DOE facility as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program)" [42 U.S.C. § 7384I(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ..." [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) restrict the "performance of duty" referred to in 42 U.S.C. § 7384n(b) to nuclear weapons work (NIOSH 2010).

The statute also includes a definition of a DOE facility that excludes "buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program" [42 U.S.C. § 7384l(12)]. While this definition excludes Naval Nuclear Propulsion Facilities from being covered under the Act, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled "Exposure in the Performance of Duty"] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally-derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external occupational radiation exposures are considered valid for inclusion in a dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposures to be occupationally derived (NIOSH 2010):

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

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

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5.1.1 <u>Purpose</u>

The purpose of this technical basis document (TBD) is to describe internal dosimetry systems and practices at Oak Ridge National Laboratory (ORNL) from late 1943 to the present. It discusses historical and current practices for use in the evaluation of internal radiation exposure of monitored and unmonitored workers. This TBD can serve as a supplement to, or substitute for, individual monitoring data.

5.1.2 <u>Scope</u>

ORNL began operations in early 1943. The startup of the Graphite Reactor and plutonium separation activities in late 1943 introduced the potential for personnel exposures from intakes of radioactive material. Laboratory operations involving radioactive materials increased over subsequent years as ORNL expanded its roles in radionuclide production and development of chemical separations processes.

Development of methods and techniques for internal monitoring (bioassay) was one of the many priorities at ORNL in its early years of operation because such methods simply did not exist. Although ORNL used air sampling and radiological contamination monitoring programs as qualitative indicators of internal exposure, urinalyses for various internal contaminants did not begin at the site until about 1945. (A limited number of *in vivo* measurements appear to have begun at ORNL in 1959.) ORNL maintained early tolerance levels for airborne contamination based on "product" (i.e., ²³⁹Pu) concentrations in the air (Cox 1944). [These early tolerance levels for alpha and beta-gamma contaminants were 3×10^{-11} and $1 \times 10^{-7} \,\mu$ Ci/cm³, respectively, during the mid-1940s.] In addition, the Laboratory later established tolerance levels for materials such as ¹³¹I and noble gases.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.4.

5.1.3 Special Exposure Cohort Class

June 17, 1943, through July 31, 1955

In its evaluation of SEC-00189, NIOSH determined, and the Advisory Board on Radiation and Worker Health concurred, that NIOSH lacks adequate information with which to perform sufficiently accurate dose reconstructions for the following worker class recommended for addition to the SEC (NIOSH 2012):

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked in any area at the Oak Ridge National Laboratory (X-10) in Oak Ridge, Tennessee, from June 17, 1943 through July 31, 1955, for a number of work days aggregating at least 250 work days, occurring either solely under this employment, or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort.

Dose reconstruction guidance in this document for periods before August 1, 1955, is presented to provide a technical basis for partial dose reconstructions for claims not covered within the SEC class through July 31, 1955. NIOSH found that it was infeasible to reconstruct doses for inadequately monitored radionuclides such as uranium, thorium, and fission products. Although NIOSH found that it is not possible to completely reconstruct radiation doses for the proposed class, NIOSH intends to use any internal and external monitoring data that might become available for an individual claim (and that can be interpreted using existing NIOSH dose reconstruction processes or procedures). Therefore, dose reconstructions for individuals who were employed at the ORNL site during the period

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from June 17, 1943, through July 31, 1955, but who do not qualify for inclusion in the SEC, may be performed using these data as appropriate.

For periods within the SEC period, partial dose reconstructions can be performed based on personal monitoring information and/or available coworker intake rates.

5.2 RADIONUCLIDES OF CONCERN

Because of the many diverse processes and experiments that took place there, a complete list of radionuclides workers encountered at ORNL would be difficult to assemble. Radionuclides likely to produce a measureable internal dose include uranium, activation products, fission products, and transuranic elements [1]. The earliest urine sample results that were provided by ORNL were for ²³⁹Pu and ⁹⁰Sr. The electronic data that were provided by ORNL for use in estimating isotopic MDAs came from a project performed in the early to mid-1990s to convert hard-copy data to a dBase IV database. Funding ran out on the conversion project in the mid-1990s; the entire set of site data was never completely converted, but a significant number of results were made available for Project use. In addition, captured information for 1945 indicated 164 urine bioassay samples for plutonium (Wirth 1945). Table 5-1 lists radionuclides that were included in *in vitro* bioassay results from ORNL for the period from 1945 to 1988. The results were provided for estimating minimum detectable activities (MDAs) for various analyses. The source of these data was an electronic database that was created by ORNL in the early 1990s from hard-copy bioassay records and the 1945 plutonium samples. The sample size values in Table 5-1 are the numbers of analyses for that nuclide in the data ORNL provided and the 1945 plutonium samples. These values do not reflect the total number of *in vitro* bioassays that were performed by ORNL in this period because not all hard-copy records are in the database.

Isotope-specific analyses for *in vitro* samples did not become routine until 1989. Before that time, chemical methods were used to separate radioelements as well as practicable, and the materials were assayed in terms of total activity. The measured activity would later be assigned to a predominant nuclide. Therefore, a result from the early years might indicate ⁹⁰Sr, when in reality it includes ⁸⁹Sr. The same is true for early plutonium results and results for transuranic materials. Therefore, "associated" radionuclides are inherently included in such results. Process knowledge of radionuclides present in various work areas was used to assign nuclides to sample results. The present ORNL internal dosimetry program uses a limited number of radionuclides for screening purposes. Positive results are followed up with additional bioassays.

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Nuclide	Sample size	Nuclide	Sample size	Nuclide	Sample size
Am-241	5,670	I-131	41	S-35	10
Am-243	12	Mn-54	2	Sb-125	1
As-74	7	Mo-99	1	Sm-151	11
BG	2	Na-24	3	Sr-85	1
Bk-249	14	Nb-95	3	Sr-89	37
Br-82	2	Np-237	55	Sr-90	12,893
Br-83	2	P-32	166	Tc-99	20
C-14	11	Pa-231	55	Th-232	1,125
Ca-45	4	Pa-233	16	TI-201	1
Ce-144	37	Pa-234	1	TI-204	1
Cf-249	3	Pb-210	2	Tm-170	6
Cf-252	14	Pm-147	80	U-232	1
CI-36	1	Po-210	66	U-233	829
Cm-242	12	Pu-238	65	U-235	3
Cm-244	299	Pu-239	15642	U-238	11,434
Co-60	83	Pu-241	112	U-239	11
Cs-134	1	Pu-242	41	Y-88	5
Cs-137	3,561	Ra-226	333	Y-90	31
Fe-59	9	Ra-228	1	Zn-65	7
Gross alpha	4,875	Rare earths	1,098	Zr-95	20
Gross beta	324	Ru-103	1		
H-3	2,070	Ru-106	65		

Table 5-1. Radioanalytical results between 1945 and 1988 [2, 6].

5.2.1 Solubility Classes

Internal dosimetrists at ORNL provided a list of assumed intake modes and clearance class information for 73 radionuclides. These assumptions, given in Table 5-2, are those used by ORNL when reevaluating historical bioassay results. They are provided as a reference for dose reconstructors, with the caveat that the assumed solubility classes are not known to be based on any specific studies. In general, they merely reflect conservative choices for dose assessment in terms of committed effective dose equivalent. Dose reconstructors therefore should not assume that these classes, which are in terms of the system promulgated in (ICRP 1966), represent choices that are favorable to claimants for tissue-specific dose evaluations. Dose reconstructors should instead make a choice for the radionuclide and tissue(s) of interest using the system of solubility classes in International Commission on Radiological Protection (ICRP) Publication 66 (ICRP 1994) [3].

Material	Intake	
class	mode	Radionuclides
V (Very soluble)	Ingestion	H-3
L (Labeled organic)	Inhalation	C-14
D (Days)	Inhalation	Na-22, P-32, P-33, S-35, Rb-86, Sr-85, Sr-89, Sr-90, I-125, I-129, I-131, Ba-133, Cs-134, Cs-137, Eu-152, Eu-154, W-188
W (Weeks)	Inhalation	Ca-45, Cr-51, Mn-54, Fe-55, Fe-59, Ni-63, Ge-68, Sc-75, Tc-99, Gd-153, Hg-203, Bi-207, Po-210, Ra-226, Np-237, Am-241, Am-243, Cm-242, Cm-244, Cm-248
Y (Years)	Inhalation	Sc-46, Co-57, Co-58, Co-60, Cu-64, Zn-65, Cu-67, Y-88, Y-90, Pd-103, Ru-106, Cd-109, Ag-110m, Pm-147, Ir-192, Os-191, Th-228, Th-229, Th–230, Th-232, Pa-231, U-232, U-233, U-234, U-235, U-236, U-238, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Cf-249, Cf-252

Table 5-2. Solubility classifications used by ORNL for reevaluation of historical bioassay results (McLaughlin 2002).

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5.2.2 Route of Intake and Particle Sizes

Unless additional information is provided, it should be assumed in all cases that the route of intake for internally deposited radionuclides was via inhalation of particles of 5 μ m activity median aerodynamic diameter.

5.2.3 Bioassay Programs

5.2.3.1 *In Vitro* Monitoring Program

ORNL has collected urine and fecal samples from individuals suspected of potential intakes from 1945 to the present. Urine samples have been and still are the preferred method. While fecal samples can provide good supplementary information to determine when an intake occurred, the chemical solubility of the material, and the particle size, there typically is more variation associated with these samples than with urine samples. When fecal samples are obtained with urine samples after a known intake, the results can be used to better understand the intake parameters and provide a more accurate estimate of intake.

Urine samples were collected in the early years of the bioassay program based on the area health physicist's knowledge of field conditions (e.g., known spills or incidents, air and contamination sample results, etc.). This practice of scheduling did not use a specified sampling frequency (Fleming 2004a). A 1961 procedure manual (UCC 1961) references procedures and practices governing the health physics program at that time, including internal and external exposure monitoring. Although referenced, the procedure detailing internal dosimetry was not among the documents available for review.

A similar manual of health physics procedures and practices from 1982 lists a detailed set of requirements for graduated routine sampling, depending on the frequency and extent of a potential exposure (ORNL 1982). Table 5-3 lists ORNL-published routine bioassay monitoring methods and frequencies from 1973 through 1982. NOTE: Historical information of the *in vivo* monitoring program (see Section 5.3) indicates that the whole-body counting frequencies in Table 5-3 were not consistently followed. Discussions with previous site personnel indicate that no formal counting frequency was used at ORNL until the later 1980s (Fleming 2004b).

		Routine sampling categories	
Radioactive material	I	II	III
Pu-241 and alpha emitters	Urine 3–4 wk	Urine 6–13 wk	Urine 6–12 mo
other than uranium	Whole-body 3–4 mo	Whole-body 6–13 mo	Whole-body 6–12 mo
Sr-90 and uranium	Urine 4–6 wk	Urine 4–13 wk	Urine 6-12 wk
H-3	Urine each wk	Urine each mo	Urine each qtr
I-131	Whole-body each wk	Whole-body each qtr	Urine each qtr
Co-60 and Cs-137	Whole-body 3 mo	Whole-body 6 mo	Whole-body each 6–12 mo
All others	Consult w/Internal Dose	e Group	· ·

Table 5-3. Routine bioassay monitoring methods and frequencies from 1973 to 1982^a (ORNL 1982).

a. Frequency of sampling should be in accord with employee's potential for exposure as determined by health physics surveyors. The following is a guide:

Category I Persons actively involved in operations or processes containing quantities of radioactive material, and when there is some evidence of contamination (i.e., positive results from smear and air samples).

Category II Employees working with relatively small quantities of materials that are confined or when there is no evidence of contamination or activity.

Category III Employees working with radioactive material or in the vicinity of material when there is no known exposure but some potential for exposure.

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Section 5.3 discusses the *in vitro* monitoring program and the data it produced.

5.2.3.2 *In Vivo* Monitoring Program

ORNL has collected whole-body, lung-, and wound-counting data for employees since 1959. For the most part, *in vivo* counting was used until the late 1980s to confirm potential intakes from known incidents or identified by the *in vitro* monitoring program. Although Morgan, Snyder, and Struxness (1965) indicated that routine *in vivo* monitoring for all site radiological workers began in 1965, it appears that a formal program did not begin until the mid- to late 1980s. Section 5.4 discusses the *in vivo* monitoring program and the data it produced.

5.2.4 Recordkeeping

ORNL used various formats on paper records to record bioassay results through most of the period of interest. In addition, ORNL has entered much historical *in vitro* monitoring data into a database. These data were used to estimate MDAs as described in this document (see Attachment A). These records will be used by dose reconstructors in estimating intakes.

It is not practicable for this document to provide examples of all record formats ORNL used in the past six decades; MMES (1995) contains a guide to historical record formats. Most bioassay records share the following information: name, badge or other identification number, division code, health physics area, date, analysis code, results (in disintegrations per minute per 24-hr sample), and a reason for the analysis. A review of ORNL claim files indicated that many bioassay forms had slight revisions throughout their use but that the information they contain is similar from one revision to the next and should be easily interpreted.

5.2.4.1 Division Codes

The division code sometimes provides information on individual locations and job assignments. Table 5-4 lists historical division codes, and Table 5-5 lists more recent codes.

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Table 5-4. Historical division codes (MMES 1995).

Γ

Codes	DIVNUM	Departments	Division name
AC	01	3390	Analytical Chemistry
BI	02	4455	Biology
СН	04		Chemistry
CT	03	3370	Chemical Technology
DI	20	3200	Directors
EC, GE	38	3060	Gen. Engr. and Construc.
ED	06	3480	Education
EL	07	3320	Electronuclear
FM	22		Finance and Materials
HE	23	3090	Health
HP	08	3810, 4193, 3193, 3490	Health Physics
IC	09	3341, 3075	Instrumentation and Controls
IE	24		Inspection Engineering
IS	25	3369, 3650, 4362, 3360	Isotopes
LP	26	3094	Laboratory Protection
MA	10	3166, 3516, 3152	Mathematics
MC	11	3470	Metals and Ceramics
MET			Metallurgy
NP	12	3410	Neutron Physics
OP	28	3639	Operations
PE	21	3016, 3078, 3003, 3004, 3062	Plant and Equipment
PH	13	3405	Physics
PI	30	3173	Public Information
PR	29	3107, 3141	Personnel
RC	14	3430, 4430	Reactor Chemistry
RE	16	4435, 3435	Reactor
RP	17		Research Participation
SS	18	3475	Solid State
TH	19	4460	Thermonuclear
TI	31	3072, 3148	Technical Information

5.2.4.2 Radioanalytical Abbreviations and Codes

Paper records contain abbreviation codes for recording analytes. These codes were often based on the initials of the analysis or the isotopic abbreviation of the radionuclide. Tables 5-6 and 5-7 list the codes for measured radionuclides in urine and fecal samples, respectively (Mani 1983) (MMES 1995).

Code	DIVNUM	Division	
AC	01	Analytical Chemistry	
AS	70	Administrative Services	
AT	07	Applied Technology	
BI	02	Biology	
BS	95	Business Systems	
CH	04	Chemistry	
CM	20	Central Management	
CS	63	Computing and Telecom	
СТ	03	Chemical Technology	
EA	93	ESA	
EC	35	Environmental Compliance	
EH	36	Env. & Health Prot.	
EN	15	Energy	
EP	12	Eng. Physics & Math	
ER	29	Employee Relations	
ES	42	Env. Sciences	
ET	16	Eng. Technology	
EX	90	Executive Offices	
FE	19	Fusion Energy	
FM	37	Finance and Materials	
FR	14	Fuel Recycle	
GE	69	ORNL Engineering	
GR	71	Graphics	
HE	23	Health	

Table 5-5. New division codes (MMI

Code	DIVNUM	Division
HS	08	Health & Safety RSRH
IC	09	Instrmt & Controls
IF		Isotopes
IR	43	Info. Resource Org.
IS	72	Isotopes Division
LP	26	Lab Prot.
MC	11	Metals and Ceramics
OC	62	Controller Office
OP	27, 32	Operations
OS	22	Operational Safety
PC	64	Procurement
PE	21	Plant & Equipment
PH	13	Physics
PU	73	Publications
QA	24	Quality
RE	87	Env. Restoration
RP	14	Robotics
RR	06	Research Reactors
RU		Rust
SS	18	Solid State
TR	60	Treasurer Office
VI		Visitor
WM	27	Waste Management

Table 5-6. Urinalysis abbreviations and codes (Mani 1983; MMES 1995).

Alphanumeric code			Imeric code		Alph	anumeric code	Numeric code			
CM0	Cm-244	000	Other	r PA3 Pa-233 beta		017	Np-237			
CO0	Co-60	001	S-35		PH2	P-32	018	Ag-110m		
CS0	Cesium B (Cs-137)	002	Co-60		PM7	Pm-147				
CS7	Cs-137	003	Pb-210		PO0	Po-210				
EU	Eu-154	004	Na-24		PU0, PU	Pu-239 alpha				
FP0, FP	Fission products (Cs- 137)	005	Zr-95/Nb-95		PU1	Pu-241				
FU0	Total rare earths ()	006	Tc-99		PU9	Pu-239				
GA0, GA	Gross alpha (Pu-239)	007	As-74		RA0	Ra-226				
GB0, GB	Gross beta (Sr-90)	008	Br-82/Br-83		RU6	Ru-106				
GD0	Gd-153	009	Fe-59		SR0	Sr-90				
GG0	Gross gamma (Cs-137)	010	Mn-54		SR5	Sr-85				
GU0	Gross alpha (Pu-239)	011	I-131		SR9	Sr-89				
HY3	H-3	012	Cs-132		TA0	Ta-180				
125	I-125	013	Gross beta		TH	Th-232				
131	I-131	014	Ba-140		TP0	Trans plutonium alpha (Am-241)				
NP0	Np-237	015	Sb-125		TRE	Total rare earths ()				
PA0	Pa-230 alpha	016	TI-204]	URO	U-234 alpha				

The ORNL database contains values for isotopic activities during times when isotope-specific analyses were not possible or routinely performed. In earlier years, the element of concern was extracted chemically from the biological sample and the total radioactivity of the element in the extract was measured. At some point after the extraction and sample count, the total sample activity was attributed to a specific radionuclide. Many of the isotopic assignments were based on process knowledge.

Table 5-7.	Fecal analysis codes (Mani
1983; MME	

1303,	1905, MINILO 1995).						
	Alphanumeric code						
GF0	Pu-239 gross alpha (includes Th)						
PF0	Pu-239						
RF0	Rare earths ()						
SF0	Sr-90						
SF9	Sr-89						
TF0	Trans plutonium (Am-241)						
UF0	U-234						
OF0	Other						

5.2.4.3 Whole-Body Counting Results Codes

Table 5-8 lists codes used on some older cards and forms for documenting whole-body counting results. Many hard-copy records have been consolidated into individual personal records folders. However, this compilation is incomplete, with records only for employees with last names beginning with A through G.

	Before 1971	1971–1978					
Code	Description ^a	Code	Description ^a				
1	Normal human spectrum	0	= <15% MPOB				
2	Less than 10% MPBB	2	= <25% MPOB				
3	Less than 25% MPBB	4	= <50% MPOB				
4	Less than 50% MPBB	6	= <100% MPOB				
5	Greater than 50% MPBB	8	>100% MPOB				
		N	Insignificant & indeterminate				
		S	Significant & indeterminate				

Table 5-8. Result codes for whole-body counting [4].

a. MPBB = maximum permissible body burden; MPOB = maximum permissible organ burden.

Recovered data show that analytical MDAs tended to remain fairly consistent for a number of years. Abrupt changes in MDAs can be identified for groups of radionuclides during specific years. After these changes, the MDAs remain generally consistent in subsequent years. This "step-wise" pattern allowed MDAs from several years to be grouped to obtain a single, representative MDA. Table A-2 in Attachment A provides annual averages for periods when bioassay data were available.

To reflect the performance of the instrumentation and analytical methods more accurately during the period before 1990, Table 5-9 assigns MDAs for some isotopes to their corresponding radioelement rather than the specific radionuclide. For example, routine separation of the alpha emitters ²³⁸Pu and ²³⁹Pu did not occur until alpha spectrometric analyses became routine in 1989, but the recovered database reports both separately. They have been combined in this document as "Plutonium." NOTE: Plutonium-241 is reported separately because it is a beta emitter that can be assayed separately from the alpha-emitting isotopes of plutonium.

		Urine MDAs (dpm/24h sample)									Fecal MDAs (dpm/24h sample)											
	Gross	Gross	Am-	Cm-	Cs-		I-	Np-	Pm-			Pu-	Ra-	Rare	Ru-	Sr-89 +		Gross	Am-	Cm-		Th-
Year	alpha	beta	241	244	137	H-3	131	237	147	Plutonium	Polonium	241	226	earths	106	Sr-90	Uranium	alpha	241	244	Plutonium	232
1943																						
1944																						
1945										3.1												
1946																						
1947										0.38												
1948										0.38							1.4					
1949										0.38							1.4					<u> </u>
1950					33					0.21						34	1.4					0.53
1951			0.33		33					0.21				129	0.30	34	6.3					0.53
1952	0.26		0.33		33					0.21			31	129	0.30	34	6.3					0.53
1953	0.26		0.33		33					0.21	58		31	129	0.30	34	6.3					0.53
1954	0.26	1,135	0.33		33					0.21	58		1.3	129	0.30	34	6.3					0.53
1955	0.26	1,135	0.33		197					0.21	58		1.3	129	0.30	34	6.3					0.53
1956	0.26	1,135	0.33		197					0.21	58		1.3	129	0.30	34	6.3	0.64				0.53
1957	0.26	1,135	0.33		197			1.1		0.21	58		1.3	129	0.30	34	6.3	0.64			0.34	0.53
1958	0.26	1,135	0.33		197			1.1		0.21	58		1.3	129	0.30	34	6.3	0.64			0.34	0.53
1959	0.26	1,135	0.33		197			1.1	396	0.21	58		1.3	129	0.30	34	6.3	0.64			0.34	0.53
1960	0.26	1,135	0.33		197			1.1	396	0.21	58		1.3	129	53	34	6.3	0.64			0.34	0.53
1961	0.26	288	0.33		197	320,282		1.1	396	0.21	17		1.3	129	53	6.3	6.3	0.64			0.34	0.53
1962	0.26	288	0.33		197	320,282		1.1	396	0.21	17		1.3	129	53	6.3	6.3	0.64			0.34	0.53
1963	0.26	288	0.33		197	320,282		1.1	396	0.21	17		1.3	129	53	6.3	6.3	0.64			0.34	0.53
1964	0.26	288	0.33	0.22	197	320,282		0.09	396	0.21	17		1.3	129	53	6.3	1.1	0.64			0.34	0.53
1965	0.26	288	0.33	0.22	197	320,282	10,358	0.09	396	0.21	17		1.3	129	53	6.3	1.1	0.64	0.46	1.05	0.34	0.53
1966	0.26	288	0.33	0.22	197	320,282	10,358	0.09	396	0.21	17		1.3	129	53	6.3	1.1	0.64	0.46	1.05	0.34	0.53
1967	0.26	288	0.33	0.22	197	320,282	10,358	0.09	396	0.21	17		1.3	129	53	6.3	1.1	0.64	0.46	1.05	0.34	0.53
1968	0.09	39.8	0.08	0.10	51	55,681	10,358	0.09	396	0.06	17	9.2	1.3	129	53	6.3	0.09	0.64	0.46	1.05	0.34	0.53
1969	0.09	39.8	0.08	0.10	51	55,681	10,358	0.09	396	0.06	17	9.2	1.3	129	53	6.3	0.09	0.64	0.46	1.05	0.34	0.53
1970	0.09	39.8	0.08	0.10	51	55,681	10,358	0.09	396	0.06	17	9.2	1.3	129	53	6.3	0.09	0.64	0.46	1.05	0.34	0.53
1971	0.09	39.8	0.08	0.10	51	55,681	10,358	0.09	396	0.06	17	9.2	0.07	5.4	53	4.0	0.09	0.64	0.46	1.05	0.34	0.53
1972	0.09	39.8	0.08	0.10	51	55,681	10,358	0.09	396	0.06	17	9.2	0.07	5.4	53	4.0	0.09	0.64	0.46	1.05	0.34	0.53
1973	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	17	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1974	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	17	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1975	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1976	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1977	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1978	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1979	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1980	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1981	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1982	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1983	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1984	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1985	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1986	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1987	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1988	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53
1989	0.09	39.8	0.05	0.10	51	55,681	10,358	0.09	396	0.04	0.52	9.2	0.07	5.4	53	4.0	0.06	0.64	0.46	1.05	0.34	0.53

Table 5-9. Recommended in vitro MDAs (dpm/24-hr sample) for radionuclides from 1947 to 1989.

5.3 *IN VITRO* MINIMUM DETECTABLE ACTIVITIES, COUNTING METHODS, AND REPORTING PRACTICES

5.3.1 *In Vitro* Minimum Detectable Activities

5.3.1.1 Minimum Detectable Activities between 1945 and 1989

Table 5-9 lists historical MDAs for radionuclides of concern in urine and feces. These MDAs were calculated from analytical records that were recovered from the *in vitro* sample databases that support the ORNL historic workforce dose assessment project (MMES 1995). In addition, captured information for 1945 indicated 164 urine bioassay samples for plutonium (Wirth 1945). This 1945 data was also used in the MDA calculation for that year. Attachment A contains details of data recovery and additional information on specific radionuclides. Blank MDA entries in the early years in Table 5-9 indicate that no analytical results for that radionuclide were recovered from that year. This could be because the records have not been found, or because the analysis was not performed.

5.3.1.2 Minimum Detectable Activities after 1989

MDA values for present (since 1989) samples are available with each sample. Table 5-10 lists typical, current MDAs for radionuclides of concern in urine and feces.

MDA
(dpm/24-hr sample)
9,100
4,480
1
3
200
20
0.02
0.02
0.02
0.02
0.02
0.02
0.02
0.02
0.02
0.02
0.02
0.02
26

Table 5-10. MDAs for *in vitro* samples after 1989 (McLaughlin 2002).

5.3.2 <u>Counting Methods for *In Vitro* Samples</u>

Several counting methods have historically been available at ORNL for determining radioactivity in *in vitro* samples. The following sections discuss alpha spectrometry, liquid scintillation, zinc sulfide scintillation, gamma spectrometry, and beta counting using a gas flow proportional counter.

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5.3.2.1 Alpha Spectrometry

ORNL uses alpha spectrometry in the analysis of nuclides that decay primarily by alpha emission, with only very low-energy photons or none at all. A tracer is added to the bioassay sample before analysis begins to determine the chemical yield of the process. The radioelements are chemically separated from the sample and electrodeposited on stainless-steel disks. Plutonium and the other transuranic elements can be analyzed sequentially; uranium analysis requires a separate sample.

5.3.2.2 Liquid Scintillation

Liquid scintillation is used for the analysis of low-energy pure beta emitters, specifically ³H and ¹⁴C. One milliliter of a urine sample is mixed with a scintillation cocktail for analysis.

5.3.2.3 Zinc-Sulfide Scintillation

ORNL used zinc-sulfide scintillation counting to count alpha emitters such as the trivalent alpha actinides (see Sections 5.3.3.1 and 5.3.3.2).

5.3.2.4 Gamma Spectrometry

ORNL uses gamma spectrometry to identify and quantify radionuclides that emit photons with energies greater than 60 keV. A high-resolution, hyper-pure, intrinsic germanium (HPGe) detector with a beryllium end-window is used. Urine samples are placed in a 1-L Marinelli beaker, which is placed over the detector for counting. If the total sample volume is less than 1 L, distilled or deionized water is added to bring the volume up to 1 L. Fecal samples were counted directly in the sample counter for screening purposes, and they might have been ashed and placed into a 2-in. Petri dish for quantitative results.

5.3.2.5 Gas Flow Proportional Counter (Beta Counting)

ORNL uses the gas flow proportional counter system for the analysis of strontium. Strontium is chemically separated from the sample and filtered onto a glass fiber filter. The filter is placed on a planchet for insertion into the counter. The counting system does not distinguish between beta energies, so the reported result is total strontium (⁸⁹Sr plus ⁹⁰Sr).

5.3.3 Notes on Measurements of Alpha Emitters

5.3.3.1 Trivalent Alpha Actinides

Before 1989, ORNL did not perform radionuclide-specific analyses for americium, curium, and other high atomic number elements beyond plutonium. [These radionuclides were typically recorded on the HP Body Fluids Analysis Request cards as transplutonium (TPO or TPL).] Rather, the Laboratory separated trivalent alpha actinides as a group and analyzed by zinc-sulfide scintillation counting. Therefore, monitoring of transplutonium elements was unable to differentiate between such nuclides as ²⁴¹Am and ²⁴⁴Cm. The default radionuclide to use with measurements involving trivalent alpha actinides is ²⁴¹Am. The detection sensitivity of the transplutonium analysis technique is not well documented for samples processed before 1985. However, laboratory records suggest that the transplutonium detection level was about 0.2 dpm through 1985.

For analyses after 1985, an estimate of the sample-specific MDA is generally reported. Alpha spectroscopy analysis for transuranic elements in bioassay samples began in the early to mid-1980s. Differentiation by alpha energy separation for isotopes such as ²³⁴U, ²³⁵U, ²³⁸U, ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, and ²⁴⁴Cm became possible.

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5.3.3.2 Plutonium

Before 1989, ORNL did not routinely perform isotope-specific analyses for plutonium. Rather, the Laboratory separated plutonium as an element and analyzed it by zinc-sulfide scintillation counting. Therefore, the historic plutonium analysis technique was unable to differentiate among the alphaemitting ²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu. Site analytical personnel assert that in the early 1980s, positive total plutonium measurements were recounted on the limited number of alpha spectrometers that were available at the site. Although many HP Body Fluids Analysis Request cards in the claims files gave results for ²³⁸Pu and ²³⁹Pu, this was not consistently observed. The default isotope for positive, total plutonium measurements should be ²³⁹Pu [5].

5.3.3.3 Environmental Uranium

The following paragraphs were taken entirely from the *Oak Ridge National Laboratory Internal Dosimetry Program Technical Basis Document* (McLaughlin 2002). They are included as an aid to dose reconstructors in the interpretation of uranium bioassay results for ORNL workers.

Environmental levels of naturally-occurring uranium are found throughout eastern Tennessee. The environmental activity levels in the immediate area surrounding Oak Ridge are sufficiently high such that dietary intake of uranium is detectable with 24hour urine samples. A urinary uranium background study was conducted in the mid 1990's using non-occupationally exposed employees to quantify the range of typical background uranium excretion. Based upon the results of that study, a discrimination level (set at the 99th percentile level) of 0.14 dpm/day was established for both U-234 and U-238 to differentiate between environmental and occupational exposure to uranium. A value of 0.25 dpm/day is applied to total uranium results. Plots of the observed uranium excretion distributions for U-234, U-238, and total uranium are provided below.

An activity ratio of U-234 to U-238 of approximately 2 to 1 has been observed within the analyzed local potable water samples. Activity ratios in this range have been reported for various aquifers (Osmond). Though U-234 and U-238 should be in secular equilibrium within nature, the observed enrichment of the isotope U-234 is believed to be caused by several factors which include the direct transfer of U-238 decay products across a solid/liquid phase boundary by alpha recoil and differences in solubility between uranium decay chain members. Recognizing this trend, uranium bioassay results that are less than 0.2 dpm/day that do not exhibit a U-234 to U-238 activity ratio of 2:1 should be considered suspect and investigated.

5.4 *IN VIVO* MINIMUM DETECTABLE ACTIVITIES, COUNTING METHODS, AND REPORTING PRACTICES

5.4.1 Shielded Counting Room

The ORNL Whole-Body Counting (WBC) facility, sometimes referred to as the In Vivo Gamma-Ray Spectrometer), is in Building 2008 in the northwest corner of the Main Plant area. Several Health Physics Division annual progress reports (Morgan 1961; Morgan, Snyder, and Struxness 1963, 1966) indicate that the WBC facility began operation in June 1960 and another report (Brown, Patterson, and Abee 1971) indicates a May 1960 date; a more recent document (Watts et al. 1995) indicates several counts occurred earlier. Therefore, it is likely that the WBC facility began limited operations in 1959.

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The main counting room has inner dimensions of 10 by 10 by 10 ft. Its walls consist of four layers of pre-World War II steel with a total thickness of 14 in. Some documents called it the iron or big room. The first recorded count in the WBC facility occurred on July 13, 1959, and, "was a background count [conducted] in the corner of the steel room with the door not in place and the roof incomplete" (Watts et al. 1995). The room was completed on July 24, 1959 (with the exception of a 0.125-in. layer of "special low-radioactivity lead" added to all inner surfaces in 1960 to reduce background radiation levels) and was used for a time to conduct background studies of paint and interior samples and to count biological samples such as milk, grass, and cow thyroids. The first recorded lung count of an employee occurred on May 19, 1960. Almost immediately after (May 19 to 20, 1960), it was used to conduct lung counts of three employees involved in an onsite contamination incident. The original counting facility used a 4- by 4-in. Nal (TI) crystal, but it is not clear what the counting geometry was. There is a photograph of an individual laying on a nylon-strapped, aluminum beach chair, but it was not clear whether that was the geometry initially employed in the facility. The "Argonne Chair" counting geometry was set up for whole body counting on February 27, 1961 (Morgan 1961, p. 240). The tubular steel chair was tilted so the individual's body was in a V position with the detector placed approximately 50 cm directly over the hips (Morgan 1961, p. 241).

Before construction of the WBC facility, health physicists realized that there could be problems with siting a low-level radiation counting facility in the plant environment, with radioactive effluents from operations being generated immediately adjacent to the facility. Therefore, ORNL designed and installed a recirculating air treatment system that pumped air from inside the counting room through charcoal traps, cooling coils, and heaters to remove radon, odors, and excess moisture (Brown, Patterson, and Abee 1971; Morgan 1961). A slight positive pressure was maintained in the counting room by using cylinders of "aged" breathing air to make up for leakage from the system.

To improve the detection sensitivity, additional layers of material were applied to the floor in the WBC facility counting room over the years. Morgan, Snyder, and Struxness (1963) indicates that 0.04-in.-thick layers of tin and cadmium were laid over the interior surface of the lead, only on the floor, with a 0.01-in.-thick layer of copper over the tin and cadmium. The original vinyl tile was placed over the copper. These materials were installed to reduce background radiation emitted from the lead shield as a graduating shield to minimize the contribution of low-energy X-rays. In the mid-1990s, the vinyl tile surface was removed and a 0.03-in.-thick layer of stainless steel was placed directly over the other metal layers. Site personnel indicated that the stainless-steel layer was primarily for aesthetic purposes to cover the oxidized copper layer. With the addition of frictional surfaces for slip reduction, this was the latest form of the floor. Table 5-11 lists the construction history of the counting room.

Installation date	Material	Thickness (in.)
1959 (entire facility)	Pre-WW II steel	14
1960 (entire facility)	Special low-radioactivity lead	0.125
1963 (floor only)	Tin	0.04
1963 (floor only)	Cadmium	0.04
1963 (floor only)	Copper	0.01
Mid-1990s	Stainless steel	0.03

Subsequent improvements to the shielded counting room, detectors, and counting geometries are described below. The shielded room was used to conduct all *in vivo* measurements at ORNL until 1992, when a Canberra scanning bed counter (SBC) was installed in Room 16 of Building 2008.

A discussion with Berger (Fleming 2004b), who was responsible for the facility from the mid-1970s until the early 1980s, indicated that although the facility was in operation, a formal *in vivo* monitoring program was not in place until the late 1980s. Before that time, the WBC facility was used almost exclusively to either confirm known or suspected intakes of radioisotopes or for research purposes.

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Selection of individuals for counting was performed by field health physics personnel based on expected contaminants of concern and the probability of exposure until the early 1990s, when the selection of individuals for *in vivo* monitoring became the responsibility of the Internal Dosimetry staff.

5.4.2 <u>Detectors, Geometries, and Techniques</u>

As described above, the initial *in vivo* counter at ORNL used a 4- by 4-in. Nal(TI) crystal and a tilted chair counting arrangement (Morgan 1961). In July 1961, counting activities in the WBC facility were suspended to modify the detector arrangement with the installation of an 8- by 4-in. Nal(TI) crystal to replace the smaller detector to increase efficiency and reduce counting times. In 1962, several thin (5-in.-diameter by 0.0625-in.) Nal(TI) crystals were installed in the facility to quantify low-energy photons (e.g., mainly isotopes of plutonium). Also in that year, calibration studies were conducted using an arc-shaped geometry. This geometry was expected to result in less variation in counting efficiency than the chair geometry. (The arc geometry placed an individual laying in an arced position with the anterior portion of the body facing the detector at a distance of about 1 m, so each portion of the body was approximately the same distance from the detector (Mani 1983).

Morgan, Snyder, and Struxness (1963) indicated that the thin crystal detectors could see approximately 40 nCi of ²³⁸Pu if there was a preexposure chest count. If there was no preexposure count, the detection capability was approximately 80 nCi. In 1963, an SBC replaced the chair geometry using the 8- by 4-in. Nal(TI) crystal for whole-body counting. The bed and individual were moved under the stationary detector. This geometry was used to determine roughly the part of the body in which the gamma-emitting radioisotopes were located. The detection efficiency was approximately equivalent to that of the chair geometry (Morgan, Snyder, and Struxness 1963). Morgan, Snyder, and Struxness (1963) noted that the computer output provided a "gross" spectrum as well as having the ability to "strip the ⁴⁰K and ¹³⁷Cs background counts." Several spectra observed during that period indicated a large portion of the "net" spectra with negative values. Morgan, Snyder, and Struxness (1964) noted that in March 1963 "written weekly reports of *in vivo* counting activities and results was begun." The research to generate this document did not find any of these weekly reports.

Morgan, Snyder, and Struxness (1965) reported that "Baseline counts on essentially every person with a potential for future exposure was completed in May 1965." By the third week of May 1965, a prioritizing system for selecting individuals for whole-body counting was initiated. Table 5-12 describes this system. Although several located documents stated that baseline and specified monitoring frequencies were used to make *in vivo* measurements, Fleming (2004b) indicated that a full *in vivo* monitoring program did not exist at ORNL until approximately 1994, when site internal dosimetrists became responsible for identifying personnel for counting. Before that time, the area health physicists selected individuals for *in vivo* monitoring. The area health physicists were responsible for determining radioisotopes to which the worker could have been exposed and counting frequencies. This led to inconsistent approaches to the selection of individuals for monitoring.

Priority	Selection criteria
1	Persons suspected of having sustained exposure
2	Persons being recounted as follow-up to initial elevated in vivo results
3	Persons who work directly with radioactive material (once every 3 mo)
4	Persons who work in areas where radioactive materials are handled, but do not work directly with the material (once every 6 mo)
5	New hires or other persons requiring a baseline count and a limited number of persons prior to termination

Table 5-12. 1965 selection criteria for whole-body counting.

Morgan, Snyder, and Struxness (1966) indicated that the "job of determining which employees should be counted, the frequency with which they should be recounted, and preparing the necessary

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schedule cards and lists is requiring more time than had been anticipated." For this reason, the WBC facility was still being used to verify if intakes had taken place rather than for routine monitoring of site personnel. A new Health Physics Division Report (Morgan, Snyder, and Davis 1967) begun in 1966 included data on numbers of *in vivo* counts and basic statistics for *in vivo* monitoring. It included numbers of individuals whose results exceeded the U.S. Atomic Energy Commission (AEC) reporting level (50% of the permissible body burden averaged over the year) or some other specified amount. Table 5-13 summarizes this information. NOTE: The use of annual averages could be misleading in cases involving short-lived or rapidly cleared materials. Mani (1983, Table 15) provided similar data but reported all *in vivo* counts that exceeded specified levels rather than annual averages. Table 5-14 lists this information.

Table 5-13. Qualitative information concerning radioactive material detected using the WBC facility	,
from 1966 to 1983.	

	Number of persons exceeding the permissible body burden
Year	(based upon <i>in vivo</i> measurements)
1966 ^a	0 persons exceeded 50% of permissible body burden
1967 ^a	0 persons exceeded 50% of permissible body burden
1968 ^a	0 persons exceeded 50% of permissible body burden
1969 ^a	0 persons exceeded 50% of permissible body burden
1970 ^b	0 persons exceeded 25% of permissible body burden
1971 ^b	0 persons exceeded 25% of permissible body burden
1972 ^b	0 persons exceeded 25% of permissible body burden
1973 ^b	0 persons exceeded 25% of permissible body burden
1974 ^b	2 (employees inhaled Cm-244 believed to range from 20 (15 pCi) to 40% (30 pCi) of organ (lung)
	burden)
1975 [⊳]	4 (1 - Cm-244 of ~50%, 1 – U-238 of <15%, 1 – Zn-65 and Co-57 of <15%, and 1 – Co-60 of <15% of
	lung burden)
1976 ^b	6 (all six appear to have had detectable amounts of Cm-244 of <15% of the lung burden)
1977 ^b	20 (8 $-$ ²³⁹ Pu and ²⁴¹ Am of <25%, 8 $-$ ¹³⁷ Cs, ⁶⁰ Co, ¹⁵³ Gd, and ¹⁰⁹ Cd of <15%, and 4 $-$ ¹²⁵ I and ¹²³ Te
	<25% of the lung burden)
1978 ^b	6 (1 - Pu-241 and Am-241 of \sim 70% (lung), 1 – ¹³¹ I-131 minor amount that could not be quantified
	(thyroid), and 4 – Co-60 of <25% (lung)
1979 ^b	0 persons exceeded 25% of permissible lung burden
1980 ^b	0 persons exceeded 10% of permissible lung burden
1981 ^b	0 persons exceeded 10% of permissible lung burden
1982 ^b	0 persons exceeded 10% of permissible lung burden
1983 ^b	0 persons exceeded 10% of permissible lung burden
a. The	AEC permissible level was 50% of the permissible body burden from 1966 through 1969.

b. From 1970, ORNL indicated in reports that in vivo measurements did not exceed specified amounts.

Table 5-15 summarizes the maximum activity measured through *in vivo* monitoring for various nuclides for the period from 1961 to 1966.

The results for a given *in vivo* analysis contained the following information: name, badge number (or equivalent), division code, health physics area, date, analysis code, and results code. Table 5-8 lists result codes for the period before 1978. The reason for the analysis was given. The ORNL Division codes are the same as those listed in Tables 5-4 and 5-5. [NOTE: The ICRP Publication 2 methodology of reporting percentages of body or lung burdens (ICRP 1959) was used at ORNL through 1983.] AEC Manual Chapter 0502 required "an evaluation of the radiation exposure status of an employee when monitoring techniques indicated that a body burden equaled or exceeded 50% of a maximum permissible limit".

Count year	Insignificant and indeterminate	Significant and indeterminate	<15% of MPOB	Normal spectrum	<10/25% of MPOB ^a	<25% of MPOB	<50% of MPOB	>50% of MPOB	<100% of MPOB	>100% of MPOB	Total counts
1962				59	9	3					71
1963				906	15						921
1964				1,485	6	20	13	31			1,555
1965				1,111	16	8	23	11			1,169
1966				632	11	12	28	13			696
1967				918	13	6	25	4			966
1968				815	135	12	6				968
1969				905	126	8	18				1,057
1970				499	14	2	1	2			518
1971	310	4	9					1			324
1972	255		4								259
1973	321	1	3								325
1974	246	2	8		3		6		7	6	278
1975	297	23	5		4		1				330
1976	215	6	29				3			1	254
1977	256	14	15		5		1				291
1978	257	12	30		3		3		1		306
Total	2,157	62	103	7,330	360	71	128	62	8	7	10,288

Table 5-14. Qualitative information concerning radioactive material detected using the WBC facility through 1978 (Mani 1983).

a. 10% before 1971 and 25% from 1971 to 1978.

Isotope	1961	1962	1963	1964	1965	1966
Na-24			140			
Sc-46						4
Cr-51		320		42	Trace ^a	
Co-56					15	
Co-57						65
Co-58		20		38	Trace	
Fe-59		40		15		
Co-60	80	13	2	220	5	12
Co-64				<5		
Zn-65		40	3	16	Trace	
Se-75				250		
Sr-90			40	4,500	800	626
Zr-95/Nb-95	30	11	14	47	18	39
Ru-106/Rh-106		131		47	200	30
Sb-125		162	6	18		
I-131	12	200	28 ^b	73	54	6
Cs-137	440	360	570	310	104	92
Ce-144/Pr-144		30		<7	75	50
Eu-155				Trace		
Au-198						
Hg-203	800	0.0005			169	22
Ra-226				<5		< 0.5 [°]
Pa-233	2,800					
U (enriched)				Trace		Trace

Table 5-15. Maximum measured *in vivo activity* (nCi) from 1961 to 1966.

a. *Trace* is not defined within any of the annual reports, but is provided here to give a qualitative "feel" for the amount present.

b. Thyroid.

c. Micrograms.

Since 1989, each *in vivo* measurement showing a positive result has been assessed, including making estimates of intake and dose. The *Oak Ridge National Laboratory Internal Dosimetry Program Technical Basis Document* (McLaughlin 2002) states that dose estimates of less than 1 mrem were reported as zero. The overwhelming number of *in vivo* measurements obtained at the facility indicate no elevated activity.

Mani (1983) indicated that a NaI-CsI phoswich detector was installed and operational in 1967, although Brown, Patterson, and Abee (1971) state, "The several detectors used for measuring gamma radiation from the human body are all NaI(TI) crystals." The 1974 annual report of the Health Physics Division (Auxier, Davis, and Turner 1975) stated that a phoswich detector was in place and apparently was installed between 1971 and 1974, but no other information was found stating when the detector was placed in the WBC facility. A 9- by 9-in. Nal(TI) crystal was installed in the WBC facility in 1974 (Auxier, Davis, and Turner 1975) and placed under the chest area of the counting bed. This detector was used for stationary chest counts. [A statement in Brown, Patterson, and Abee (1971) indicated that a distortion of the spectrum occurred if the large detector was used with the bed moving. Thus, it was not used for whole-body scans.] The whole-body scans performed during this period used the 8- by 4-in. detector at a distance of 12 in. from the bed surface and an elapsed counting time of 20 minutes. (If transuranic isotopes were suspected, the thin crystal assembly was used.) Stationary chest counts used either the 8- by 4-in. or thin crystal assembly in contact with the chest depending on what contaminants were expected. When the 9- by 9-in. Nal(TI) crystal was used the individual was positioned either face-up or -down on the bed and the bed was lowered just shy of contact with the face of the large detector. The time to conduct stationary chest counts typically ranged from 10 to 40 minutes and sometimes longer to ensure "better counting statistics" (Brown,

Patterson, and Abee 1971). The arc-counting geometry was described in Brown, Patterson, and Abee (1971) with the 8- by 4-in. detector positioned 6 ft above the center point of the counting bed. This geometry differs from that described in Mani's assessment of ORNL internal dosimetry data (Mani 1983). The counting bed was placed on the floor with the ends supported to give the proper curvature. This geometry was intended for use only when the count rate was too high for the scanning or stationary chest counting methods. (No evidence of the arc-counting geometry actually being used was noted in the references reviewed in the generation of this document.)

In addition to the NaI(TI) detectors described above, Brown, Patterson, and Abee (1971) indicated that three other detectors were in use in the counting room in 1971. A 3- by 3-in. Nal crystal was used to measure radiation emanating from specific organs (e.g., mainly for the thyroid, kidneys, liver, and spleen). Because of its smaller size, it could be placed directly over the organ of interest. Two other Nal crystals were employed in the WBC Facility as wound probes: 2 by 2 in. and 1 by 0.03125 in., with the latter used to measure low-energy photons less than about 200 keV.

The next bulk sources of information describing instruments and procedures in the WBC Facility were provided by Berger and Goans (1981), Berger and Lane (1981, 1982, 1984), and Fleming (2004b). Fleming (2004b) indicated that use of the phoswich detector began in late 1976 to early 1977. Until that time, the three thin NaI(TI) crystal detector assembly was in use for detection of low-energy photons. The phoswich detector had the following dimensions: 5.25 by 0.0625 in. Nal - 2 in. Csl. Mani (1983) indicates that in 1978, an 80-cm² HPGe counting array was in the WBC facility, but it was not fully operational until May 1980 (Berger and Lane 1981). The array consisted of six separate detectors in an aluminum block in a close-packed rectangular array (e.g., a six-die pattern) designed to cover one lung (Berger and Lane 1981). The phoswich and HPGe detectors were used together from the early 1980s to obtain lung count data; they were positioned over the right and left lungs, respectively. Berger and Lane (1984) indicated that there were problems with getting all the HPGe detectors positioned close to the chest surface due to the construction of the aluminum block. Detectors on the chest surface were almost a factor of 3 more sensitive than those farther away from the chest. This lung count geometry was used until approximately 1987, when problems developed with the HPGe/liquid nitrogen feed system. At that time, the HPGe detector array was taken out of service, and the phoswich detector was the only device used for routine chest counts until 1994.

In 1992, a Nuclear Data SBC was put in service, primarily to measure mixed fission and activation products ranging in energy between 100 and 2,000 keV; it is in use today. This counter is in Room 16 of Building 2008, and is the only *in vivo* monitoring device outside the shielded counting room (Watts et al. 1995). It has three germanium detectors facing downward and two Nal detectors on the sides of the bed. As described below, it can also be used as a backup chest counter, if necessary, due to its 3%- to 50% efficient germanium detectors (Watts et al. 1995). The detectors are in a solid center shield. The shield is designed so there is a minimum of 4 in. of lead around each detector.

During this time, the 5.25- by 0.0625-in. Nal - 2-in. CsI phoswich detector was used to conduct chest counts. Sometime between 1977 and 1994, another phoswich detector with similar dimensions was procured and used until three 70% efficient germanium detectors were installed in the counting room in 1994. A chair counting geometry was reinitiated at that time, with the individual reclined 45 degrees from the vertical. The three germanium detectors are positioned with two detectors over the right lung (one over the upper portion and the other over the lower portion) and one over the left lung. The third detector is over the upper portion of the left lung.

In addition to the above, there are three other detector arrangements in the WBC facility. A germanium thyroid counter can be set up using one of the detectors used for lung counting. The individual is in the same chair used for lung counting, but the chair is inclined at a 25-degree angle. Other organ-specific geometries can be used as necessary (skull, liver, hand, etc.). The 5.25- by

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0.0625-in. Nal - 2-in. Csl phoswich and the SBC (in a fixed-bed position) are backup systems that can be used for lung counts if needed.

Until 1994, field health physicists selected individuals for *in vivo* monitoring. At that time, the internal dosimetry group became responsible for the selection of personnel for *in vivo* monitoring with guidance from field health physicists. Fleming (2004b) indicated that the *in vivo* monitoring program might not have been effective for monitoring ORNL personnel until the late 1980s because of reliance on field health physicists to determine the individuals to be monitored.

5.4.3 Cesium Counting Artifact

Many ORNL workers had measured body burdens from intakes of ¹³⁷Cs from nonoccupational sources (e.g., fallout and consumption of local venison). At present, ORNL uses a value of 20 nCi ¹³⁷Cs in a whole-body count for consumers of venison as the decision level to follow up or conduct a dose assessment (McLaughlin 2002). However, the Laboratory has never used it to negate an individual's occupational dose that was received on the site.

Fallout affects everyone in North America, and body burdens of ¹³⁷Cs measurable in whole-body counters were common in the 1960s and 1970s. National Council on Radiological Protection and Measurements (NCRP) Report 94 provides mean body burdens of ¹³⁷Cs for the United States for the years most likely to produce interference with occupational whole-body count results (NCRP 1988). Table 5-16 lists these values.

United	States (NCRP 1988).		
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	1975	1.1
1963	11	1976	1.6
1964	19	1977	1.1
1965	16		

Table 5-16. Mean body burdens of ¹³⁷Cs from fallout in the United States (NCRP 1988).

5.4.4 Minimum Detectable Activity

As stated above, the thin (5-in.-diameter by 0.0625-in.) Nal(TI) crystal array used in 1963 to detect low-energy photons emitted primarily from transuranic elements could detect approximately 40 nCi of ²³⁸Pu, if there had been a preexposure chest count. If there was no preexposure count, the detection capability was approximately 80 nCi (Morgan, Snyder, and Struxness 1963).

Table 5-17 lists MDAs for the HPGe array of six individual detectors in the aluminum block (used around early- to mid-1980s).

Tables 5-18, 5-19, and 5-20 list MDA values for the ORNL SBC, germanium thyroid counter, and fixed bed counter, respectively (Watts et al. 1995). MDA values were calculated for each individual measurement made using the germanium lung counter, SBC, and fixed bed counter. Figure 5-1 is a

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Table 5-17.	MDA values for six-detector
HPGe arrav	(Berger and Lane 1981).

nr Ge anay (nr Ge anay (Derger and Lane 1901).				
Nuclide	Organ	MDA (nCi)			
Pu-239	Lung	21.5			
Am-241	Lung	1.1			
I-125	Thyroid	0.04			
Cs-137	Lung	16.4			
Gd-153	Lung	0.87			
Eu-152	Lung	0.71			
U-233	Lung	1.42			
Co-57	Lung	0.94			

Table 5-18. Current MDA values for ORNL scanning bed counter (Nal/Ge) (Watts et al. 1995).

Isotope	MDA (nCi)	Isotope	MDA (nCi)	Isotope	MDA (nCi)
Cr-51	24/30	Mn-54	2.1/2.4	Co-57	4.9/5.9
Fe-59	3.9/4.5	Co-60	2.2/1.9	Zn-65	4.3/6.0
Rb-88	12/15	Y-88	1.8/1.9	Nb-95	2.1/2.7
Zr-95	3.7/4.1	Tc-99m	4.4/5.3	Ru-103	2.6/2.9
Ru-106	21/30	Cd-109	NG/224*	Sn-113	3.4/4.0
Sb-124	2.2/3.1	I-131	2.7/3.1	Ba-133	3.7/4.5
I-133	2.7/2.9	Cs-134	2.2/3.0	Cs-137	2.4/2.8
Ce-139	4.3/4.6	Ba-140	9.2/9.1	La-140	2.3/2.4
Ce-141	7.7/8.4	Ce-144	36/43	Eu-152	8.5/9.7
Gd-153	21/21	Eu-154	6.1/7.2	Pb-212	6.3/7.6
Bi-214	5.8/9.8	U-235	5.9/6.1		

*NG = not given.

Table 5-19. Current MDA values for

ORNL germaniu	m thyroid counter.
Isotope	MDA (nCi)
I-125	0.06
I-131	0.12

Table 5-20. Current MDA values for ORNL fixed bed counter.

Isotope	MDA (nCi)
Co-60	0.31
Ce-144	6.28

plot of MDA values for different nuclides for the ORNL germanium lung counter as a function of chest wall thickness. Figure 5-2 shows MDA values for ²³⁸Pu and ²⁴¹Am for the ORNL phoswich lung counter as a function of chest wall thickness. Table 5-21 lists the frequencies of *in vivo* counting since 1960 and, for the various detectors, a summary of the best estimates of the applicable periods of use.

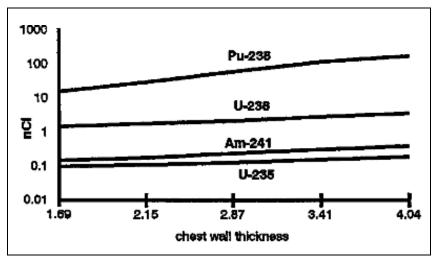


Figure 5-1. Current MDA values (nCi) for ORNL germanium lung counter versus chest-wall thickness (cm) (Watts et al. 1995).

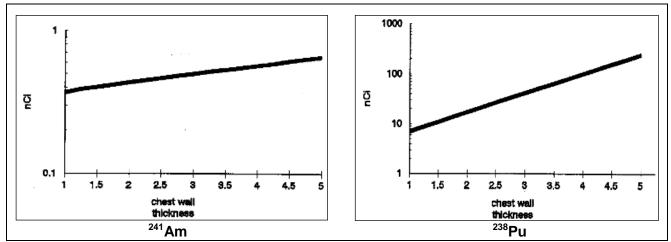


Figure 5-2. Current MDA values (nCi) for ORNL phoswich lung counter versus chest-wall thickness (cm) for ²⁴¹Am and ²³⁸Pu (Watts et al. 1995).

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Table 5-21. Frequency of in vivo monitoring.

Monitoring type		Frequency
	1960–1961	No set frequency. (Period of facility development – limited
	4- x 4-in. Nal(TI)	routine human counting, typically of incidents.)
	1961–1962	No set frequency. (Development activities to change over
	8- × 4-in. Nal(Tl)	to larger detector – typically incidents.)
	1962 to ca. 1967–1971	No set frequency. (Period spent obtaining baseline counts
	8- x 4-in. NaI(TI)	onsite employees and incident counts)
Lung counting	1965–??	Frequency stated in Annual Report (see Table 5-12), but
Lung counting	8- x 4-in. Nal(Tl)	discussions with site personnel indicate that it was not
		consistently implemented.
	ca. 1967–1971 to 1976	Large detector was added below counting table and used
	8- x 4-in. NaI(TI) and 9- x 9-in.	when conducting stationary lung counts only.
	Nal(TI)	
	1962–1976	No set frequency. (Thin Nal crystals used to quantify low
	5-indiameter × 1/16-in. NaI(TI)	energy photons.)
Lung counting	1976–1994	Monitoring frequency was not effectively implemented.
(cont'd.)	5.25- × 1/16-in. Nal - 2- Csl	This phoswich detector was used to monitor for low energy
, ,	phoswich	photons.
	1980–1987	Monitoring frequency was not effectively implemented.
	Germanium array and phoswich	Detectors used together.
	1992-present	Fixed Bed Counter is SBC in fixed position. Used only to
	combination germanium and Nal	measure fission and activation products (e.g., ⁶⁰ Co and
		¹⁴⁴ Ce) immediately after assimilation.
	1994-present	Varies from annual to semiannual depending on exposure
	Germanium system	potential set by Internal Dosimetrist.
	1994–present	Phoswich detector can be used, but germanium detection
	5.25- × 1/16-in. Nal - 2- Csl	system noted above is routinely used.
	phoswich	
	1960–1961	No set frequency. (Period of facility development –limited
	4- x 4-in. Nal(Tl)	routine human counting, typically incidents.)
	1961–1962	No set frequency. (Development activities to change to
	8- x 4-in. Nal(Tl)	larger detector – typically incidents.)
Whole-body	1963–1976	Table 5-12 selection criteria used in 1965 for WBC.
counting	8- x 4-in. Nal(Tl)	
	1992-present	SBC. Monitoring frequency implemented in 1994 to
	combination germanium and Nal	include annual and semiannual counts depending on
	<u> </u>	exposure potential set by Internal Dosimetrist.
	1963-??	Incident counting and research. May have been used for
	3- × 3-in. Nal(Tl)	frequent monitoring of workers potentially exposed to
Thyroid counter		iodine, but this information was not found.
(and other	1994-present	Typically, site research personnel would be placed on this
organs)	germanium (uses one of three	program and monitored on semiannual frequency, as well
5 5 5 7	germanium detectors used for	as persons involved in known or potential iodine intake.

5.5 ATTRIBUTIONS AND ANNOTATIONS

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

- [1] Burns, Robert E. ORAU Team. Senior Health Physicist. April 2007. Section 2 of this Site Profile (ORAUT 2007) provides a more detailed discussion of the key radionuclides workers encountered.
- [2] Burns, Robert E. ORAU Team. Senior Health Physicist. April 2007. This list was compiled from queries of the database of *in vitro* bioassay results provided by ORNL.
- [3] Burns, Robert E. ORAU Team. Senior Health Physicist. April 2007. The system of absorption types promulgated in ICRP Publication 66 (ICRP 1994) is used for all inhalation dose reconstructions under EEOICPA, as this represents use of the best available science. Use of older solubility class information or assumptions to maximize committed dose could result in organ dose assessments that would not be favorable to the claimant.
- [4] Burns, Robert E. ORAU Team. Senior Health Physicist. April 2007. These codes are from worker dosimetry records.
- [5] Burns, Robert E. ORAU Team. Senior Health Physicist. April 2007. The selection of ²³⁹Pu as the default radionuclide, when the true isotopic mix is not known from measurement, is made to ensure assessments favorable to claimants.
- [6] Varnado, Keith W. ORAU Team. Senior Health Physicist. October 2012. Captured documents indicated 164 urine samples for plutonium in 1945. Calculations of 1945 plutonium MDA performed by Tom LaBone. Table 5-9 updated to incorporate a 1945 plutonium MDA value of 3.1 dpm/d.

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GLOSSARY

absorption type

Categories for materials according to their rate of absorption from the respiratory tract to the blood, which replaced the earlier inhalation clearance classes. Defined by the International Commission on Radiological Protection, the absorption types are F: deposited materials that are readily absorbed into blood from the respiratory tract (fast solubilization), M: deposited materials that have intermediate rates of absorption into blood from the respiratory tract (moderate rate of solubilization), and S: deposited materials that are relatively in the respiratory tract (slow solubilization). Also called solubility type.

activation

Creation of a different isotope by bombarding the parent isotope with neutrons, protons, or other types of radiation that results in the formation of a different isotope or element.

alpha radiation

Positively charged particle emitted from the nuclei of some radioactive elements. An alpha particle consists of two neutrons and two protons (a helium nucleus) and has an electrostatic charge of +2.

beta radiation

Charged particle emitted from some radioactive elements with a mass equal to 1/1,837 that of a proton. A negatively charged beta particle is identical to an electron. A positively charged beta particle is a positron.

contamination

Radioactive material in an undesired location including air, soil, buildings, animals, and persons.

curie

Traditional unit of radioactivity equal to 37 billion (3.7×10^{10}) becquerels, which is approximately equal to the activity of 1 gram of pure ²²⁶Ra.

dosimeter

Device that measures the quantity of received radiation, usually a holder with radiationabsorbing filters and radiation-sensitive inserts packaged to provide a record of absorbed dose received by an individual.

dosimetry

Measurement and calculation of internal and external radiation doses.

fission

Splitting of the nucleus of an atom (usually of a heavy element) into at least two other nuclei and the release of a relatively large amount of energy. This transformation usually releases two or three neutrons.

fission product

(1) Radionuclides produced by fission or by the subsequent radioactive decay of radionuclides. (2) Fragments other than neutrons that result from the splitting of an atomic nucleus.

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gamma radiation

Electromagnetic radiation (photons) of short wavelength and high energy (10 kiloelectron-volts to 9 megaelectron-volts) that originates in atomic nuclei and accompanies many nuclear reactions (e.g., fission, radioactive decay, and neutron capture). Gamma photons are identical to X-ray photons of high energy; the difference is that X-rays do not originate in the nucleus.

in vitro bioassay

Measurements to determine the presence of or to estimate the amount of radioactive material in the excreta or in other biological materials removed from the body.

in vivo bioassay

Measurements of radioactive material in the human body utilizing instrumentation that detects radiation emitted from the radioactive material in the body.

ionizing radiation

Radiation of high enough energy to remove an electron from a struck atom and leave behind a positively charged ion. High enough doses of ionizing radiation can cause cellular damage. Ionizing particles include alpha particles, beta particles, gamma rays, X-rays, neutrons, high-speed electrons, high-speed protons, photoelectrons, Compton electrons, positron/negatron pairs from photon radiation, and scattered nuclei from fast neutrons.

isotope

One of two or more atoms of a particular element that have the same number of protons (atomic number) but different numbers of neutrons in their nuclei (e.g., ²³⁴U, ²³⁵U, and ²³⁸U). Isotopes have very nearly the same chemical properties.

neutron

Basic nucleic particle that is electrically neutral with mass slightly greater than that of a proton. There are neutrons in the nuclei of every atom heavier than normal hydrogen.

nucleus

Central core of an atom, which consists of positively charged protons and, with the exception of ordinary hydrogen, electrically neutral neutrons. The number of protons (atomic number) uniquely defines a chemical element, and the number of protons and neutrons is the mass number of a nuclide. The plural is nuclei.

nuclide

Stable or unstable isotope of any element. Nuclide relates to the atomic mass, which is the sum of the number of protons and neutrons in the nucleus of an atom. A radionuclide is an unstable nuclide.

radiation

Subatomic particles and electromagnetic rays (photons) with kinetic energy that interact with matter through various mechanisms that involve energy transfer.

radioactivity

Property possessed by some elements (e.g., uranium) or isotopes (e.g., ¹⁴C) of spontaneously emitting energetic particles (electrons or alpha particles) by the disintegration of their atomic nuclei.

radionuclide

Radioactive nuclide. See radioactivity and nuclide.

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rem

Traditional unit of radiation dose equivalent that indicates the biological damage caused by radiation equivalent to that caused by 1 rad of high-penetration X-rays multiplied by a quality factor. The sievert is the International System unit; 1 rem equals 0.01 sievert. The word derives from <u>roentgen equivalent in man</u>; rem is also the plural.

U.S. Atomic Energy Commission (AEC)

Federal agency created in 1946 to assume the responsibilities of the Manhattan Engineer District (nuclear weapons) and to manage the development, use, and control of nuclear energy for military and civilian applications. The U.S. Energy Research and Development Administration and the U.S. Nuclear Regulatory Commission assumed separate duties from the AEC in 1974. The U.S. Department of Energy succeeded the U.S. Energy Research and Development Administration in 1979.

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A.1 RECOVERY OF HISTORICAL DATA

A large amount of raw bioassay data exists at the ORNL complex. A portion of these data was transcribed into an electronic database hosted on ORNL equipment and maintained by ORNL personnel.

As part of this dose reconstruction effort, this database was queried for historical urinalysis records on October 30, 2003. A supplemental query was performed on December 12, 2003. These queries requested information on the samples and the instrumentation used in the analysis. No personal information or individual dose results were requested or received.

These database queries generated 66,204 records from the period between 1945 and 1988. These records were compiled into a single database. A data validation and comment field was added to each record. Then the records were examined and classified according to usability.

Further NIOSH data capture identified 164 plutonium urine samples taken from February 1945 through May 28, 1945, and include some control samples (Wirth 1945). These 1945 plutonium samples were not identified within the electronic database records. These records were also evaluated separately [3] using the MDA calculational methods described in LaBone (2012) and are not included in Attachment A with the exception of Section A.5.

A.1.1 Data Description

The database queries retrieved 17 parameters from the ORNL internal dosimetry database:

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- NUCLIDE A unique abbreviated identifier for specific nuclides.
- SAMPLEID A unique identifier for individual samples.
- SAMPLETYPE A one-character identifier designating the type of sample (urine, fecal, etc.).
- ALIQUOTVOL The volume or mass of the sample analyzed.
- SAMPLEVOL The sample volume or mass collected.
- COUNTDATE The date the aliquot was counted.
- STARTEDATE The date sample processing began.
- STOPDATE The date sample processing ended.
- RECOVERY The chemical yield, in percent.
- EFFICIENCY The detector's counting efficiency, in percent.
- EFF_FACTOR The reciprocal of the detector's counting efficiency.
- COUNTMIN The sample count time, in minutes.
- T_BKG_RATE The total background count rate, in counts per minute.
- BKG_MIN The background count time, in minutes.
- BKG_RGNT_R The count rate of any reagent used.
- BKG_COUNTS The number of background counts.
- MDA The minimum detectable activity.

A.1.2 Data Classification

The 66,204 individual records in the database were classified into one of four categories:

- **Duplicate Records** 272 duplicate records were found. These were marked with a "D" in the record's validation field.
- Usable without Modification 58,972 of the records collected (89%) were usable without qualification.
- Usable with Modification Some of the records collected contained input errors that were easily identified. Examples include a nuclide entry of C-137, instrument efficiencies listed in fractions instead of percent, and dates from the turn of the century. Other information in the database enabled correction of some of these records. Such modifications were designated by placing an "M" in the records validation field and entering the reason for the change in the comment field. These records comprise 3% (2,193) of the database.
- **Unusable Records** 4,767 of the records recovered (7%) were determined to be unusable in their present form. Most of these records are incomplete or contain gross errors. These were marked with an "R" in the validation field. An explanation for a record's rejection was entered in the comment field.

The resulting database provided all the raw data used to determine the MDAs of radionuclides in urinalysis calculated in this attachment.

A.1.3 Synopsis of Recovered Data

The database queries recovered 61,165 usable records. Of these, 59,288 are clearly records of urinalyses and 1,859 are records of fecal analyses. Special analyses made up the remaining 18 records. Seventy-five different analyte labels are listed, although some are clearly different names for

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the same analyte (e.g. GA, GAO, and G_ALPHA are all gross alpha measurements). Table A-1 lists the recorded analyses performed by radionuclide.

A.2 CALCULATION METHOD FOR SAMPLE MDAS

Equations A-1 and A-2 show the derivation of the estimated MDA, which was calculated from 61,165 analytical records. MDAs were calculated using the method in Brodsky (1992):

$$MDA = \frac{3 + 4.65 \times SD_{b}(\text{counts})}{T_{b}(\text{min}) \times k \left(\frac{\text{counts} \times 24 - \text{hr sample}}{\text{disintegration}}\right)}$$
(A-1)

where

= standard deviation of the total background counts (cpm) SD_b

= duration of the background count (min) T_b

= calibration factor (counts x 24-hr sample/disintegration) k

$$k\left(\frac{\text{counts} \times 24\text{-hr sample}}{\text{disintegration}}\right) = InstrEff\left(\frac{\text{counts}}{\text{disintegration}}\right) \times Yield \times \frac{V_a(\text{mL})}{V_r\left(\frac{\text{mL}}{24\text{-hr sample}}\right)}$$
(A-2)

where:

InstrEff = instrument efficiency (counts/disintegration)

Yield = fractional chemical recovery

$$V_a$$
 = volume of the aliquot analyzed (milliliters for urine, grams for feces)

 V_r = volume of the sample submitted, or the volume of a standard 24-hr sample from Reference Man (1,400 mL/24-hr sample for urine, 135 g/24-hr sample for feces) (ICRP Publication 23 [ICRP 1975] default values for 24-hr voids), whichever is larger

MDAs for radionuclides in urine and feces.										
Nuclide	Fecal	Urinalysis	Row totals							
Totals	1,859	59,288	61,147							
Alpha	1	3	4							
Am-241	130	5,540	5,670							
Am-243	6	6	12							
As-74		7	7							
BG		2	2							
Bk-249	2	12	14							
Br-82		2	2							
Br-83		2	2							
C-14		11	11							
Ca-45		4	4							

Table A-1. Data used to determine historical MDAs for radionuclides in urine and faces

Nuclide	Fecal	Urinolygig	Pow totals				
		Urinalysis	Row totals				
Ce-144	4	33	37				
Cf-249	2	1	3				
Cf-252	3	11	14				
CI-36		1	1				
Cm-242	1	11	12				
Cm-244	26	273	299				
Co-60	2	81	83				
Cs-134		1	1				
Cs-137	4	3,557	3,561				
Fe-59	1	8	9				
GA	69	1,875	1,944				
G-alpha		6	6				
GAO	154	2,771	2,925				
GB		278	278				
GBO		46	46				
H-3		2,070	2,070				
I-131		41	41				
Mn-54		2	2				
Mo-99		1	1				
Na-24		3	3				
Nb-95		3	3				
	1	54	55				
Np-237	1						
P-32	4	166	166				
Pa-231	1	54	55				
Pa-233	3	13	16				
Pa-234		1	1				
Pb-210	10	2	2				
Pm-147	16	64	80				
Po-210	9	57	66				
Pu-238	4	61	65				
Pu-239	124	15,352	15,476				
Pu-241	1	111	112				
Pu-242		41	41				
Ra-226	1	332	333				
Ra-228		1	1				
RE	39	698	737				
Ru-103		1	1				
Ru-106		65	65				
S-35		10	10				
Sb-125		1	1				
Sm-151		11	11				
Sr-85		1	1				
Sr-89		37	37				
Sr-90	142	12,751	12,893				
T	1	,. • .	1				
Tc-99	•	19	19				
Th-232	1,079	46	1,125				
TI-201	1,073	UTU	1,125				
TI-201	1	1	1				
11-204		I I	I				

Nuclide	Fecal	Urinalysis	Row totals
Tm-170	1	5	6
TPU		1	1
TRE	2	197	199
TRE B		1	1
Nuclide	Fecal	Urinalysis	Row totals
TRE-B		4	4
U-232		1	1
U-233	3	826	829
U-235		3	3
U-238	25	11,409	11,434
U-239		11	11
Y+RE		157	157
Y-88		5	5
Y-90		31	31
Zn-65		7	7
Zr-95	1	19	20

To use this equation one must know, or be able to calculate, the standard deviation of background counts, the duration of the count, the yield of any chemical extractions used, the efficiency of the detector used, and the amount of the sample analyzed. The method of recovering this information from the historical database is discussed in the following sections.

A.2.1 <u>Standard Deviation of Background Counts</u>

The data field BKG_COUNTS contained recorded background counts for a sample and was the preferred source of background count information. When a record's BKG_COUNTS field was blank, the background count information was calculated as the product of the T_BKG_RATE and BKG_MIN fields. If the BKG_MIN field was blank, but the T_BKG_RATE field contained a value, the background count information was calculated as the product of the T_BKG_RATE and COUNTMIN fields.

The variability in repeat counts, such as background counts, follows the Poisson distribution (Evans 1955, Chapter 26). It is a property of the Poisson distribution that its standard deviation is the square root of its mean value. Therefore, in this analysis, the standard deviation of the total background counts was calculated as the square root of the total background counts.

A.2.2 Background Count Time

The data field BKG_MIN contained the recorded background count time for a sample and was the preferred source of background count time information. If the BKG_MIN field was blank, the time in the COUNTMIN field was used. This last method of estimating the background count time is likely to be favorable to claimants because in almost all cases where there is paired data, the COUNTMIN value is less than the BKG_MIN value. This produces a higher MDA when substituted into Equation 1.

A.2.3 Instrument Efficiency

Information on instrument efficiencies was contained in two fields. The EFFICIENCY field contained the percent of disintegrations detected by the instrument. The EFF_FACTOR field contained the reciprocal of the EFFICIENCY field.

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A.2.4 <u>Yield</u>

The chemical yield for a given radiochemical analysis was contained in the RECOVERY data field. This value was given in percent recovery.

Many of the records had blank recovery fields. However, due to the repetitive nature of the chemical extractions used, application of an average, radionuclide-specific, chemical recovery to records with no recovery value was judged to be acceptable. This approach is supported further by the lack of variability observed in daily runs on known standards recorded in laboratory logbooks recovered from the late-1950s through the mid-1970s.

A.2.5 Volume of Aliquot Analyzed

The ALIQUOTVOL field usually contained a value for the amount of material subjected to radioanalysis. When this field was blank, it was assumed that the entire sample provided was used in the analysis. The amount of the sample was recorded in the SAMPLEVOL field.

A.2.6 Volume of 24-Hour Sample

Excretion rates and concentrations vary greatly within a 24-hr period and between individuals. The models developed for calculating intakes from *in vitro* analyses are based on 24-hr samples. The daily urine excretion rate is 1,400 mL/24-hr sample; for feces, this value is 135 g/24-hr sample.

Often, the volume analyzed (V_a) was less than the reference excretion rate from Reference Man. When this was observed, the calculated activity in the sample was normalized to the Reference Man volume to compensate. This adjustment increased the MDA and, therefore, would be favorable to claimants.

A.3 CALCULATION METHOD FOR MDAS FOR VARIOUS PERIODS BETWEEN 1945 AND 1988

Once the MDAs were calculated for individual samples, they were segregated by radionuclide and imported into Microsoft Excel spreadsheets. Using Excel, the values were plotted for visual inspection. Figures A-1 through A-18 at the end of this attachment show plots of data the was obtained and calculated for the following radioisotopes: (urine) ²⁴¹Am, ²⁴⁴Cu, ¹³⁷Cs, gross alpha, gross beta, ³H, ¹⁴⁷Pm, ²³⁸Pu, ²³⁹Pu, ²⁴¹Pu, rare earths, ⁹⁰Sr, ²³³U, ²³⁸U, and (feces) ²⁴¹Am, gross alpha, ²³⁹Pu, ²³²Th. In the first plot, the MDA values were plotted against the date of their analysis. This provided a visual representation of individual MDAs over time. The density of the available data and a gross approximation of typical sample MDAs for a given period were quickly observed using this format.

Two bar charts were created for each radionuclide. One shows the annual average MDAs between 1945 and 1988. The other shows the number of analytical results recovered for each year in the same period. These two charts provide additional information on gross trends exhibited by sample MDAs, and on the temporal distribution of the data.

These visual aids were used to establish general periods during which sample MDAs were similar. The sample MDAs in these periods were grouped as similar populations. Using Excel's statistical functions, the following statistical parameters for each group of samples were determined:

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- The population size, ٠
- The 50th-percentile value of the population, •
- The population's arithmetic mean, ٠
- The arithmetic standard deviation of the population, and
- The 95% confidence level on the mean. •

The 95% confidence levels on the mean for each radionuclide are the recommended MDAs for that radionuclide during the period evaluated.

A.4 ADDITIONAL CONSIDERATIONS

The recovered data indicate the analytical MDAs tended to remain fairly consistent for years. Abrupt changes in the MDAs were identified for groups of radionuclides during specific years. After the change, the MDAs remained generally consistent in the several succeeding years. This "step-wise" pattern allowed MDAs from several years to be grouped as one MDA for a specified period. Table 5-9 highlights these groupings by enclosing similar MDAs in a box.

Some MDAs changed frequently, creating a pattern of similar MDAs in short adjacent periods. When this occurred, the largest estimated MDA from that period was selected to be the representative MDA for the entire period. The tritium (³H) MDA listed between 1968 and 1981 in Table 5-9 is an example of this adjustment.

Most recovered data contained isotope-specific entries in the NUCLIDE field. However, the ability to differentiate routinely between isotopes of the same radioelement did not always exist before 1989. Many of the isotopic assignments were based on process knowledge. Therefore, during the period before 1989, the MDAs for some isotopes are reported as the MDAs for the radioelement instead of the radionuclide. For example, the alpha emitters ²³⁸Pu and ²³⁹Pu are reported separately in the database. They have been combined in this TBD as plutonium. NOTE: Plutonium-241 is reported separately because it is a beta emitter that can be measured separately from the alpha-emitting isotopes of plutonium. If the recovered data did not extend to 1989, the MDA calculated from the last known data was extrapolated forward (see Table 5-9).

A.5 **CALCULATION METHOD FOR SAMPLE MDAS – 1945 PLUTONIUM**

NIOSH data capture identified 164 plutonium urine samples taken from February 1945 through May 28, 1945, including some control samples (Wirth 1945). The records were examined and classified according to usability.

The 1945 plutonium MDAs were calculated using the method described by LaBone (2012).

			Urine MDAs (dpm/24-hr sar										mple)							Fecal MDAs (dpm/24-hr sample)				
	Gross	Gross	Am-	Cm-	Cs-		I-	Np-	Pm-			Pu-	Ra-	Rare	Ru-	Sr-89 +		Gross	Am-	Cm-		Th-		
Year	alpha	beta	241	244	137	H-3	131	237	147	Plutonium	Polonium	241	226	earths	106	Sr-90	Uranium	alpha	241	244	Plutonium	232		
1943																								
1944																								
1945												-										<u> </u>		
1946																								
1947										0.33		-					0.005							
1948 1949										0.40		-					0.965							
1949					24.34					0.34						53.95	1.401					0.176		
1950					29.54					0.19				22.24	0.036	22.34	3.600					0.176		
1951		74.44	0.27		34.93					0.18			25.44	31.41	0.030	27.50	5.988							
1952	0.17	/4.44	0.27		29.49					0.23	2.98		25.44	30.45		27.01	6.196							
1954	0.17	351.14			32.90					0.19	9.42		0.32	41.97		29.00	6.296							
1955	0.23	960.93			26.52					0.17	33.32		0.32	41.37	0.142		6.810							
1956	0.22	889.95	0.25		32.05					0.19	57.11		0.87		0.142		7.651							
1957	0.69	998.73	0.44		30.61			1.07		0.22	57.11	1	0.80		0.272	28.08	5.257	0.055			0.072	0.226		
1958	0.35		0.42		38.61			0.26		0.19			0.34		0.255		7.423	0.728			0.012	0.537		
1959	0.31	1321.55	0.35		312.8			0.10	587	0.23			0.29	331.74			5.636	0.647				0.303		
1960	0.19	938.01			147.3			0.27	196	0.18	10.67		0.17	167.87			5.740	0.568				0.573		
1961	0.21	290.98			42.62	71382				0.18	12.74	1	0.29	50.04		5.86	5.686	0.512				0.424		
1962	0.32	252.50				641672				0.14				54.69		8.88	8.687					0.422		
1963	0.45	146.59			72.18			0.49	25.81	0.10				87.30		14.79	7.747	1.075			0.789	0.633		
1964	0.24	198.28		0.57	62.09					0.19					78.91		2.561				0.118	1.052		
1965	0.46	140.95	0.21	0.17	55.66		353.2	0.08		0.14				63.41		13.78	0.992	0.848	0.208	0.172	0.163	0.112		
1966			0.12	0.16	52.71	153077				0.08						6.58	0.128	0.447		0.268				
1967			0.082	0.099	75.55	150699	35718			0.08						7.30	0.405				0.769			
1968	0.056	36.17	0.073	0.080	55.59	54872	1131	0.07		0.07	2.55	25.76	15.70		49.40	6.68	0.112		0.079	1.059	0.349			
1969	0.066	38.82	0.099	0.082	50.80	53036	1281	0.09	23.10	0.07	31.75	7.66		56.96	46.95	4.73	0.094		0.768	0.306	0.536			
1970	0.038	32.31	0.074	0.096	44.32	48763	1140	0.05		0.05		5.67				4.05	0.071		0.097	0.271	0.097			
1971		37.06	0.072			49037		0.06	677	0.06		6.00			2.82	4.33	0.076		0.230		0.351			
1972	0.18		0.046		75.05	45882		0.04	24.06			9.18		5.00		3.95	0.061	0.093	0.035		0.033			
1973	0.10		0.039			50535			6.44	0.04		6.94		5.44		4.14	0.071		0.105		0.060			
1974			0.093			46154			5.72			4.53		4.86		4.80	0.054		0.543		1.073	1.040		
1975			0.040			44192				0.04	0.46			1.49		3.26	0.054		0.047	0.129	0.073	0.046		
1976				0.080	46.12	45554				0.04		1.84	0.014			3.58	0.057		0.173		0.053			
1977			0.037			36984			2.82			9.65	0.049			3.62	0.053		0.121		0.100	ļ		
1978				0.016		31838		0.14		0.03		8.16		3.39		3.30			0.007			───		
1979				0.046		35406		0.03		0.03	0.23	12.40				3.06			0.029		0.032	───		
1980			0.032	0.036		48449										3.05						───		
1981			0.015																			┝───		
1982												 	<u> </u>					ļ				──		
1983			0.000					<u> </u>					ł						l			───		
1984			0.026					<u> </u>					ł						l			───		
1985			0.07																					
1986 1987			0.07																					
1987			0.07																			<u> </u>		
1900	l	1	0.028				1	1	I	1	l	1	L	1				L	I			<u> </u>		

Table A-2. MDAs (dpm/24-hr sample) calculated from recovered data for radionuclides.

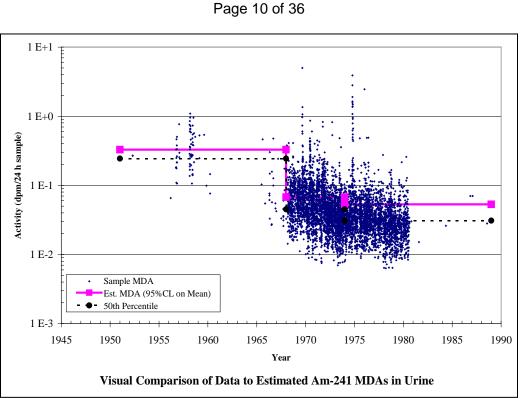


Figure A-1. MDA data for ²⁴¹Am in urine (part 1 of 3).

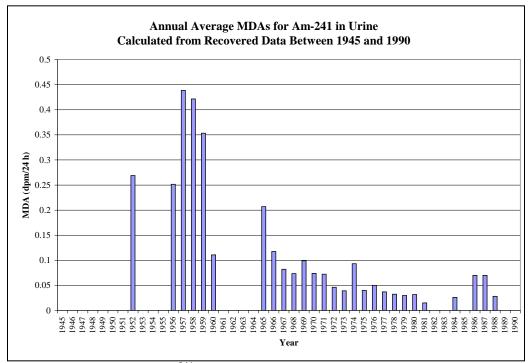


Figure A-1. MDA data for ²⁴¹Am in urine (part 2 of 3).

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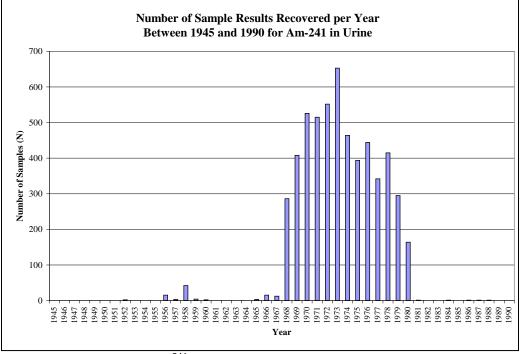


Figure A-1. MDA data for ²⁴¹Am in urine (part 3 of 3).

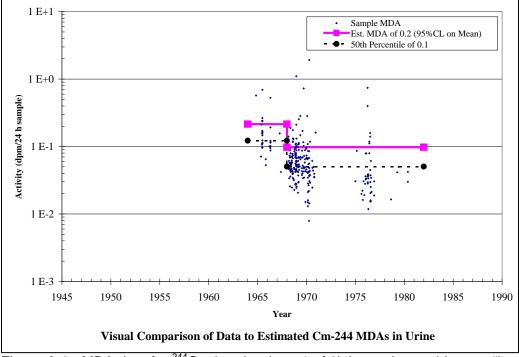


Figure A-2. MDA data for ²⁴⁴Cm in urine (part 1 of 3) (some later tables are "in feces").

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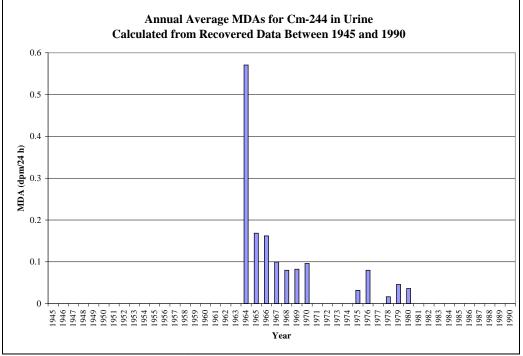


Figure A-2. MDA data for ²⁴⁴Cm in urine (part 2 of 3).

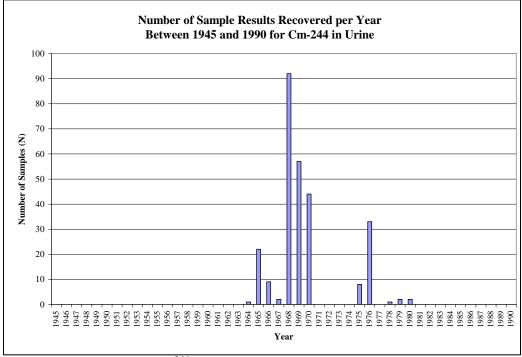
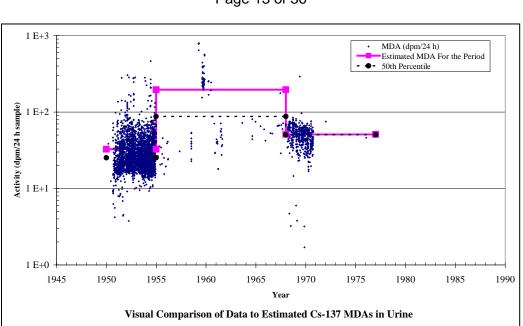
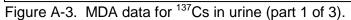


Figure A-2. MDA data for ²⁴⁴Cm in urine (part 3 of 3).



ATTACHMENT A CALCULATION OF MDAs FROM HISTORICAL DATA Page 13 of 36



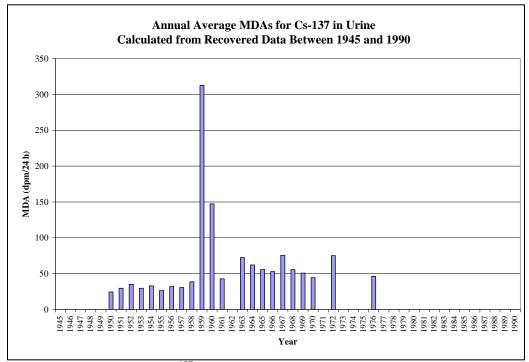


Figure A-3. MDA data for ¹³⁷Cs in urine (part 2 of 3).

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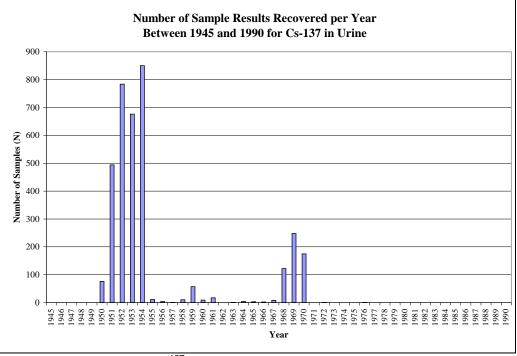


Figure A-3. MDA data for ¹³⁷Cs in urine (part 3 of 3).

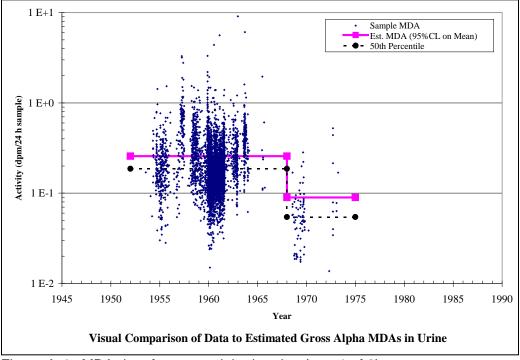


Figure A-4. MDA data for gross alpha in urine (part 1 of 3).

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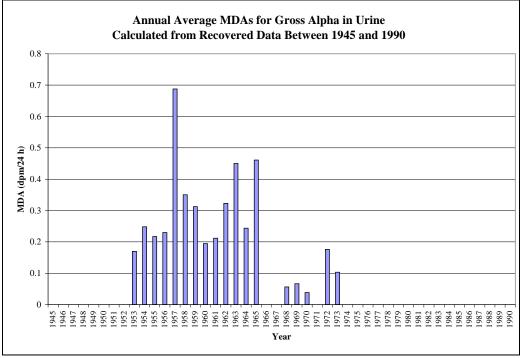


Figure A-4. MDA data for gross alpha in urine (part 2 of 3).

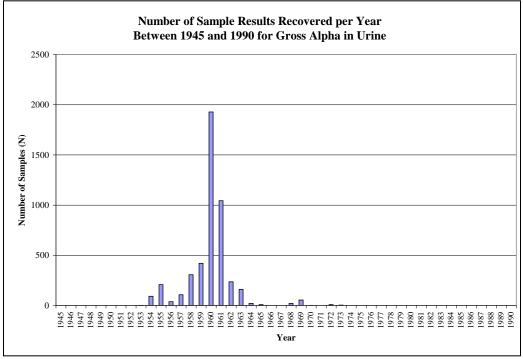


Figure A-4. MDA data for gross alpha in urine (part 3 of 3).

ATTACHMENT A

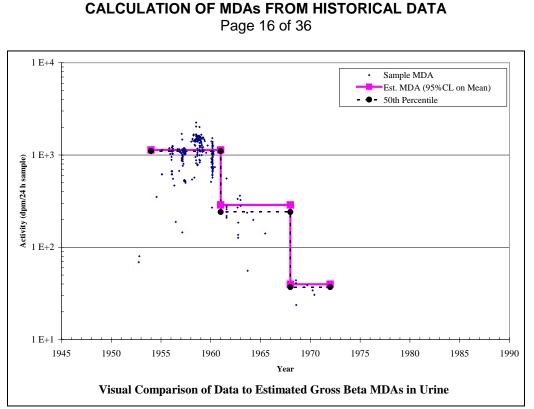


Figure A-5. MDA data for gross beta in urine (part 1 of 3).

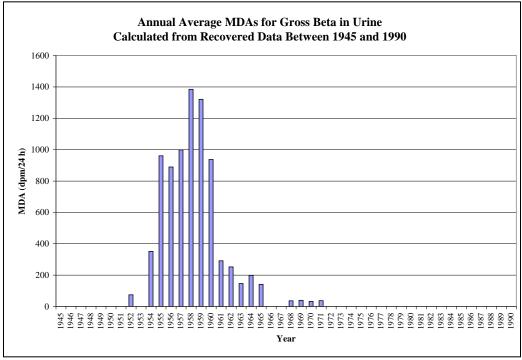


Figure A-5. MDA data for gross beta in urine (part 2 of 3).

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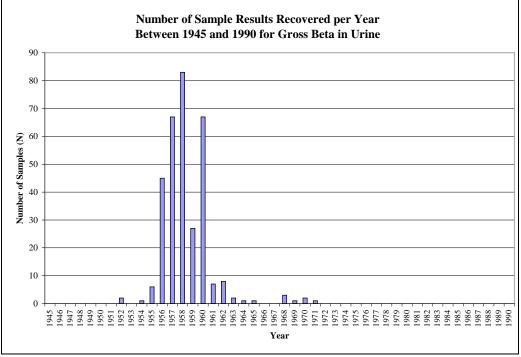


Figure A-5. MDA data for gross beta in urine (part 3 of 3).

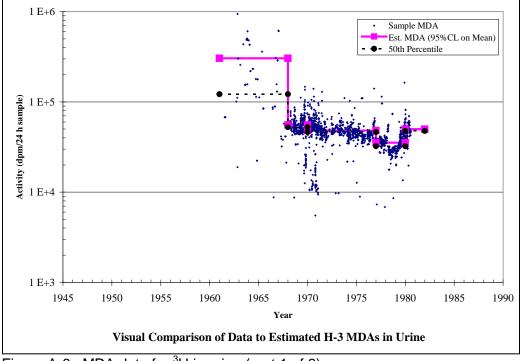


Figure A-6. MDA data for ³H in urine (part 1 of 3).

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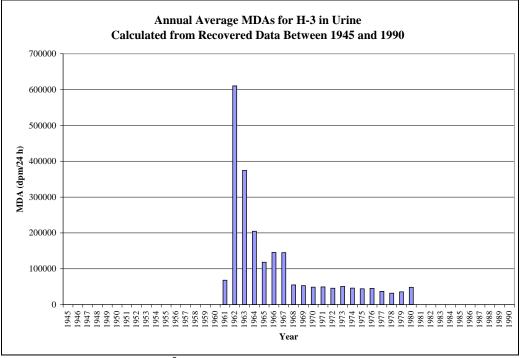


Figure A-6. MDA data for ³H in urine (part 2 of 3).

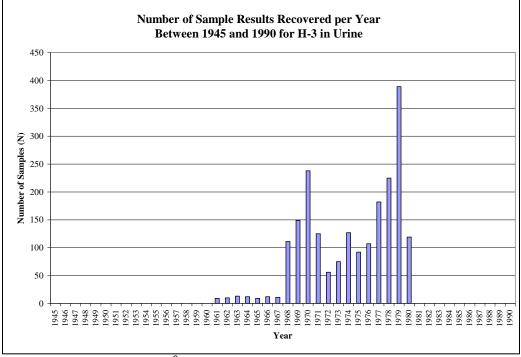


Figure A-6. MDA data for ³H in urine (part 3 of 3).



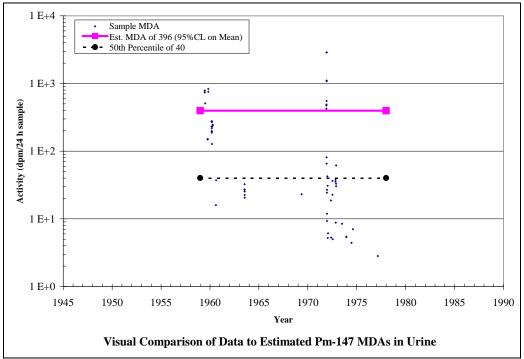


Figure A-7. MDA data for ¹⁴⁷Pm in urine (part 1 of 3).

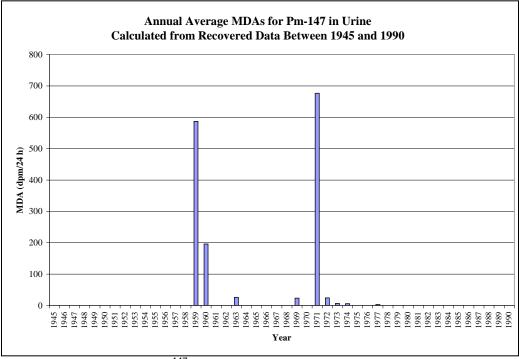


Figure A-7. MDA data for ¹⁴⁷Pm in urine (part 2 of 3).

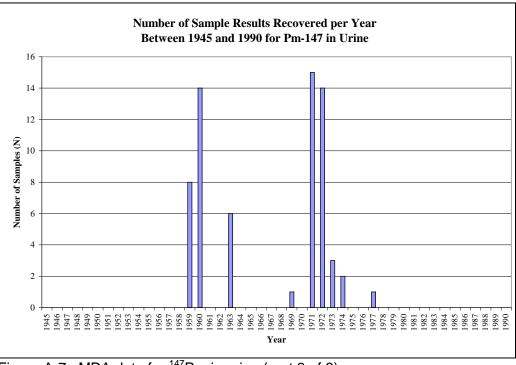


Figure A-7. MDA data for ¹⁴⁷Pm in urine (part 3 of 3).

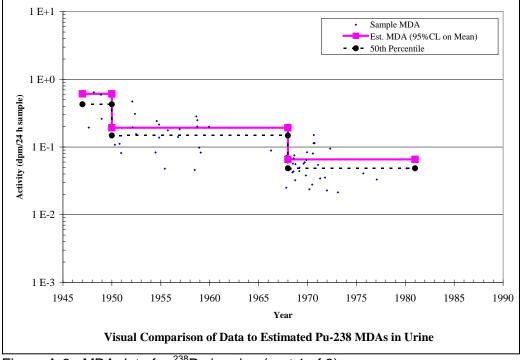


Figure A-8. MDA data for ²³⁸Pu in urine (part 1 of 3).

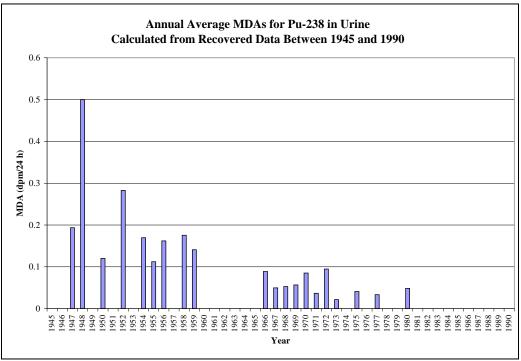


Figure A-8. MDA data for ²³⁸Puin urine (part 2 of 3).

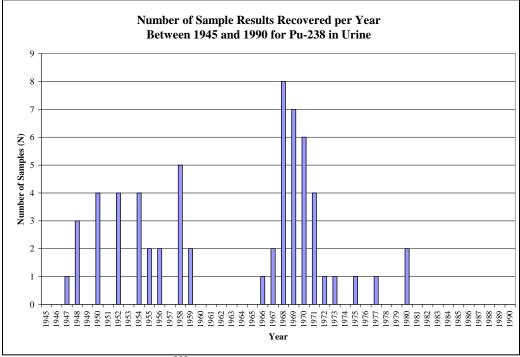


Figure A-8. MDA data for ²³⁸Pu in urine (part 3 of 3).



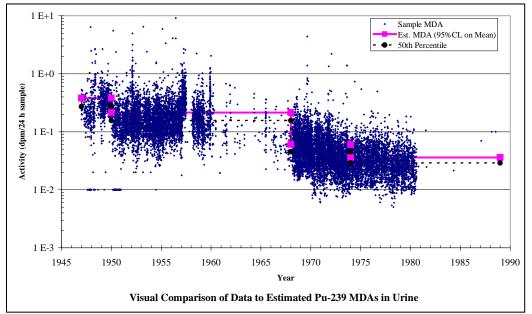


Figure A-9. MDA data for ²³⁹Pu in urine (part 1 of 3).

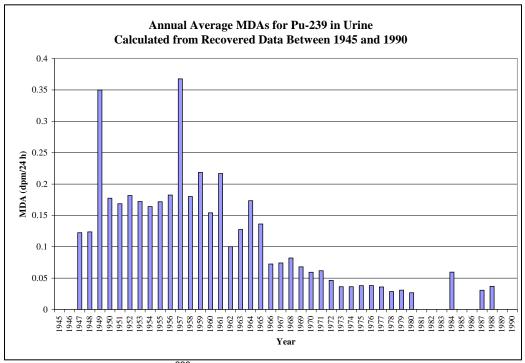
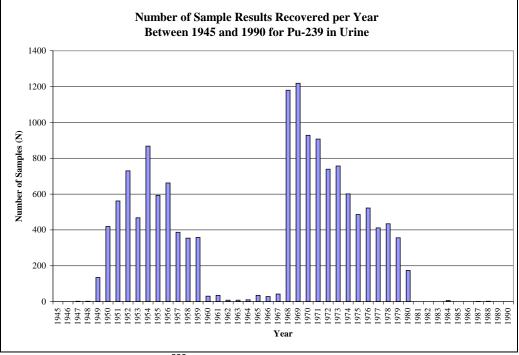
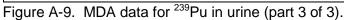


Figure A-9. MDA data for ²³⁹Pu in urine (part 2 of 3).

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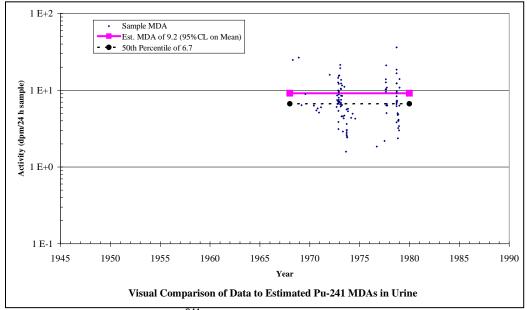


Figure A-10. MDA data for ²⁴¹Pu in urine (part 1 of 3).

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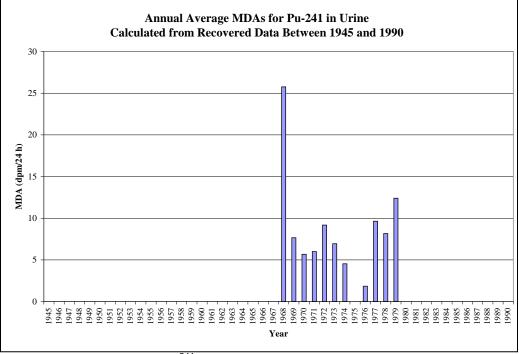


Figure A-10. MDA data for ²⁴¹Pu in urine (part 2 of 3).

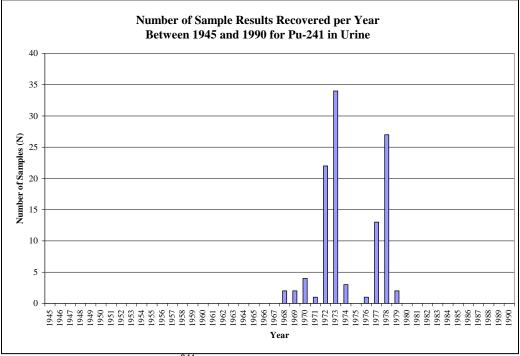
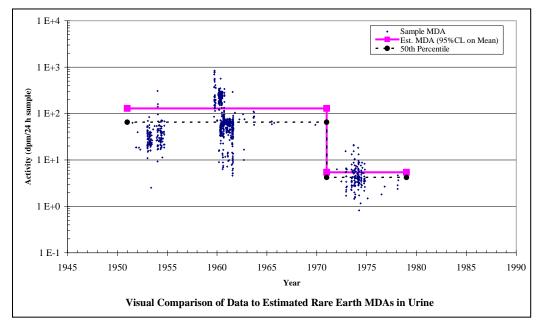


Figure A-10. MDA data for ²⁴¹Pu in urine (part 3 of 3).







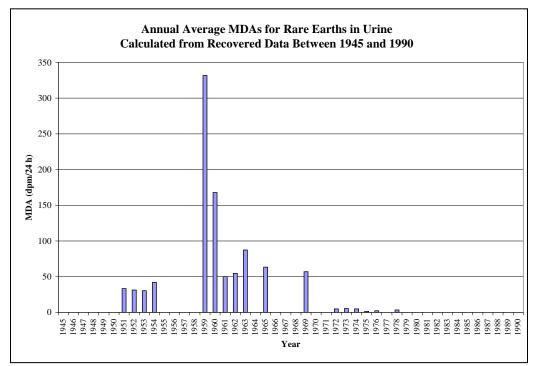


Figure A-11. MDA data for rare earths in urine (part 2 of 3).

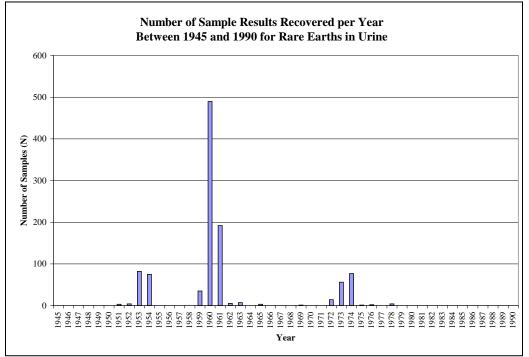


Figure A-11. MDA data for rare earths in urine (part 3 of 3).

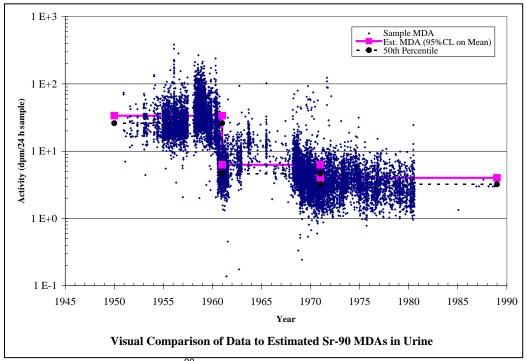


Figure A-12. MDA data for ⁹⁰Sr in urine (part 1 of 3).

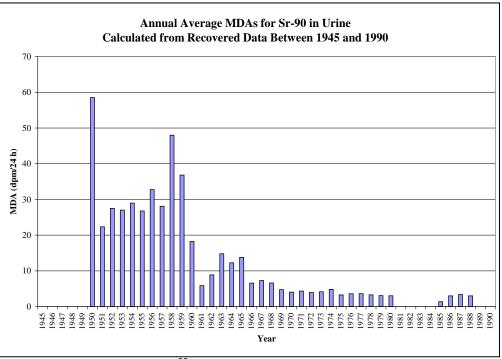


Figure A-12. MDA data for ⁹⁰Sr in urine (part 2 of 3).

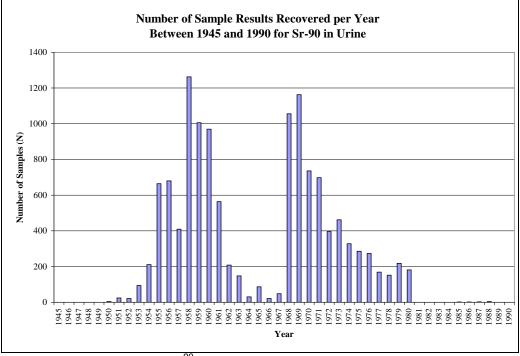


Figure A-12. MDA data for ⁹⁰Sr in urine (part 3 of 3).



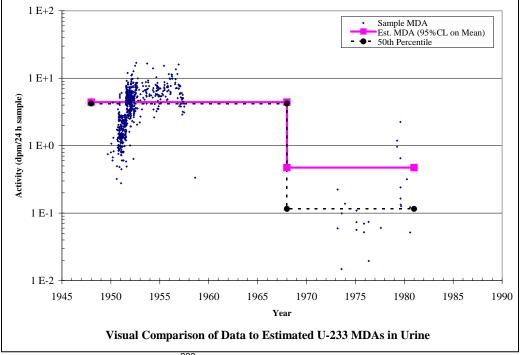


Figure A-13. MDA data for ²³³U in urine (part 1 of 3).

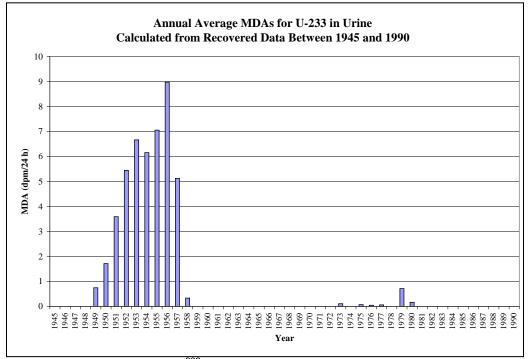


Figure A-13. MDA data for ²³³U in urine (part 2 of 3).

 Number of Sample Results Recovered per Year Between 1945 and 1990 for U-233 in Urine

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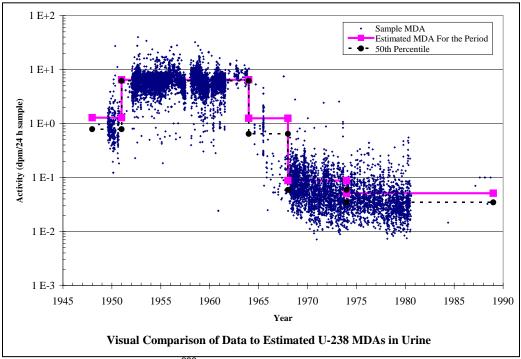


Figure A-14. MDA data for ²³⁸U in urine (part 1 of 3).

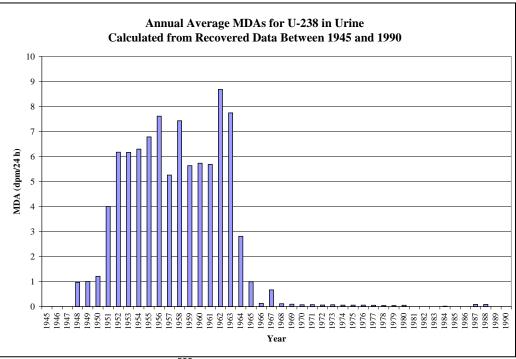


Figure A-14. MDA data for ²³⁸U in urine (part 2 of 3).

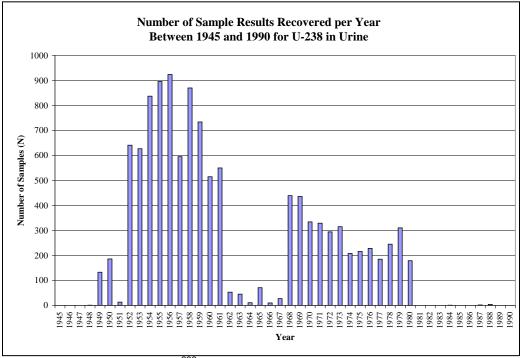


Figure A-14. MDA data for ²³⁸U in urine (part 3 of 3).

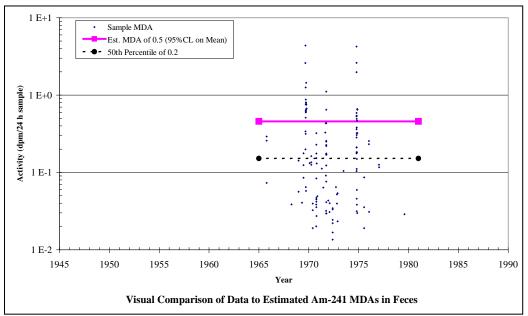


Figure A-15. MDA data for ²⁴¹Am in feces (part 1 of 3).

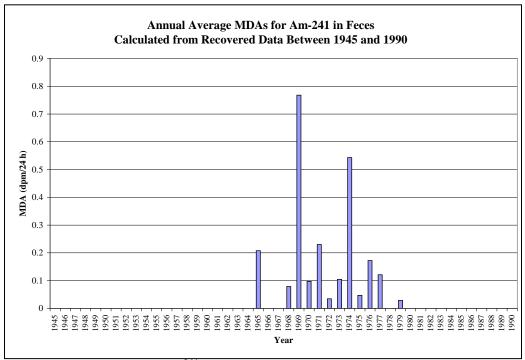


Figure A-15. MDA data for ²⁴¹Am in feces (part 2 of 3).

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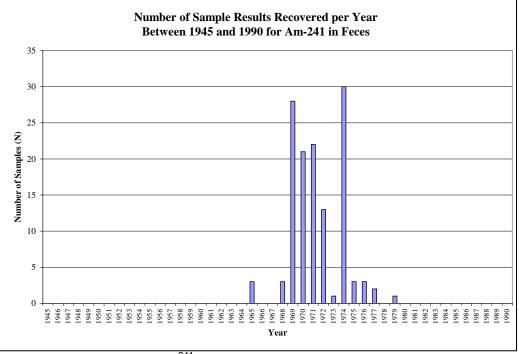


Figure A-15. MDA data for ²⁴¹Am in feces (part 3 of 3).

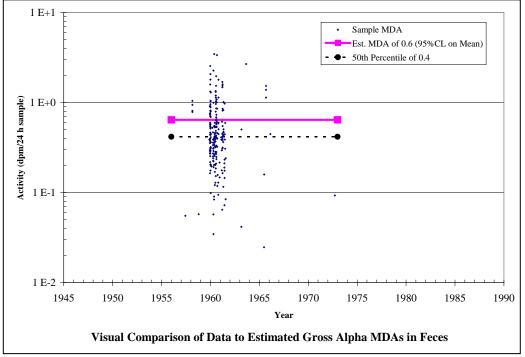


Figure A-16. MDA data for gross alpha in feces (part 1 of 3).

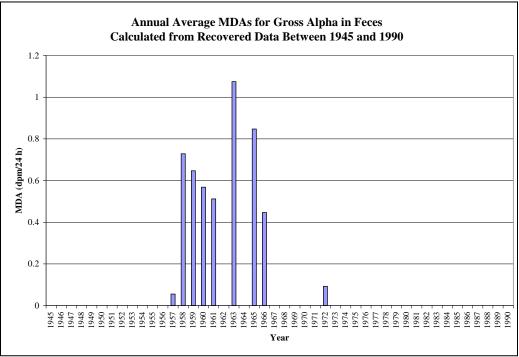


Figure A-16. MDA data for gross alpha in feces (part 2 of 3).

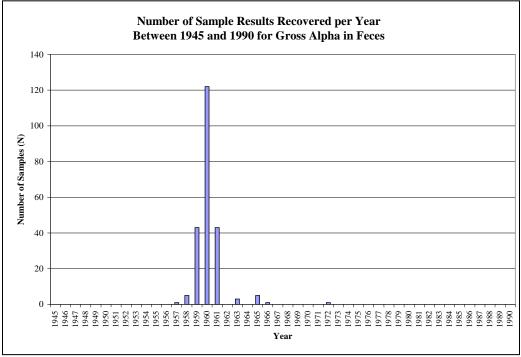
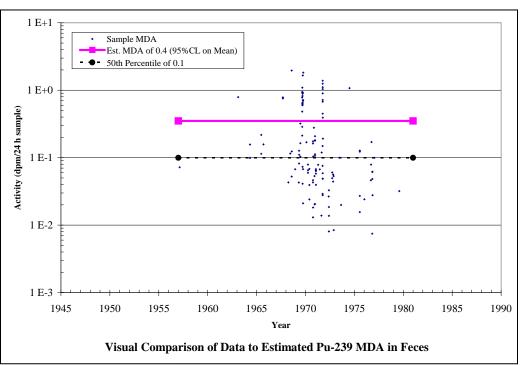


Figure A-16. MDA data for gross alpha in feces (part 3 of 3).



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Figure A-17. MDA data for ²³⁹Pu in feces (part 1 of 3).

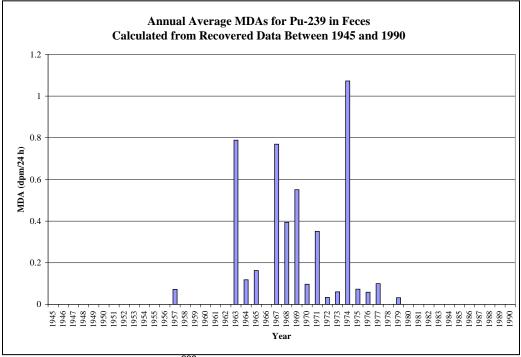


Figure A-17. MDA data for ²³⁹Pu in feces (part 2 of 3).

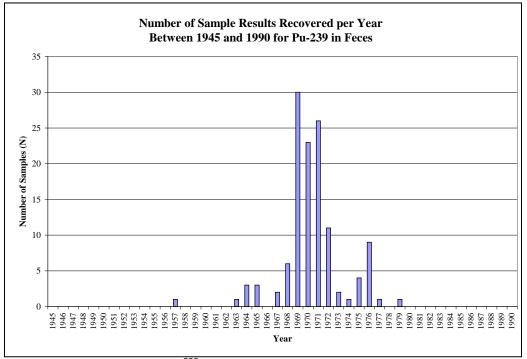


Figure A-17. MDA data for ²³⁹Pu in feces (part 3 of 3).

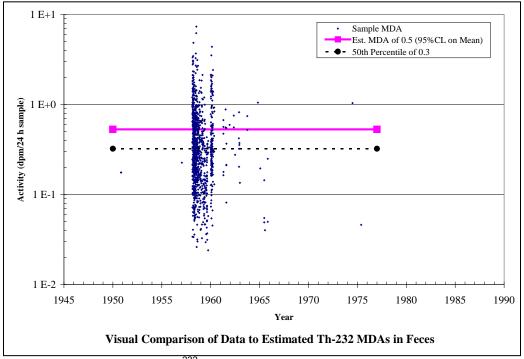
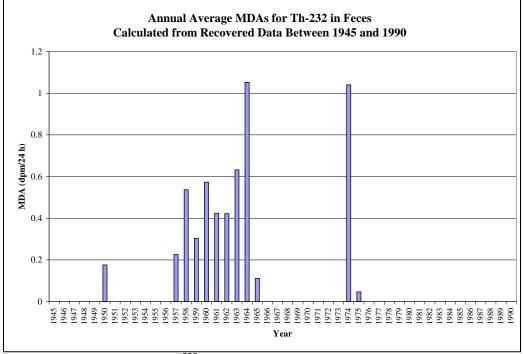
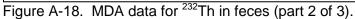


Figure A-18. MDA data for ²³²Th in feces (part 1 of 3).





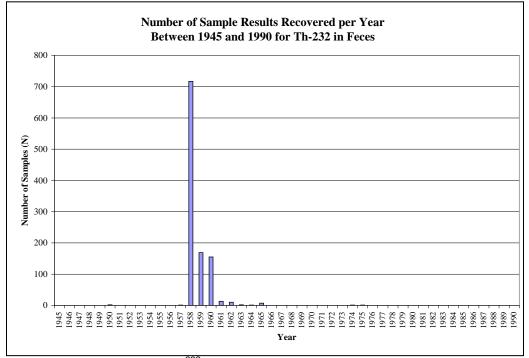


Figure A-18. MDA data for ²³²Th in feces (part 3 of 3).