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Subject Expert(s):	Edward D. Scalsky and Donald E. Bihl

Approval:	Signature on File	Approval Date:	01/19/2012
	Edward D. Scalsky, Document Owner		
Concurrence:	Signature on File	Concurrence Date:	01/19/2012
	John M. Byrne, Objective 1 Manager		
Concurrence:	Signature on File	Concurrence Date:	01/19/2012
	Edward F. Maher, Objective 3 Manager		
Concurrence:	Signature on File	Concurrence Date:	01/19/2012
	Kate Kimpan, Project Director		
Approval:	Signature on File	Approval Date:	01/20/2012
	James W. Neton, Associate Director for Science		

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PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
01/20/2012	00	New document initiated in response to questions raised by SC&A that not all radionuclides have been identified and considered in the analysis of personnel exposures at the Savannah River Site. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Edward D. Scalsky.

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ACRONYMS AND ABBREVIATIONS

AEC   U.S. Atomic Energy Commission
ANL   Argonne National Laboratory

BNL   Brookhaven National Laboratory

d   day
DNA   delayed neutron analysis
DU   depleted uranium

EU   enriched uranium

g   gram
GT   Georgia Tech

HDEHP   di,2ethylhexylophosphoric acid
HLC   High Level Cave
hr   hour

in.   inch

keV   kilo-electron volt
kg   kilogram

LSC   liquid scintillation counter

MCi   megacurie
mg   milligram
mm   millimeter
MPBB   maximum permissible body burden
MPLB   maximum permissible lung burden
mrem   millirem

nCi   nanocurie

ORNL   Oak Ridge National Laboratory

SRL   Savannah River Laboratory
SRP   Savannah River Plant
SRS   Savannah River Site

TIOA   tri-isooctlyamine

UCB   University of California, LRL Berkeley
UCL   University of California, LRL Livermore
UCLRL   University of California Lawrence Radiation Laboratory

wk   week

yr   year

μg   microgram

1.0 EXECUTIVE SUMMARY

The first reactor (R-Reactor) at Savannah River started operation on December 28, 1953 and shut down in June 1964. The K-, P-, and L-Reactors all started in 1954 and operated until 1988, while the L reactor shut down in 1968 and restarted in 1985. The C reactor operated from 1955 through 1985.

During the years the Savannah River Site (SRS) reactors were operating, there were many nuclides that were irradiated. Originally, the irradiations were in support of the nuclear weapons program. However, when military requirements slowed down, other missions were sought. Some missions included the production of transplutonium elements; development of isotopic power and heat power sources; and development of sources for food sterilization, medical uses, radiography, and irradiating samples for national laboratories, universities, and other countries in support of their research.

In light of the many radionuclides that resulted from these irradiations, questions have been raised that not all radionuclides have been identified and considered in the analysis of personnel exposures (SC&A 2010). According to the SC&A report, the main issues relate to the production, irradiation in reactors, and processing (if any) of target materials for the production of special radionuclides to clarify the dose reconstruction issues associated with them. SC&A classifies these special radionuclides as the "exotic" radionuclides that were produced in the C- and K-Reactors during the high flux years of operation (i.e., 1965 to 1970). This report attempts to identify the radionuclides, the bioassay methods that were used to evaluate the potential exposures to personnel handling and processing the radionuclides and, if possible, the approximate quantities that were available at SRS.

2.0 INTRODUCTION

In addition to the materials that were irradiated in the SRS reactors for the weapons program, a great deal of material was irradiated for use in commercial, industrial, and medical applications. This report discusses the nuclides that were irradiated and processed for various periods in the SRS facilities. The bioassay methods that were used to evaluate the potential exposure to these radionuclides are also discussed.

3.0 RADIONUCLIDE PRODUCTION AT SAVANNAH RIVER

The term "exotic radionuclides" was used first in the SC&A report (SC&A 2010). SC&A states their memo concerns "exotic radionuclides" that are not explicitly addressed as matrix issues for which NIOSH is developing coworker models. Radionuclides excluded from analysis include ^{232}Th, ^{241}Am, ^{244}Cm, ^{252}Cf, ^{237}Np, fission and activation products,^{210}Po, recycled uranium, natural uranium, $^{239/240}$Pu, and ^{241}Pu. SC&A does include among the "exotic radionuclides" those materials that were irradiated for off-site customers during the high flux demonstration program. However, for purposes of this report any radionuclide with some exceptions (e.g., tritium, thorium) that were produced as a result of irradiation in the SRS reactors is considered to be an "exotic radionuclide", some of which are only briefly mentioned.

The SC&A report (SC&A 2010) identified the following radionuclides of concern:

The alpha emitters were:

- Americium-243
- Protactinium-231
- Plutonium-238
- Plutonium-242
- Uranium-233

The beta emitters were:

- Cesium-137
- Europium-153
- Iodine-131
- Iridium-192
- Promethium-147
- Technetium-99
- Thulium-170

In the early 1950s, five heavy-water reactors (R, P, L, K, and C) were constructed at SRS. All five reactors were operating by 1955. Reactor shutdown began in 1964 with R-Reactor. C-Reactor was shut down in 1987; L-Reactor was shut down in 1968, restarted in 1985, and shut down again in 1988. K- and P-Reactors operated with few interruptions until 1988. A restart of K-Reactor began in 1991, but was not completed (Till et al. 2001). None of the reactors are currently operational.

Originally, the primary function of the SRS reactors was the production of plutonium and tritium for nuclear weapons. However, even in the early 1950s, the reactors were producing radioisotopes for nonweapons uses, which continued in the 1960s with the reduction of the Savannah River Site's military mission (Reed et al. 2002). Depleted uranium (DU), ^{237}Np, and lithium targets were irradiated to produce ^{239}Pu, ^{238}Pu, and tritium, respectively. The targets were irradiated and transferred to the Tritium Facilities, H-Area Separations (^{238}Pu), and F-Area Separations (^{239}Pu) for processing. Spent reactor fuel was sent to H-Area Separations for processing. The reactors were also used to irradiate targets that produced transplutonic elements such as curium and californium.

Between 1955 and 1967 large quantities of special radionuclides were produced in the SRS reactors for various campaigns. For example, a series of campaigns produced 66 MCi of ^{60}Co for Oak Ridge National Laboratory (ORNL), the U.S. Bureau of Mines, the U.S. Army Food Process Development Irradiator, Japan Atomic Energy Research Institution, Brookhaven National Laboratory (BNL), and Marine Products Development and 630 kg of ^{233}U from irradiation of thorium were produced for the thorium breeder reactor program. In addition, 275 kg of ^{238}Pu from irradiation of ^{237}Np, 600 g of ^{210}Po, 1,700 Ci of ^{152}Eu, and 370 MCi of ^{170}Tm were produced. Other radionuclides produced during this time period and in subsequent years included more ^{210}Po, ^{121}Sn, ^{139}Ce, ^{166}Ho, and ^{41}Ca for BNL, the University of Illinois, and Denmark (Bebbington 1990).

From 1965 through 1970, C- and K- reactors were modified to be able to produce a very high flux of neutrons for production of unusual (e.g., ^{170}Tm, ^{171}Tm, ^{46}Sc) radionuclide sources. The High Flux Demonstration Program began in C-Reactor in February 1965 and lasted 1 year. The High Flux Program in K-Reactor began in August 1969 to produce ^{252}Cf. A peak flux of 7×10^{15} n/cm^2-sec was achieved and 2.1 g of ^{252}Cf were produced before termination of the production campaign in November 1970. A second Californium Production Campaign was planned but never executed because the market for ^{252}Cf never materialized as expected (Boswell 2000). The target material in the C- and K-Reactors consisted primarily of ^{242}Pu. In addition, three 1-in.-diameter thimbles contained ^{243}Am, ^{244}Cm, and 150 target materials of 66 elements for nine universities and laboratories (Boswell 2000). Table 3-1 lists the target materials that were used to supply irradiated samples to Argonne National Laboratory (ANL), ORNL, University of California, LRL, Berkeley, and University of California, LRL, Livermore (Smith et al. 1965). There were additional targets; for example, one tritium-producing core (Mark VI) was designed with a removable inner lithium target so fuel could be irradiated to a very high exposure. The high exposure depletes 70% of the ^{235}U and thus concentrates ^{236}U, which is formed from every sixth neutron capture in ^{235}U. Irradiation of the ^{236}U forms ^{237}Np, which is the target material for the production of plutonium (Boswell 2000). In addition, the irradiation of the target material ^{242}Pu, after a succession of neutron captures and beta decays, successively produces other target materials – ^{242}Pu, ^{243}Pu, ^{243}Am, ^{244}Am, ^{244}Cm, ^{245}Cm, ^{246}Cm,

^{247}Cm, ^{248}Cm, ^{249}Cm, ^{249}Bk, ^{250}Bk, ^{250}Cf, ^{251}Cf, and ^{252}Cf (Reed et al. 2002). There were also special irradiations during both the high-flux charge and the Curium I campaign to produce test quantities of ^{170}Tm, ^{171}Tm, ^{46}Sc, and ^{204}Tl.

Table 3-1. Examples of general samples irradiated during the high flux demonstration.[a,b]

Sample	Lab	Sample	Lab	Sample	Lab
Si-30	A	Yb-168	OR	Os	UCL
S-33	OR	Tm-168	UCB, UCL	W	UCL
Ca-40	OR, A	Er-170	UCB	Re	UCL
Ca-48	A	Yb-176	A, UCB, UCL	Be	A
Fe-54	OR	Lu-176	UCB	Be$_3$N$_2$	OR
Fe-58	OR, A, UCB	Hf-177	UCB	Xe	UCL
Ni-58	OR, A	Hf-179	A, UCL, UCB	TiO$_2$	A
Ni-62	OR	Hf-180	A, UCL, UCB		
Nb-93	OR	Ta-180	UCB		
Nb-96	A	Ta-181	UCB		
Ag-107	UCB	W-186	A, UCL, UCB	**Additional isotopes[c]**	
Ag-108	UCB	Re-187	UCL, UCB	Zn-67	A
Sn-118	OR, UCB, A	Os-180	UCB	Te-124	A
Sn-124	A	Os-182	A, UCL, UCB	Lu-175	UCL
Ba-130	UCB	Bi-209	UCB	Be-9	A
Sm-144	OR, UCB	Am-241	A, UCL		
Nd-146	OR	Cm-244	A, UCL, UCB	Sn-120	A
Nd-150	A	Bk-249	A		
Sm-150	UCB, UCL	Cf-252	A, UCL, UCB	Cd-160	UCL
Sm-154	OR	Es-254	UCB	Am-243	UCL
Dy-156	UCB, UCL	LiCl	UCB		
Dy-158	A	MgO+Cr-50	UCL		
Tb-158	UCB, UCL	KPO$_3$	UCL		
Gd-160	A, UCL, UCB	Ar	UCL		
Er-162	UCB, UCL	RbCl	OR		
Dy-164	A, UCB	Hf	A		
Ho-165	UCB, UCL	HFO$_2$	UCL		

a. A=Argonne National Laboratory; OR=Oak Ridge National Laboratory; UCB=University of California, LRL, Berkeley; UCL=University of California, LRL, Livermore.
b. Source: Smith et al. 1965.
c. Other isotopes from Table 3-2.

Some development work was done mostly with cold Tm$_2$O$_3$ (DuPont 1974, p. 12) but a little work was done with ^{170}Tm, ^{171}Tm, ^{46}Sc, and ^{204}Tl (DuPont 1974, pp. 7–8). Scandium-46 is a gamma-emitter. Thallium-204 is a beta-only emitter. Thulium-170 and -171 have low-yield, low-energy gammas that might show up in chest counting if someone was looking closely, but mostly are beta emitters. Thulium oxide was characterized as target material for irradiation to form ^{170}Tm, with potential applications for short-term missions in space (128-day half-life). As with cobalt, the irradiated product is useful without chemical processing. Conditions were established for fabricating wafers, and the properties of Tm$_2$O$_3$ were studied to prepare and irradiate it to yield a radioactive product suitable for use in heat sources. There was some activity that involved breaching of the source or doing chemistry on the ^{170}Tm and ^{171}Tm. These were small research and development experiments before actual large-scale production of the sources. There was some breakage of ^{170}Tm oxide wafers in the early experiments. Metal targets did not have that problem. There was no evidence of ongoing, large-scale breaching or chemistry on the ^{170}Tm or ^{171}Tm sources. Thulium-170 was mentioned as a possible heat source early in the program; it emits a gamma ray with about 3% yield. Thulium-171 only emits a beta in sufficient abundance for bioassay.

SC&A mentioned [147]Pm and ^{192}Ir as two radionuclides of concern (SC&A 2010). The National Aeronautics and Space Administration's interest in aged [147]Pm as a source of Isotopic heat for space missions led to a study of the quantity and quality of [147]Pm that could be produced annually at the SRP. The recovery of [147]Pm from current high-activity waste was also studied. The major source of [147]Pm would be fission product waste; the quantity available after aging to allow for decay of associated [148]Pm would be about 1,700 W(t) per reactor-year of operation. Product quality from current waste, as represented by a low ratio of [148]Pm to [147]Pm, would presumably be the same as that produced at the Hanford Site (about 5×10^{-7}). Promethium-148 emits gamma rays that could require extra shielding. Irradiation of naturally occurring neodymium would produce only 50% as much [147]Pm (DuPont 1974). No further mention of [147]Pm has been found in this historical reference. DuPont (1974) mentions that iridium was experimented with but indicates that this experimentation was with several inactive iridium capsules. The report makes no mention of [192]Ir.

Table 3-1 shows a list of materials that were irradiated for the AEC laboratories only, i.e., Argonne National Laboratory (ANL), Oak Ridge National Laboratory (ORNL), University of California, Berkeley (UCB), and the University of California, Livermore (UCL). Some of the materials were irradiated for the duration of the high-flux campaign (i.e., from February to November 1965). Others were irradiated for single fuel cycles of about 1 week (Smith et al. 1965). Table 3-1 does not include isotopes that were irradiated for BNL, the University of Illinois, or Denmark, such as [121]Sn, [138]Ce, and [139]Ce, or those irradiated for all off-site customers.

Table 3-2 lists some of the specific target elements and compounds that were irradiated for outside customers during the months and years indicated. Many of the samples are repeated in subsequent months; for example, March and May 1966 and March 1967 have many repeat samples. There were five shipments during January 1966. The [257]Fm to LLNL and the 2 µg of [252]Cf to ANL are listed in the table. The other shipments, not listed, included 380 mg of iridium to ORNL, 200 mg of calcium to Berkeley, and 1 µg of [253]Es to ANL.

The procedure for handling these samples is described by Gray (2006) as follows:

> *All samples containing the material to be irradiated for off-site customers were in welded closed aluminum cans. They came to SRP in that condition. They were placed in a basket and the basket lowered into the spent fuel storage basin where the deep water provided shielding for handling radioactive components removed from the reactor. The research samples were placed in small perforated tubes about 1 inch in diameter; maybe five or so samples per a 12" tube. The tubes were then placed in a larger (about 1 3/8ths inch diameter) perforated tube about 21 feet long, called a thimble. The reactor had many of these thimbles for general use with safety rods, instruments, etc. Three of them near the center of the reactor were dedicated to my [Gray] use for research samples. When it came time for either insertion or removal (or both) of a sample, the thimble was removed by remote machine from the reactor and placed in the basin. It was moved remotely under water to a location where I could have it turned upside down and the tubes unloaded onto a well-lit work table about 12 feet down. I sorted through the tubes and found the one from which a sample would be unloaded (or a new one loaded); all under water. The process was reversed to put the whole thing back together and return the loaded thimble to the reactor for the next cycle. Of the 15-18 hours for shut down, I had about 3-5 hours in the middle of it to do my work.*

> *A removed research sample, without any opening of the welded-closed aluminum cans, was placed directly in a shielded shipping cask. The cask lid was put in place and sealed (or locked) down for transport. As the cask was removed from the basin it*

Table 3-2. Samples irradiated during the high flux program for selected months.[a]

August 1965[b]		January 1966[c]		March 1966[d]		May 1966[e]		March 1967[f]	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
ORNL	280 mg natural iridium	**SRP-SRL**	1.4 g Co-59	UCLRL	11 mg Ag-107	UCLRL	11 mg Ag-107	UCLRL	11 mg Ag-107
SRP-SRL	1.4 g Co-59	ANL	40 mg Cm-244	UCLRL	1.52 g LiCl	UCLRL	1.52 g LiCl	UCLRL	1.52 g LiCl
ANL	40 mg Cm-244	ANL	2 µg Cf-252	UCLRL	217 mg MgO and Cr-50	UCLRL	217 mg MgO and Cr-50	UCLRL	217 mg MgO and Cr-50
ANL	2 µg Cf-252	UCLRL	240 mg Cm-244, 20 mg Cm-246	UCLRL	10 mg Fe-58	UCLRL	10 mg Fe-58	UCLRL	10 mg Fe-58
UCLRL	400 mg Cm-244	ANL	77.2 mg Be-9, 0.6 mg Ca-48, 2 mg S-30	UCLRL	2 mg Dy-156	UCLRL	2 mg Dy-156	UCLRL	2 mg Dy-156
UCLRL	450 mg Cm-244; 40 mg Cm-246	ANL	3.95 mg Fe-58, 1.4 mg Sn-124, 5.26 mg Hf-180	UCLRL	9 mg Hf-179	UCLRL	9 mg Hf-179	UCLRL	9 mg Hf-179
UCLRL	60 mg Cm-244; 25 mg Cm-246	UCLRL	1.9 mg Cf-252	UCLRL	10.3 mg Hf-180	UCLRL	10.3 mg Hf-180	UCLRL	10.3 mg Hf-180
ANL	240 mg Cm-244; 20 mg Cm-246	UCLRL	231 mg Gd-160	UCLRL	58 mg Sm-150	UCLRL	58 mg Sm-150	UCLRL	58 mg Sm-150
ANL	77.2 mg Be-9; 0.6 mg Ca-48; 2 mg Si-30	UCLRL	48 g Bi-209	GT	200 mg 27% Pb-204 metal, 1 mg 99.7% ^{204}PbO	GT	200 mg 27% Pb-204 metal, 1 mg 99.7% ^{204}PbO	GT	200 mg 27% Pb-204 metal, 1 mg 99.7% ^{204}PbO
UCLRL	3.95 mg Fe-58; 1.4 mg Sn124; 5.26 mg Hf-180	UCLRL	48 gm Bi-209	BNL	0.5 g C (diamond in graphite)	UCLRL	1.5 mg Pu-240, 0.1 mg Pu-244	UCLRL	1.5 mg Pu-240, 0.1 mg Pu-244
ANL	1.9 mg Cf-252	UCLRL	99 mg Yb-176	ANL	40 mg Cm-244	UCLRL	48 g Bi-209 (2.5 in.)	UCLRL	48 g Bi-209
UCLRL	0.8 g Hf-180	UI	8.3 mg Ba(NO$_3$)$_2$, 12.5 mg CeO$_2$, 600 mg Sn-118	UCLRL	1.5 mg Pu-240, 0.1 mg Pu-244	UCLRL	48 g Bi-209 (2.54 in.)	UCLRL	48 g Bi-209
UCLRL	231 mg Cd-160	UCLRL	1 g Am-243[c]	UCLRL	48 g Bi-209	UCLRL	10.8 mg Hf-177	UCLRL	10.8 mg Hf-177
UCLRL	48 g Bi-209	UCLRL	1 g Am-243	UCLRL ANL	240 mg Cm-244; 20 mg Cm-245	UCLRL	10 mg Os-192	UCLRL	10 mg Os-192
UCLRL	48 g Bi-209	UCLRL	5.7 g Lu-175	UCLRL	48 g Bi-209	UCLRL	10.9 mg Er-162	UCLRL	10.9 mg Er-162

August 1965[b]		January 1966[c]		March 1966[d]		May 1966[e]		March 1967[f]	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
UCLRL	99 mg Yb-176	ORNL	216 mg Sm_2O_3 (enriched to 94.5% Sm-144) and 7.828 mg type 302 SS monitor	UCLRL	10.8 mg Hf-177	UCLRL	9.6 mg W-186	UCLRL	9.6 mg W-186
SRP	13.47 mg Al	ORNL	700 mg gold foil and 7.61 mg type 302 SS monitor	UCLRL	10 mg Os-192	UCLRL	9.74 mg Gd-160	UCLRL	9.74 mg Gd-160
UI	8.3 mg $Ba(NO_3)_2$; 12.5 mg CeO_2; 600 mg Sn-118	ORNL	418 mg La_2O_3 and 7.718 mg type 302 SS monitor	UCLRL	10.9 mg Er-162	UCLRL	9.74 mg Re-187	UCLRL	9.74 mg Re-187
ORNL	280 mg natural iridium	ANL	1 gm Sn-120	UCLRL	9.6 mg W-186	UCLRL	7.65 mg Lu-176	UCLRL	24 mg Sm-144
UCLRL	1 g Am-243	UCLRL	40 mg Ba-130 as $BaCO_3$	UCLRL	9.74 mg Gd-160	UCLRL	24 mg Sm-144	UCLRL	50 mg Sn-118
UCLRL	1 g Am-243	UCLRL	40 mg Ba-130 as $BaCO_3$	UCLRL	9.74 mg Re-187	UCLRL	9.6 mg Ag-109	GT	77 g lead
UCLRL	1 g Am-243	UCLRL	40 mg Ba-130 as $BaCO_3$	UCLRL	24 mg Sm-144	UCLRL	50 mg Sn-118	**SRL**	1 g Er_2O_3 enriched in Er-170
UCLRL	1 mg Hg-203 (as nitrate)	UCLRL	1.5 mg Pu-240, 0.1 mg Pu-244	UCLRL	9.6 mg Ag-109	ORNL	100 mg Te-128 (99.16% enriched)	ANL	60 mg Te-126 enriched (98.69%)
SRL	90 mg $^{146}Nd_2O_3$ (enriched)	BNL	0.5 g C (diamond in graphite)	UCLRL	50 mg Sn-118	GT	77 g lead	Livermore	460 mg Cm-244, 90 mg Cm-246, 9 mg Cm-248
		BNL	0.5 g C (diamond in graphite)	ORNL	100 mg Te-128 (99.16% enriched)			NC	500 mg SnO_2 enriched 99% Sn-120
		BNL	0.5 g C (diamond in graphite)	ORNL	100 mg ^{128}Te (99.16% enriched)			Livermore	300 µg Cf-252
		ANL	^{40}Ca, ^{42}Ca, ^{58}Ni, ^{62}Ni,	GT	77 g lead			ANL	Er-162, Yb-176, Mo-92

August 1965[b]		January 1966[c]		March 1966[d]		May 1966[e]		March 1967[f]	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
		ANL	Zn-67, Te-124, Pb-204[d, g]					ANL	Fe-54, Yb-168, Cr-50, Os-192
		ANL	Se, Yb-176 [g]					ANL	Nd-150, Ti-50, W-186, Yb-176
		ANL	Nd-150, Os-190[g]					ANL	Nb, Ce-136, Cf-249, N-236p
		ANL	U-236, Pa, Ni [g]					ANL	90 mg Cm-244, 30 mg Cm-246
		ANL	Np-236, Cf-249, Th-229[g]					UCLRL	5 mg Ln-176 (70% enriched)
		UCLRL	200 mg calcium					ORNL	103.2 mg $^{186}WO_3$ (97.2% enriched)
		UCLRL	0.7 mg platinum metal						
		UCLRL	1 mg Hg-202 in quartz capsule						
		UCLRL	Fm-257 rabbit sample in Al capsule (10 µg)						
		ORNL	8.5 mg type 302 SS monitor						
		ORNL	150 µg $^{236}U_2O_3$, 3 - 8.5 mg type 302 SS monitor						

a. ANL-Argonne National Laboratory, ORNL-Oak Ridge National Laboratory, UCLRL-University of California Lawrence Radiation Laboratory, BNL-Brookhaven National Laboratory, GT-Georgia Tech, UI-University of Illinois, SRL-Savannah River Laboratory, SRP-Savannah River Plant.
b. DuPont (1965a).
c. DuPont (1966a).
d. DuPont (1966c).
e. DuPont (1965b).
f. DuPont (1967a).
g. The sample mass and chemical form of one or more nuclides were unreadable in the referenced document.

was washed down. Then it was placed on a truck for transport, usually to the Atlanta airport, for a flight to the outfit that sent it to us originally for irradiation. Many samples, because of short half lives of the desired product, were returned in well less than 24 hours. No samples were opened at SRP [Savannah River Plant].

As discussed by Gray (2006), all samples that were to be irradiated for external customers were in welded closed aluminum cans. They came to SRS in that condition. After irradiation they were prepared under water for shipment, so the potential for intake was minimal. Similarly, because the sources were inspected and loaded into casks under water, the potential for external exposure was reduced. As noted above, Table 3-2 lists specific samples that were irradiated for the sponsors listed in the odd numbered columns for the month and year indicated. As can be seen in the table, the samples are generally small for research purposes at the sponsor's facilities. However, there are five samples in the 5-month period listed in the table that were transferred to SRS-SRL. These samples would have been handled using SRS procedures. Based on the limited number of radionuclides transferred, it would appear that such irradiations were not routine.

In addition to the radionuclides and compounds listed in Tables 3-1 and 3-2, a request was made of the Isotopes Development Center at SRS to look at the possibility of using Site irradiation facilities as fully as possible for the production of unusual or quite large amounts of isotopes for the potential benefit to research and commerce. In response to that request, Rupp (1969) discussed some of the characteristics of the radionuclides in Table 3-3, the target materials or compounds that would be used to produce some of them, and difficulties that might be encountered. He noted that some of these radionuclides were already being produced. As noted above in this section, these radionuclides have been identified by SC&A (2010) as being of concern for dose reconstruction. The entire list discussed in Rupp (1969) is included in Table 3-3. No documentation has been found indicating that program was ever pursued.

Table 3-3. Radionuclides discussed for possible irradiation (Rupp 1969).

Plutonium-238	Thulium-170 and 171	Calcium-41	Barium-133	Samarium-154
Americium-242m	Potassium-40	Iron-55	Lanthanum-137	Samarium-155
Cesium-134	Argon-38 (stable)	Nickel-59	Terbium-157	Europium 155
Europium-152	Chlorine-36	Cadmium-109	Holmium-166m	
Europium-154	Argon-39	Cadmium-113m	Samarium-151	

In addition to the information above, a review of monthly reports was conducted to identify the radionuclides that were being irradiated or processed at the SRS from 1954 through 1971. Table 3-4 lists the results of this review (Isotope Charts 2011). Some of these isotopes were part of the production process and many were shipped to off-site customers.

The cells containing an x indicate that the isotope was present several times during the year. Some isotopes were present in every month of the year, others only during several months. Those identified by an L or M indicate they were present on a limited basis during 1 or 2 months during the year. The letters represent the potential for exposure to that radionuclide [e.g., either low (L) or medium (M) during that period]. This represents the highest estimated exposure potential for those radionuclides. Because the radionuclides indicated by an x appeared many times during the year, an indication of their exposure potential could not be identified in the table. Most were in the low to medium range but others, such as ^{241}Am, ^{252}Cf, ^{244}Cm, and ^{237}Np, were often listed as a high exposure potential. The # symbol indicates the isotopes were identified either through existing SRS calculations or in plans. There are 4 radionuclides added to the table that were not identified in the monthly reports but were identified in History of the Savannah River Laboratory, Volume IV – General (DuPont 1984a) and were observed in the bioassay results. These are ^{106}Ru, ^{233}Pa, ^{95}Zr, and Zr/Nb-95 and are included for completeness.

Table 3-4. Isotopes identified in SRS documents from 1954 through 1971.

Isotope	1954	1955	1956	1957	1858	1959	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971
Am-241					x					x		x				x	x	x
Am-242m[a]															#[b]	M[c]		
Am-243						x	x	x		x	x	x	x	x	x	x		
Al[a]											L[d]							
Ba-133[a]										L								
Bi-209[a]												L						
Bk-249											x	x	x	x	x	x		
Bk-250[a]																L	M	
Ce-144[a]						x				x	x			x	x			
Cf-252						x					x	x	x	x	x	x	x	x
Cm-244						x	x	x	x		x	x	x	x		x	x	x
Cm-248[f]														f				
Co-59												x	x					
C0-60		x								x	x				x	x	x	x
Cs-137[a]										x	x	x						
Er-170											x	x		x				
Es-253														x	x	x	x	
Es-254												x	x	x	x			
Es-255													L					
Eu-152[a]														L	L			
Eu-154[a]											L							L
Fm-254[a]																	M	
Flux wire														L				
Ir-192[a]												L						
Nd-148[a]											L							
Np-237[a]		x	x			x	x	x	x	x	x	x	x	x	x	x	x	x
Np-238[a]																		M
Np-239										L								
Pa-231[a]													M					
Pa-233[e]										e								
Pm-147[a]						x	x					x	x	x	x	x		
Po-210[a]								x			x	x	x	x	x	x		
Pu-238[a]								x			x	x	x	x	x	x	x	
Pu-239[a]												x						
Pu-240[a]		x					x	x	x			x	x	#	#			
Pu-241[a]												x						
Pu-242[a]										x		x	x	x	x	x	x	x
Ru-106[a,e]														e				

Isotope	1954	1955	1956	1957	1858	1959	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971
Sb-124[a]																		L
Sb-125[a]																		L
Sc-46[a]										L		L						
Si (quartz)[a]													L					
Ta[a]												L						
Tc-99[a]												L						
Tl-204[a]												L	L					
Tm-170[a]										x		x	x	x	x			
Tm-171[a]											x	x	x	x	x			
U-233[a]		x							x	x		x	x	x	x			x
Y-91[a]						L												
Zr-95[e]													e					
Zr-95-Nb-95[e]														e				
Thorium[a]	x	x			x	x	x	x	x	x	x	x	x	x	x			

a. These isotopes are not included in Table 3-2.
b. # Identified in calculations or plans.
c. M Represents an estimated moderate potential of exposure.
d. L Represents an estimated low potential of exposure.
e. Not identified in the review of the monthly reports but was identified by the SRL bioassay program (DuPont 1984a).
f. During separation and purification of transcurium nuclides, 1.7 µg ^{248}Cm was obtained from a filter from which 14 µg ^{252}Cf had previously been obtained.

4.0   <u>SAVANNAH RIVER LABORATORY OPERATIONS</u>

The Savannah River Laboratory (SRL) (Building 773-A) and related organizations began organizing in July 1953. Over the next couple of years the Engineering' Services that were provided in support of research and development personnel included operation of all building auxiliaries and waste disposal facilities, a glass shop, instrument pool, and a supply room. Close liaison was established with Works Engineering Department personnel to coordinate all maintenance, machine shop, and project work required for the Savannah River Laboratory. As radioisotopes were brought into 773-A to start operations in radiochemical laboratories, the Radiation Control group assisted in establishing necessary procedures and handling techniques.

Operations formally accepted occupancy of the cave wing, 773-A, Part II, on December 7, 1953 and all construction work was completed in February 1954. Experiments with radioactive materials began in the caves in July 1954. In October 1954 the first plutonium produced at the Savannah River Plant was isolated in the caves.

A review of the operations in the SRL was conducted because that was where the products of reactor operations were evaluated. While the SRL did not evaluate the radionuclides that were shipped to off-site customers, they did handle, evaluate, experiment with, and store "production" radionuclides such as ^{244}Cm, ^{252}Cf, ^{60}Co, etc. Reactor incidents are not covered in this report because they would primarily involve reactor functionality, such as fuel failures, valve failures, etc., rather than exposure to the handlers of the transfers of highly radioactive radioisotopes, which was, of necessity, performed under water due to the high exposure potential. There were a considerable number of potentially serious incidents in the SRL, as discussed below. Several of the incidents have been investigated and have an associated Special Hazard Investigation report (Dosimetry Special Hazard Incident Investigations #'s 200-299), while the nature of many of the incidents has not been identified. However, there was an extensive radiation control program in terms of identifying potential problems and steps taken to evaluate any internal or external exposure as a result of these incidents.

Radiation Control was established in Technical Division facilities so all the survey, monitoring, and "containment' services usually assigned to Health Physics could be provided in the Technical Division organization, together with a wholly integrated approach and direct responsibility (DuPont1984a). The containment service function required close liaison work with technical personnel in the design and construction of the special equipment required to contain the radioactive materials completely during experimental work. This function was performed by the Apparatus Development Group, a subgroup of Radiation Control (DuPont 1984a). As radioisotopes were brought into Building 773-A to start operations in radiochemical laboratories, the Radiation Control Group assisted in establishing necessary procedures and handling techniques. Major effort was directed to extensive training of laboratory personnel and personnel of assigned groups. The fundamentals of radiation and contamination control and aspects of personal protection were stressed. By December 1955, procedures basic to the program of personnel and area radiation protection were essentially complete (DuPont 1984a).

Much of the work in the SRL was carried out in the High-Level Caves, which were designed to perform chemical and metallurgical operations on highly radioactive materials. As indicated above, operations formally accepted occupancy of the cave wing, 773-A, Part II, on December 7, 1953. In October 1954 the first plutonium produced at the Savannah River Plant was isolated in the caves to provide basic data to substantiate and firm up calculations of reactor performance. Operations in the caves continued at the maximum obtainable rate from that date.

The following information presents a synopsis of the internal exposure history of the SRL from 1953 through December 1972, which encompasses the high-flux operational period. The information for

these years is taken from the *History of the Savannah River Laboratory, Volume IV – General* (DuPont 1984a).

In the period from July 1953 to the early part of 1960 evaluation of internal doses was based on *in-vitro* analysis. In 1960 the SRS whole body counter was completed and provided another means of assessing internal doses. From 1953 through 1972 there were thousands of urine samples and whole body counts but relatively few documented uptakes in the SRL that resulted in significant doses. There were no significant biological uptakes from 1953 through 1959 and none identified in 1963 and 1968. Table 4-1 shows the number of positive bioassay results that have been documented in DuPont (1984a). Not all positive bioassay results were confirmed to be significant uptakes.

Two Special Hazards Investigation (SHI) reports have been identified during this period. Special Hazards Investigation No. 273 reported a general service operator received a positive uptake of ^{244}Cm when he released airborne activity to the decontamination room environment while removing solid radioactive waste from the Decontamination Chamber in Building 773-A, Decontamination Facility, Room C-003 (DuPont 1970a). Special Hazard Investigation No. 283 reported an Analytical Chemistry Division Chemist assimilated some curium while removing samples from storage in Building 773-A, Room B-15 (DuPont 1971a).

Table 4-1. Positive bioassay results.

Year	Number of positive results	Radionuclide	% MPBB
1960	5	Not identified	< 1
1961	3	Not identified	< 1
1962	51	Not identified	< 1
1964	1	Pa-233	< 1.5
	1	Ce-144	< 1.5
		Pu	< 1.5
	1	Not identified	Not identified
1965	1	Am-Cm	30[a]
	1	Not identified	< 5
1966	1	Zr-95	3.2
		Ru-106	1.2
	4	Pa-231	< 1
1967	1	Ru-106	4.2
		Ce-144	1.4
		Zr-Nb-95	0.36
1969	11	Cm-244	Not identified
1970	31	Cm-244	Not identified
	1	Cm-244	SHI No. 273[b]
1971	21	Not identified	Not identified
	1 of the 21	Cm-244	SHI No. 283[c]
1972	4	Not identified	Not identified
	2	Cf-252	Not identified

a. No Hazard Investigation Report was found for this uptake.
b. DuPont (1970a).
c. DuPont (1971a).

5.0   BIOASSAY PROGRAMS AT SRS

Workers at SRS were monitored for internal deposition of radionuclides from the beginning of operations. The methods used for evaluation of potential intakes included analysis of the amount of radioactivity excreted in the urine or feces (*in vitro* bioassay) and the measurement of the photon radiation emitted from the body by chest counting or whole-body counting (*in vivo* bioassay).

In the beginning of operations at SRS, the only method available to evaluate the potential internal exposure of personnel was *in vitro* bioassay, primarily analysis of urine samples. This method was typically a gross beta activity analysis although a gross gamma analysis was included from at least 1959 (Boni undated). Over the years, the bioassay analyses methods changed as new techniques were developed.

In Vitro Bioassay
In vitro bioassay methods changed considerably over time as new techniques and analyses were developed and refined. Some of the methods used for analysis of some of the radionuclides are briefly discussed below.

Before completion of a whole-body counting facility in 1960, only *in vitro* bioassay, primarily urinalysis, was performed. *In vitro* urine bioassay for nuclear byproducts performed during this period was typically a gross beta activity analysis and/or gross gamma analysis. Since that time, *in vitro* bioassay analysis has become more radionuclide-specific, and can be divided into the following categories:

- Plutonium urinalysis
- Uranium urinalysis
- Trivalent actinide urinalysis
- Neptunium urinalysis
- Fission product urinalysis
- Fecal bioassay

Plutonium Urinalysis
The first procedure for plutonium urinalysis at SRS was implemented in 1954 and used until 1959. Changes in the processes to prepare the solution for counting were made but the method for counting the alphas tracks on the nuclear track emulsion (NTA) remained the same until 1966. In 1964 solid-state surface barrier detectors (gross alpha counting) replaced the NTA detectors. In the early 1980s experimentation began with a new technique that used alpha spectroscopy, which allowed the detection of the individual radionuclides in the sample. Alpha spectroscopy was used until 1988 (Taylor 2000).

Uranium Urinalysis
Analysis of uranium by measurement of emitted alpha radiation is referred to as an analysis for "enriched uranium." The analysis of uranium based on its chemical properties is referred to as a "depleted uranium" analysis. Enriched uranium (EU) was determined starting in the mid-1950s. Analyses for DU were performed with the ORNL fluorophotometric method from 1954 to 1982. This method involved using a spectroscope to measure total uranium. The delayed neutron analysis (DNA) method was adopted for both EU and DU analyses around 1982 (Taylor 2000).

Trivalent Actinide Urinalysis
The trivalent actinides consist of americium, curium, and californium. There was increased production of these materials at SRS in the early 1970s. Analysis was accomplished by extracting the trivalent actinides from the plutonium-tri-isooctlyamine (TIOA) raffinate with di,2ethylhexylophosphoric acid (HDEHP), gross alpha counting. According to a radiochemistry expert who reviewed the method, this procedure would also have captured elements higher than californium if present, specifically berkelium, fermium, and einsteinium. In 1990, ion exchange resin was used to separate trivalent actinides from other actinides. The sample was direct-mounted (placed on a planchet with rimmed edges and heated to evaporation), and was "gross-alpha" counted by using a wide window on the alpha spectrometer. Since 1994, extraction chromatography resin has been used to separate trivalent actinides from other actinides. With the implementation of improved software and use of an [243]Am tracer, chemical recovery and counting efficiency can be determined for each sample (Taylor 2000).

Neptunium Urinalysis

Starting in 1959, neptunium was coprecipitated, ion exchanged, electrodeposited, and counted on Kodak film emulsion. In the mid-1960s (Butler 1968), the TIOA/gross alpha method was adopted. This method was replaced in 1993 by anion (negatively changed ions) exchange followed by direct mounting and gross-alpha counting.

Since 1994, extraction chromatography resin has been used to separate neptunium from fission products and other actinides, and electrodeposition has been used to mount the sample and gross alpha counted (Taylor 2000).

Fission Product Urinalysis

Fission products measurements in urine date to at least January 1957, although the method used at that time has not been discovered. Since 1959, select fission products in urine were analyzed by beta counting on a Geiger-Müller counter or proportional counter. The beta count was primarily used to determine the presence of ^{89}Sr, ^{90}Sr, and ^{90}Y, which emit no gamma radiation. Because high-energy betas are associated with ^{89}Sr, a reliable measurement of its activity was made even though an average of one gram of precipitate was mounted on the planchet. Yttrium-90, which emits a strong beta, also yields a reliable count, and affords a satisfactory estimate of its parent isobar, ^{90}Sr, since some ^{90}Sr betas are not counted due to absorption by the solid. The low energy beta of most of the induced activities along with the beta of zirconium-niobium and the rare earths will also contribute to the total gross beta count. No interference from ^{40}K was observed. The chemistry on the beta sample resulted in quantitative recovery of beta-emitting isotopes of iron, cobalt, strontium, niobium, zirconium, ruthenium, and cerium. Gross gamma counting was performed using sodium iodide detectors. The gamma count was primarily used to determine the presence of induced activity. A gamma count was used because of the low counting efficiency of the weak beta energies associated with the induced activities. The gamma from zirconium-niobium and rare earths also contributed to the total gross gamma count. Either the beta or the gamma counting statistics are good enough to accurately estimate 1% of the MPA (maximum permissible amount in the total body) of any of the following radionuclides (i.e., ^{90}Sr/Y, ^{89}Sr, ^{95}Zr/Nb, ^{95}Nb, ^{144}Ce/Pr, ^{60}Co, ^{59}Fe, ^{51}Cr, ^{65}Zn) with the exception of zirconium-niobium. The mixed beta-gamma isotope method provided a simple evaluation of the sample regarding the need for an extensive specific analysis, thus eliminating unnecessary and time consuming work (Boni undated).

SRS logbooks show that radionuclide-specific concentrations and associated uncertainties were quantified for some radionuclides for some of the gamma-counted samples starting in December 1965 (DuPont 1965–1968). The reported radionuclides were:

- Cr-51
- Mn-54
- Co-60
- Zn-65
- Zr/Nb-95
- Ru-106
- I-131
- Cs-137
- Ba/La-140
- Ce-144.

Strontium Urine Bioassay

In the late 1950s and early 1960s, $^{89/90}$Sr was determined by beta counting on automatic and Geiger-Mueller counters along with zirconium, rare earths, and niobium. From 1969 to 1997, strontium was analyzed by a liquid ion-exchange method that extracted ^{90}Y, which was counted on a beta

proportional counter. In 1998, the strontium method was modified by using an ion exchange column to extract ^{90}Sr, which is also counted on a proportional counter. Table 5-1 lists the isotopes that were listed as being present, their radiations, bioassay method for detection, half-life, and the number of times they were identified in the monthly reports during the 1954–1971 period. An X indicates they were listed numerous times.

Table 5-1. Selected isotopes bioassay methods.

Radionuclide	Times identified	Half-life[a]	Principal radiation	Bioassay method used or available for use	Bioassay period of availability
Am-243	X	7.4E3 yr	Alpha, gamma	Trivalent actinide urinalysis	From mid 1960s
				Whole-body counting of Np-239	From 1960
				Chest counting	From 1966[b]
Bk-249	X	320 d	Beta	None discovered[c]	
Co-60	X	5.26 yr	Beta, gamma	Whole-body counting	From 1960
				Gamma urinalysis IA	From 1960
Cs-137	X	30.2 yr	Beta, gamma	Whole-body counting	From 1960
Es-253	X	20.5 d	Alpha	Trivalent actinide urinalysis[d]	From mid 1960s
Es-254	X	276 d	Alpha	Trivalent actinide urinalysis[d]	From mid 1960s
				Whole-body counting of Bk-250 progeny	From 1960
Fm-254	X	3.24 h	Alpha	None discovered[e]	
Pm-147	X	2.63 yr	Beta	None discovered[f]	
Pu-242	X	2.9E5 yr	Alpha	Pu urinalysis[g]	From 1954
Tm-170[h]	X	129 d	Beta, gamma	Chest counting[i]	From 1966
Tm-171[h]	X	1.92 yr	Beta	None discovered[j]	
U-233	X	1.6E5 yr	Alpha	EU urinalysis	From 1959
Bk-250	2	3.22 hr	Beta, gamma	Whole-body counting[k]	From 1960
Ce-144			Beta, gamma	Fission product urinalysis FP	From 1950s
	2	284 d		Whole-body counting	From 1960
Eu-152			Beta, gamma	Gamma urinalysis IA	From 1960
	2	12.7 y		Whole-body counting	From 1960
Eu-154			Beta, gamma	Gamma urinalysis IA	From 1960
	2	8.8 yr		Whole-body counting	From 1960
Tl-204	2	3.8 yr	Beta	Fission product urinalysis FP	From 1950s
Am242m[l]	1	152 yr	Beta (Am-242)	Trivalent actinide urinalysis	From mid 1960s
Ba-133[h]	1	10.5 yr	Gamma	Gamma urinalysis	From 1960
Cm-248	1	3.4E5 yr	Alpha	Trivalent actinide urinalysis	From mid 1960s
Es-255	1	38 d	Alpha, beta (including progeny)	Trivalent actinide urinalysis[d]	From mid 1960s
Np-239	1	2.35 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Pa-231	1	3.2E4 yr	Alpha, gamma (7%)	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Sb-124	1	60.2 yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole body counting	From 1960
Sb-125	1	2.73 yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Sc-46	1	83.8 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Tc-99	1	2.1E5 yr	Beta	None[m]	
Y-91	1	58.8 d	Beta	Fission product urinalysis FP	From 1950s
Ir-192	Shipment only	74.3 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Cd-113m[h]	0[n]	13.6 yr	Beta	Fission product urinalysis FP	From 1950s
Cs-134[h]	0	2.06 yr	Beta, gamma	Fission product urinalysis FP	From 1950s
				Whole-body counting	From 1960
Eu-155[h]	0	5.0 yr	Beta, gamma	Chest counting	From 1966
				(also whole-body counting of associated Eu-154)	From 1960

Radionuclide	Times identified	Half-life[a]	Principal radiation	Bioassay method used or available for use	Bioassay period of availability
Ho-166m[h]	0	1.2E3yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
I-131	0	8.06 d	Beta, gamma	Whole-body counting	From 1960
				Thyroid counting	
K-40[h]	0	1.3E9 yr	Beta, gamma	Whole body counting[o]	From 1960
Np-238	1	2.12 d	Beta, gamma	Whole-body counting if soon after intake	From 1960

a. Half-lives are taken from Lederer, Hollander, and Perlman (1967) and the *Radiological Health Handbook* (PHS 1970).
b. The 40-cm arc whole-body detector had a lung calibration for the 98-keV gamma from Nd-147, which also would have detected the 99- 111-keV X-rays from Np-239.
c. See Attachment C.
d. The trivalent actinide method was reviewed by an expert radiochemist who confirmed that the elements higher than Cf (specifically Bk, Fm, and Es) would also have been captured. Alpha-emitting isotopes would have been counted.
e. The half-life of Fm-254 is so short that bioassay would not have been feasible. However, the cooling of the target before handling in the laboratory resulted in little or no leftover [254]Fm. See Attachment C.
f. See Attachment B.
g. Pu-242 was used as an internal tracer starting in about 1981, but would have been detected in Pu urinalysis prior to that.
h. Isotopes that were discussed by Rupp (1969) in response to the request to use the SRS irradiation facilities for commercial and industrial development.
i. Because Tm is a bone seeker, the 84-kev gamma from the ribs would have been detectable in chest counts, despite the low yield (3.3%).
j. See Attachment D.
k. The half-life for Bk-250 is so short that the whole body count would have had to occur shortly after intake; however, the DAC (1E-7 μCi/cm^3) is 50 times larger than for Sr-90 so a significant intake would have been readily detectable in the workplace.
l. See Attachment E.
m. See Attachment A.
n. Not identified in the referenced monthly reports but were available along with bioassay methods for their detection.
o. An occupational intake would have been detectable only if the count was significantly larger than natural background.

Fecal Bioassay

Chemical bioassay of urine and fecal samples has been carried out at SRS since the 1950s to determine the individual's uptake of uranium and actinide elements such as americium, plutonium, and californium. Over the years, several techniques have been employed for these types of analyses. Methods ranging from autoradiography to high-resolution solid-state alpha spectroscopy have all been used at one time or another.

Many different methods of sample preconcentration, treatment, and preparation have been evaluated and used over the years. Each technique had some particular advantage at the time it was being used, either in terms of time required for analysis, the number of samples that could be processed simultaneously, the physical form of the final material to be counted, compatibility with other analyses that could be carried out sequentially or in parallel with the main analysis, compliance with more stringent detection levels, etc. (WSRC 1990). In contrast to a urine bioassay, a fecal bioassay can (and usually does) contain material that was never in the systemic body (i.e., insoluble material cleared from the lungs and gastrointestinal tract). Fecal bioassay is thus complementary to urine bioassay. In fact, at SRS, fecal bioassay is always used in conjunction with a urine bioassay and never alone (WSRC 2001).

The primary method for fecal bioassay used in the early and mid-1980s was gamma spectrometry with a phoswhich detector. Some of the samples analyzed by this method were subsequently dried and sent to offsite laboratories for analysis. Since 1994, fecal samples have been analyzed by a modified urinalysis procedure (Taylor et al. 1995; Taylor 2000).

In Vivo Bioassay - History of Whole-Body Counting at SRS (Taylor 2000)

In vitro bioassay was the only method available for assessing occupational intakes of radionuclides in defense facilities like SRS before the mid-1950s. Before the development of large NaI detectors, *in vivo* bioassay was performed with ionization chambers or Geiger-Mueller tubes that were rather insensitive and not practical for assessing occupational intakes of radionuclides, with the possible exception of [131]I. In 1957, Sanders proposed that a whole-body counting facility similar to that at ANL using NaI detectors be built at SRS. Before the completion of the facility in 1960, only *in vitro* bioassay, primarily urinalysis, was performed at SRS. *In vitro* urine bioassay for fission and activation products performed during this period was typically a gross beta or gross gamma analysis.

The original SRS Whole Body Counter in Building 735-A was completed in 1960 and ceased operations in September 1995. Radionuclides counted were fission products and neutron activation products with a minimum energy of 135 keV from [144]Ce and a maximum energy of 1,332 keV of [60]Co. Sometime during the early 1970s, the original chair geometry was replaced with a recumbent (bed) geometry. The single 8-in. by 4-in. NaI detector was replaced with five 4-in. by 4-in. cylindrical NaI detectors under the bed. The NaI detectors were in an arc under the bed to provide constant efficiency for all detectors (Taylor 2000).

History of Chest Counting at SRS (Taylor 2000)

The whole-body counter, which was useful for monitoring workers for intakes of gamma-emitting fission and activation products, was of no use for detecting or quantifying intakes of many actinides such as plutonium and americium. The 40-cm arc whole body counter was calibrated for [147]Nd in the lung as well as in the whole body so low-energy gamma emitters (around 100 keV) could be quantified, although the sensitivity was poor. Efforts began at SRS in the mid-1960s to develop low-energy photon detectors that would permit detection of actinides in the chest. Most efforts involved either thin NaI detectors or xenon-filled proportional counters. The xenon proportional counters, with their high resolution, showed great promise but were eventually abandoned because of their high background and low efficiency. Development of the thin NaI detector continued, and in 1966 a 1-mm-thick by 5-in.-diameter detector with a 0.001-in. window was first used to count workers involved in an incident. Thin NaI detectors were used with varying degrees of success from 1966 until 1971, when they were replaced with phoswich detectors. The results of the thin NaI counts were verified to be officially recorded in the personnel files of the workers counted during this time period.

In the late 1960s, the phoswich detectors were developed for low-energy photon and X-ray measurement. Phoswich detectors consist of a "sandwich" of phosphorescent materials (NaI and CsI), hence, the name "phoswich". This detector took advantage of its thin construction and inherent anti-coincidence counting capabilities to greatly reduce the background in the low-energy region of the spectrum. In 1972, a dual phoswich system composed of two 5-in.-diameter detectors was placed in service. Although improvements were made in the analysis of data and calibration over the years, this basic system was used for chest counting for nearly 15 years until the adoption of planar germanium detectors in the late 1980s.

In the late 1970s, small coaxial germanium detectors were installed in the steel room. These detectors were pointed at the sides of the chest in an effort to detect the 186-keV photon from the [235]U in enriched uranium, which was not readily detected by the phoswich or NaI detector. These detectors were also used to identify high-energy photon emitters, taking advantage of the superior resolution of the germanium detectors. In the mid-1980s, thick phoswich detectors specifically designed for enriched uranium were purchased. This turnkey system was used for only a few years before the germanium chest counter replaced it. In the early 1980s, solid-state planar germanium detectors were introduced at several facilities in the complex for chest counting. Routine use of a six-detector germanium chest counter began at SRS in the 1987–1988 timeframe. This was a turnkey system purchased from a commercial vendor.

The utility of routine chest counting for actinides declined throughout the 1980s and early 1990s. This decline can be related to fundamental changes in the occupational internal dose limits. During the 1970s, the occupational limit for plutonium was expressed in terms of a quantity of plutonium that could be present in the chest at any given time – the maximum permissible lung burden (MPLB). The MPLB for plutonium was 16 nCi, which at the time was considered to be detectable with the dual phoswich system. Thus, compliance with the occupational dose limits could be demonstrated with a simple chest count. In 1980, the limits changed to annual effective dose equivalent, which caused an increase in the complexity of calculations but did not cause insurmountable problems because it is closely related to the MPLB. The routine chest count was still a valuable tool for demonstrating compliance. In 1992, the limits were changed to committed effective dose equivalent. The effect of this was to reduce drastically the amount of plutonium in the chest that corresponded to the occupational limit. The reduction was so great that chest counters no longer had any chance of detecting plutonium at compliance levels, rendering routine chest counts useless for determining compliance. However, chest counters still play an important role for special chest counts and for routine chest counts if a tracer such as [241]Am might be present.

Germanium detectors have become the detectors of choice for measurement of actinide radioisotopes in the lungs. They are less efficient but much more specific (i.e., better resolution) than NaI or phoswich detectors. The SRS germanium detector system became operational in December 1989, replacing the phoswich detector system.

6.0   SUMMARY

Based on the various isotopes that were irradiated in the SRS reactors, it is clear that more than 100 have been identified as having been irradiated for the SRL and many off-site customers. As indicated in the tables, aside from the "production" radioisotopes, most materials that were irradiated were relatively small quantities, which were, according to Gray (2006), sealed in aluminum cans on arrival, handled under water, and shipped as soon as possible on removal from the reactor. During the early years, urine bioassay (either gross alpha or gross beta analyses) was the only method of evaluating intakes of radioactive materials. SRS built a whole-body counter in 1960 that was used for fission and activation products. Because the whole-body counter had limited use for the detection of actinides, efforts began in the mid-1960s to develop low-energy photon detectors that would permit actinides to be detected in the chest. Improvements in whole-body counters and chest counters provided the means of detecting lower amounts of activities in the body. The Radiation Control Group provided training and retraining of all personnel and maintained records of internal and external exposures from the beginning of operations including exposures related to incidents.

SRL did handle high activities of actinides, such as isotopes of americium, curium, and californium, for which bioassay was established. A program involving [210]Po was conducted by a small group of personnel for whom bioassay was conducted (Mahathy 2011). Whole-body counting was used to monitor intakes of most of the exotic irradiated targets. The SRL did early development work involving chemistry on [171]Tm and broken wafers ([170]Tm), which was one-time, small-scale work. Other than that, no evidence was found that the SRL worked with exotic radionuclide sources in a manner that breached the integrity of the source. Although this was small-scale work, a method for bioassay for the [170]Tm was available as indicated in Table 5-1. No evidence was found for a bioassay for [171]Tm, [249]Bk, [254]Fm, [147]Pm, and [99]Tc. These five radionuclides are discussed in Attachments A through E. There were exotic radionuclides created at SRS for which no bioassay was performed (mostly beta-only emitters), there was no evidence found that indicated that bioassay was needed.

This report addresses [249]Bk during the High-Flux demonstration period (1965-1970) (see Attachment C). However, a recently acquired document (Gibbs, 1994, p.19) indicates there was significant inventory of [249]Bk from 1975 -1980. Additional research is required to identify its source during this period.

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ATTACHMENT A
TECHNETIUM-99
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In the latter half of 1964 and into 1965 work was performed in the Savannah River Laboratory on extraction of ^{99}Tc from high level waste (DuPont 1984b, pp. 94, 118; DuPont 1964b, p. 44). The feed material would have had plenty of fission products and monitoring by fission product urinalyses and whole body counting would have been appropriate. 1.1 g of ^{99}Tc was isolated in June 1964 and work toward extracting another 1.8 g was conducted in September (DuPont 1964a, p. 44). The purity of the product was not mentioned. No mention of Tc extraction was made for subsequent years in the historical summary document (DuPont 1984b) and no mention was made in the series of quarterly reports of the Isotopic Power and Heat Sources Project starting in July 1966 (for instance, DuPont 1966b, 1967b, and others). [Note: the October – December 1966 report has not been captured.] Additionally, no mention of work with ^{99}Tc was made in any of the Savannah River Laboratory Radiation Control Group monthly reports in 1966 (whereas there is frequent mention of work specifically with curium, thorium, ^{238}Pu, ^{237}Np and occasional mention of work with thulium and polonium in 1966 or other years).

The work with ^{99}Tc extraction appears to be limited from about midyear 1964 to about midyear 1965, when it appears work on this radionuclide was abandoned.

Because the specific activity of ^{99}Tc is small (0.017 Ci/g), the potential for consequential intakes were smaller than for the other heat source radionuclides and were very small in terms of potential for inducing cancer. Table A-1 shows potential chronic intakes and committed organ doses from 1.8 g of ^{99}Tc using the common Brodsky's "magic" number (10^{-6}) to relate activity in process to potential for intakes (Brodsky 1980).

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TECHNETIUM-99
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Table A-1. Potential committed organ doses (mrem) from 1.8 g ^{99}Tc.[a]

| Target organs | Organ committed doses (mrem) | |
| | Chronic intake (365 d), 83.7 pCi/d | |
	Type F inhalation	Type M inhalation
Adrenals	3.86E-03	3.47E-03
Urinary bladder	1.60E-02	1.44E-02
Brain	3.86E-03	3.47E-03
Breast	3.86E-03	3.47E-03
Gall bladder	3.86E-03	3.47E-03
Heart wall	3.86E-03	3.47E-03
Kidneys	3.86E-03	3.47E-03
Liver	5.14E-03	4.63E-03
Muscle	3.86E-03	3.47E-03
Ovaries	3.86E-03	3.47E-03
Pancreas	3.86E-03	3.47E-03
Testes	3.86E-03	3.47E-03
Thyroid	1.01E-01	9.07E-02
R.B.M.	3.86E-03	3.47E-03
Bone surface	3.86E-03	3.47E-03
Stomach	2.07E-01	1.89E-01
S.I.	5.18E-03	6.24E-03
U.L.I.	5.59E-02	5.99E-02
L.L.I.	1.56E-01	1.69E-01
Skin	3.86E-03	3.47E-03
Spleen	3.86E-03	3.47E-03
Thymus	3.86E-03	3.47E-03
Uterus	3.86E-03	3.47E-03
ET	1.92E-01	9.33E-01
Lung	6.10E-03	2.69E+00
Colon	9.94E-02	1.07E-01
Esophagus	3.86E-03	3.47E-03
Gonads	3.86E-03	3.47E-03

a.   Maximum doses highlighted.

ATTACHMENT B
PROMETHIUM-147
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Promethium-147 was among the unusual nuclides that were produced in research quantities and evaluated for a variety of uses from 1965-1970. Promethium-147 has a half-life of 2.6 years and is a beta emitter with an average energy of 62 keV. The low energy allows for ease of handling and does not require significant shielding, which, in turn, reduces the weight required of other radioisotopes for similar applications.

As early as 1958 there was an interest in using the fission product ^{147}Pm as a fuel for SNAP batteries. As a result, a process for the recovery of ^{147}Pm was started. At that time a method was also developed for the radiochemical determination of ^{147}Pm and for ^{144}Ce and ^{91}Y, which were the major contaminants of promethium (DuPont 1984b, p. 23). As can be seen in Table 3-4 ^{147}Pm was identified several times in 1959 and 1960. However, it was not until 1965 that studies began on the use of various radionuclides as heat source materials, including ^{147}Pm. Special irradiations were made to produce test quantities of ^{170}Tm, ^{171}Tm, ^{46}Sc, and ^{204}Tl to measure the properties of these isotopes and to predict the feasibility of their large-scale production. An estimate was made of the annual production at SRP (in the fission-product fraction) of ^{147}Pm, and the recovery of ^{147}Pm from current high- activity waste was studied. The isotopic purity of aged ^{147}Pm (from the SRP waste tanks) was also measured (DuPont 1974, pp.5-6).

Because NASA had an interest in aged ^{147}Pm as a source of isotopic heat for space missions, a study of the quantity and quality of ^{147}Pm that could be produced annually at SRP was undertaken. The major source of ^{147}Pm for its recovery would be fission product waste. The quantity available after aging to allow for decay of associated ^{148}Pm would be about 1700 W(t) per reactor-year of operation. Product quality from current waste in 1965, as represented by a low ratio of ^{148}Pm/^{147}Pm, would presumably be the same as that produced at Hanford (about 5E-7). Promethium 148 emits gamma rays that could require extra shielding while ^{147}Pm is a beta emitter as indicated above. In addition, a scoping study was made to determine the quality of ^{147}Pm in fission product sludge from SRP operation. The measured isotopic ratio of ^{148}Pm in ^{147}Pm was found to be similar to the isotope ratio in Hanford fission products (0.4 ppm) (DuPont 1974, p. 7).

A process to recover ^{147}Pm from plant high activity waste was also developed in the laboratory in 1965 for use as a power source. During the extraction process, the ^{147}Pm has to be separated from ^{144}Ce since that is a major source of heat and its removal would allow easier storage of the ^{147}Pm. After separation the ^{147}Pm was then stored until most of the ^{148}Pm impurity decayed, and was then purified by ion exchange (DuPont 1984b, p.119).

A plant test to demonstrate the initial recovery steps was begun in November 1966 and demonstrated the recovery of a ^{147}Pm-lanthanide mixture from plant waste (DuPont 1984b, p. 142). In 1967 another test of the process to recover ^{147}Pm from high activity waste was successful in producing about 500 g from a lanthanide mixture. Pressurized ion exchange columns were tested with non-radioactive solutions to isolate the ^{147}Pm from the lanthanide mixture (DuPont 1984b, p. 163).

For another application suitable-isotopic purities were calculated for ^{238}pu, ^{171}Tm, and ^{147}Pm as power sources for pumps to assist or replace the heart. A heart pump would be ~ 5 inches in diameter and installed in the abdomen. It would include a steam boiler and heat exchanger with blood as coolant (DuPont 1968a, p. 45)). Plutonium-238 appeared to be the most promising power source for such pumps because it would be available in adequate supply and could be aged to remove ^{236}Pu to sustain long cycles of operation with reasonable weight of shielding. Thulium-171 would require potentially the least shielding, but is expected to be more expensive than ^{238}Pu in the necessary, highly purified form and is also handicapped by much shorter half-life (l.9 vs. 87 yr). Promethium-147,

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PROMETHIUM-147
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readily available as a fission product, had similar disadvantages of being difficult and more expensive to produce the necessary isotopic purification required for minimum shielding and it also has a shorter half-life (2.6 yr). In 1968 ^{147}Pm was also considered as a heat source for thermoelectric and thermionic power generation. Studies of compounds of promethium were conducted to provide information on the optimum fuel form selection (DuPont 1984b, p.182). A plant test of the process recovered about 2 kg of ^{147}Pm as a lanthanide mixture. In laboratory tests pressurized ion exchange columns were used to isolate approximately 1 g of ^{147}Pm from a portion of the lanthanide mixture (DuPont 1984b, p. 182).

While there was considerable development work with ^{147}Pm there is no record that indicates it was ultimately applied to the applications mentioned above. However, the separation of 500 g and 2 kg in different operations represents multicuries of ^{147}Pm activity for which no bioassay method has been identified.

In 1969 work on the development of ^{147}Pm as a heat source was concluded and no further work was planned (DuPont 1984b, pp. 193, 200 & 201) and in 1971 about 460 g ^{147}Pm and 100 g ^{144}Ce, stored for several years, were discarded to make room for ^{244}Cm (DuPont 1971b, p. 8).

ATTACHMENT C
BERKELIUM-249 AND FERMIUM-254
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Berkelium-249 and ^{254}Fm were two of many radionuclides created as part of the Transplutonium Project, evolving out of the work to create ^{252}Cf and to produce 3 kg of ^{244}Cm by 1967 (DuPont 1964a, p. 25). Berkelium-249 resulted from decay of ^{249}Cm, which was produced from multiple (n,γ) reactions starting from ^{244}Cm. The Savannah River Laboratory monthly report for September 1964, in a section on the use the tramex process for isolation of Cf, stated "Continuing analytical development study will include determination of Bk and Es, in addition to Cf, in the dissolver solution from the second Transplutonium campaign" (DuPont 1964b, p. 34). There was no mention of Bk in the October and November monthly reports for 1964. "A series of laboratory tests was made in support of the second Transplutonium separations campaign, which was started in the H Area in October." These tests focused on the separations chemistry of Pu, Am, and Cm (DuPont 1964c, p.38).

Fm-254 resulted from multiple (n, γ) reactions starting from ^{252}Cf with the final two steps being an (n,γ) reaction of the ^{253}Es and beta decay of ^{254}Es (DuPont 1971c, p. 38). Fm-254 (half-life 3.2 hr) was an intermediate step in the pathway to produce ^{257}Fm (half-life 84 d). The same historical document states that "only ^{257}Fm can be recovered in significant amounts because, even though it is at the end of the reaction chain, its 84-day half-life is much longer than those of the other fermium isotopes" (DuPont 1971c, p. 39).

The Savannah River Laboratory monthly reports for the second half of the 1960s have not been captured. The historical summary document of the Separation Technology Section indicated that in 1965 analyses were developed for ^{249}Bk and ^{253}Es as part of the Transplutonium Project (DuPont 1984b, pp. 111, 112). However, actual separation of ^{249}Bk is not mentioned until 1969 (DuPont 1984b, p. 199). The chapter in the fifty-year symposium document that discusses creation of curium and californium mentions a broader time period: "In 1968-69, a high-pressure elution development cation exchange chromatographic process was developed . . . Milligram quantities of ^{252}Cf, submilligram quantities of ^{249}Bk, and subnanogram amounts of ^{253}Es and ^{254}Fm were separated from . . . ^{244}Cm and ^{243}Am. . ." (Harbour et al. 2000, p. 172). In this time period SRS was also irradiating targets that were shipped to ORNL for separation of the Bk, Fm, and Es (DuPont 1971c, pp. 39, 40). In April 1970, tests at the Savannah River laboratory demonstrated the feasibility of isolating ^{249}Bk, ^{254}Es, and ^{257}Fm coproduced with ^{252}Cf in SRP high-flux irradiations. Berkelium is eluted after californium and before curium . . . (DuPont 1971c, pp. 37 - 40). These tests were referred to as involving tracer levels. This process was apparently intended to be used in the Multipurpose Processing Facility, under construction in 1970 in the F Area hot canyon (DuPont 1984b, p. 209, 210). The Separation Technology Section history indicates that work on isolating these elements continued into 1971 (DuPont 1984b, p. 225). This may have been in conjunction with the Californium I campaign. The Transplutonium Project history document, drafted in February 1971, indicates that "Californium I campaign is expected to yield 51 milligrams of ^{249}Bk, 6 micrograms of ^{254}Es, and 10 picograms of ^{257}Fm (DuPont, 1971c, p. 40). Note that no mention was made of ^{254}Fm. There are conflicting reports on the time of the Californium I campaign. The February 1971 document refers to it in the future and other documents show Cf work underway in early 1971 (DuPont 1971d, pp. 5, 6, 8, 13); however the reactor history states that the Californium – I campaign occurred from August 1969 to November 1970 (DuPont 1984c, p. 158). The difference might be time the targets were in the reactor versus the time the targets were being analyzed after a cooling period. The later article on this work (circa 2000) refers to submilligram quantities of ^{249}Bk not tens of milligrams (Harbour et al. 2000, p. 172) so it appears that the predicted amounts in the Californium 1 campaign were either not created or not recovered in the amounts expected.

ATTACHMENT C
BERKELIUM-249 AND FERMIUM-254
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There is no indication that the elements higher than Cf were actually isolated in the Multipurpose Processing Facility when it was operating, circa 1972. Savannah River Laboratory monthly reports in 1972 and 1973 indicate that production was focused on ^{238}Pu heat sources and ^{252}Cf neutron sources.

From an intake perspective, it is important to note concerning general contamination in glove boxes, caves, accessible surfaces, and air contamination, the overwhelming constituents would have been ^{244}Cm, ^{252}Cf, and ^{243}Am, which were being handled in large amounts. For instance just disposal of ^{244}Cm contaminated waste to the burial grounds in 1969 totaled 12,185 Ci (DuPont 1970b, p. 8). The Radiation Control Group monthly reports show that handling of ^{252}Cf in the hundreds of Ci range in the Savannah River Laboratory was common during this period. Possible exposure to pure submilligram quantities of ^{249}Bk (1.6 Ci/mg) or the microgram/picogram quantities of einsteinium and fermium would have only occurred to those handling and packaging the final products of the chemical process, which would have been handled with extreme care because of the rarity of the products. For instance, there is no mention of contamination by Bk, Es, or Fm in the April 1970 Radiation Control Group report (DuPont 1970c) (when the tests mentioned in the preceding paragraph were conducted), or in any of that series of reports in 1969 or 1971.

Possible exposure to pure ^{249}Bk appears to have occurred intermittently during 1969 through 1971 and maybe in 1968. It is likely the dates and amounts could be determined more precisely by capture of the Savannah River Laboratory monthly reports during these years. However, general contamination in the workspace would have been dominated by other radionuclides and the risk of contamination to the small amounts of pure ^{249}Bk seem low.

Both ^{254}Fm and ^{257}Fm are alpha emitters so, if present in the body, would have been excreted in urine and in theory would have been measured by the trivalent actinide urinalysis. However, because of its 3.2-hr half-life, essentially none of the excreted ^{254}Fm would have been counted considering the time gap between excretion and counting of the sample. However, the same half-life consideration applies to time involved in separating and extracting the fermium from the californium and einsteinium. For instance, the 1971 history indicates that a 15-month cooling period between the time the targets are removed from the reactor until the start of chemical processing is normal (DuPont 1971c, p. 40). If hardly any cooling time was employed then no ^{254}Fm would be left in the target (the parent ^{254}Es also has a half-life in hours). Although the article in 2000 states "subnanogram amounts of . . . ^{254}Fm were separated" (Harbour et al. 2000, p. 172), that article does not mention ^{257}Fm, which was the actual goal of the irradiation of einsteinium. It is possible that this was a typo and the sentence should have said ^{257}Fm. The purpose of creating the higher order elements was for research on their physics and chemical natures, which would not have been possible on a very small amount of something with a 3.2-hr half-life. It is concluded that no exposure occurred to ^{254}Fm as a pure radioisotope.

ATTACHMENT D
THULIUM-171
Page 1 of 2

Interest in ^{171}Tm (and ^{170}Tm) as potential heat sources seriously started in 1964. In the September monthly report of the Savannah River Laboratory, under New Products – Special Test Irradiations, "quantities of ^{46}Sc, ^{170}Tm, ^{171}Tm, and ^{204}Tl are being made in the current ^{244}Cm-producing charge" (DuPont 1964a, p.43), a test that was authorized in July (Allen 1964). The October and November monthly reports do not mention any actual handling of the material (DuPont 1964b,c). An historical summary document also mentions the above irradiation in 1964 and states that an ion exchange process for separating the thulium from the erbium target has been developed and will be used on the irradiated material (DuPont 1984a, p. 93).

The first chemistry to extract ^{170}Tm and ^{171}Tm from the irradiated erbium targets occurred in 1965 using the aforementioned technique. This was at the laboratory development phase (DuPont 1984a, p. 118). Experimentation and improvement of this technique continued in 1966 (DuPont 1984a, p. 141). During this period the material extracted consisted of a combination of ^{170}Tm and ^{171}Tm because the target material was natural erbium. The Radiation Control Group monthly report for May indicates that a study on the radiation characteristics from and shielding design needed for ^{170,171}Tm sources was underway (DuPont 1966d, p. 8). Because ^{170}Tm is a strong beta-emitter, intakes of this mixed isotopic material would have been detected by both air sampling and the fission product bioassay.

The Radiation Control Group December 1966 monthly report indicated that proposed operations for about 100 Ci of ^{171}Tm in glove boxes B-126 – 130 were approved, and that the associated beta and low-energy gamma activity was to be easily shielded. The report added that the aluminum cladding was to be removed in the High Level Caves (DuPont 1967c, p.10).

By second quarter of CY 1967 the production method had evolved to irradiation of erbium enriched to 96% ^{170}Er (DuPont 1967d, p. 21, DuPont 1984a, p. 163). This produced ^{171}Tm directly with little contamination with ^{170}Tm. Two hundred Ci of ^{171}Tm had been produced by this method at that time. In the following quarter about 60 Ci of this material was purified from the residual erbium using cation exchange chromatography (DuPont 1967e, p. 22). No work with ^{171}Tm was mentioned in the fourth quarter CY 1967 report. That report indicated that ^{238}Pu would be a better heat source than either ^{171}Tm or ^{147}Pm because of their short half-lives, difficulty in separation and purification, and cost (DuPont 1968a, p. 45). The first quarter CY 1968 report only discusses work with ^{244}Cm and ^{170}Tm; external radiation measurements were made on a 3 kCi ^{170}Tm source that also contained 1.7 kCi of ^{171}Tm but no chemistry was done (DuPont 1968b, p. 29; (Sanders et al. 1969). The second quarter CY 1968 report states that another separation of ^{171}Tm from enriched Er was made with improved chemistry methods (30 mg ^{171}Tm, 57 Ci) (DuPont 1968c, pp. 45-46). That report also stated that this concluded the work with ^{171}Tm. That the work was "deferred" was reiterated in a July 1969 letter from E. L. Albenesius, Research Manager in the Savannah River Laboratory (Albenesius 1969). No further work with potential for intakes of ^{171}Tm was found.

No mention of the work with ^{171}Tm was found in the series of monthly reports by the Radiation Control Group of the Savannah River Laboratory in 1968 or 1969, which merely indicates that there was no significant loss of control.

Bebbington, in his history of DuPont years at Savannah River, concluded "among the unusual radionuclides produced in research quantities and evaluated as heat sources and for a number of other specialized uses were: curium-244, promethium-147, thulium 170 and 171, scandium-46, thallium-204, and polonium-210. . . . . None of these nuclides, though, found significant applications" (Bebbington 1990, p. 200).

ATTACHMENT D
THULIUM-171
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The period of work with potential for intakes of essentially pure ^{171}Tm appears to be limited to 1967 and 1968. The work took place in the Savannah River Laboratory.

ATTACHMENT E
AMERICIUM-242M

Am-242m is a beta emitter with a half-life of 152 years. It has an alpha decay branch but the yield is only 0.5%. Because it does not emit significant alpha radiation it would not be counted in the trivalent actinide bioassay method. The principal decay branch of ^{242m}Am (99.5% yield) beta-decays to ^{242}Am which beta-decays to ^{242}Cm with an 82.7% yield and 16-hour half-life. So this latter transition is nearly instantaneous, and equilibrium between ^{242m}Am and ^{242}Am is rapidly established. Curium-242 is an alpha-emitter with a 163-day half-life that would have been counted by the trivalent actinide bioassay analysis. However, it takes quite a few years for equilibrium to be established between ^{242m}Am and ^{242}Cm and equilibrium is never 100% because of the branching ratios.

The question is whether the buildup of ^{242}Cm is rapid enough and sufficient such that the trivalent actinide bioassay would have been an appropriate bioassay for ^{242m}Am exposure. Because americium and curium behave the same in the body, body biokinetics do not change the ratio of ^{242}Cm and ^{242m}Am as a function of time. Assuming workers were chronically exposed to initially pure ^{242m}Am, the rate at which ^{242}Cm activity would build up in the body is shown in Table E-1, assuming a unit activity intake per day of ^{242m}Am.

Table E-1. Activity of ^{242}Cm per unit daily intake of ^{242m}Am.

Time after start of chronic intake	Cm-242 activity
1 d	0.0018
5 d	0.0087
10 d	0.017
50 d	0.082
90 d	0.140
180 d	0.249
1 yr	0.407
1.5 yr	0.504
2 yr	0.571
3 yr	0.648

The derivations and a plot of the buildup as a function of time are shown in La Bone (2011). It is unlikely that the ^{242m}Am would have been pure in the workplace environment; i.e., airborne or surface contamination would have been a mix of ^{242m}Am and ^{242}Cm so the actual ratios would have been larger than those in Table E-1, up to an equilibrium of 0.83. The activity values in Table E-1 show that an annual or biennial trivalent actinide bioassay would have had sufficient ^{242}Cm to monitor for chronic intakes of ^{242m}Am.