
Draft

**ADVISORY BOARD ON
RADIATION AND WORKER HEALTH**

National Institute for Occupational Safety and Health

**SC&A'S EVALUATION OF Cs-137/Sr-90, FISSION AND
ACTIVATION PRODUCT, AND ACTINIDE VALUES USING
INL MONTHLY AND ANNUAL WASTE REPORTS
IN RELATIONSHIP TO ASSIGNING INTAKES**

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Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 2 of 39
--------------------------------------	----------------------------------	--	----------------------------

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Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 3 of 39
--------------------------------------	----------------------------------	--	----------------------------

TABLE OF CONTENTS

Abbreviations and Acronyms	4
Executive Summary	6
1.0 Introduction and Background	9
2.0 Cs-137/Sr-90 Values	10
3.0 FAP/Sr-90 Values	14
4.0 FAP Contribution to Internal Dose	19
5.0 Actinide/Sr-90 Values.....	20
6.0 Actinide Contribution to Internal Dose.....	25
7.0 Summary and Conclusions	26
8.0 References.....	28
Attachment A. Documents and Data Used to Evaluate Cs-137/Sr-90 Values	29
Attachment B. Documents and Data Used to Evaluate FAP/Sr-90 and FAP/Cs-137 Values	33
Attachment C. Documents and Data Used to Evaluate Actinide/Sr-90 and Actinide/Cs-137 Values	38

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 4 of 39
--------------------------------------	----------------------------------	--	----------------------------

ABBREVIATIONS AND ACRONYMS

ABRWH	Advisory Board on Radiation and Worker Health
Al	aluminum
Am	americium
ANP	Aircraft Nuclear Propulsion
ATR	Advance Test Reactor
Ce	cerium
CFA	Central Facility Area
Ci	curie
Co	cobalt
CPP	Chemical Processing Plant
Cs	cesium
D&D	decontamination and decommissioning
DR	dose reconstruction
D&D	decontamination and decommissioning
ER	evaluation report
ETR	Engineering Test Reactor
FAP	fission and activation product
I	iodine
INL	Idaho National Laboratory
La	lanthanum
Max	maximum
μ Ci	microcurie
μ Ci/ml	microcurie per milliliter
MTR	Materials Testing Reactor
Nb	niobium
NIOSH	National Institute for Occupational Safety and Health
NOCTS	NIOSH OCAS Claims Tracking System
NRTS	National Reactor Testing Station
ORAUT	Oak Ridge Associated Universities Team
OTIB	Occupational Technical Information Bulletin
POC	probability of causation

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 5 of 39
--------------------------------------	----------------------------------	--	----------------------------

Pr praseodymium
Pu plutonium
RBM red bone marrow
Ru ruthenium
SEC Special Exposure Cohort
Sr strontium
SRDB Site Research Database
TAN Test Area North
TKBS Site Technical Basis Document
TRA Test Reactor Area
TSF Technical Support Facility
U uranium
Y yttrium
Zr zirconium

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 6 of 39
--------------------------------------	----------------------------------	--	----------------------------

EXECUTIVE SUMMARY

In the National Institute for Occupational Safety and Health's (NIOSH's) evaluation report (ER) for the Idaho National Laboratory (INL) Special Exposure Cohort (SEC) petition SEC-00219 of March 12, 2015 (NIOSH 2015), one of the major assumptions was that a bioassayed indictor radionuclide (strontium-90 [Sr-90] and/or cesium-137 [Cs-137]) can be used to assign dosimetric-significant fission and activation product (FAP) and actinide (alpha emitter) intakes using the ratio method. This method requires that radionuclide ratios remain fairly constant for the many processes and locations, and over most of the operating history of INL. However, SC&A found that apparently most of the radionuclide ratios recommended by NIOSH were generated by computer modeling, and that there was no indication that the computer-generated ratio values had been compared to ratio values derived from physical measurement.

In view of INL's many types of reactors, its experimental charter, and the various fuels and materials shipped in from other sites for processing, SC&A performed a preliminary analysis of some of the available INL bioassay, Brookhaven National Laboratory fuel, and swipe data in 2015 to obtain an indication of the validity of this assumption (SC&A 2015). As outlined in that report, SC&A found there were indications that the Cs-137/Sr-90 values were not always centered on unity. SC&A then searched for documentation of quantitative analyses of the fuel elements processed in the dissolver at the Idaho Chemical Processing Plant in order to determine the radionuclide components that workers may have been exposed to; however, such documentation has not been located to date. Therefore, SC&A searched the Site Research Database for other documents that could provide quantitative radionuclide data. During this search, SC&A located many INL waste reports that contained quantitative Cs-137 and Sr-90 measurements performed on the same samples (some also contained FAP and actinide analyses); these reports spanned a relatively long time period (1961–1998) for the major operational areas within INL. Quantitative radionuclide data for waste materials (air, liquid, and solid) at the INL facilities provide a good representation of the potential intake exposures to workers. Therefore, these data were analyzed in detail to evaluate if there is a reasonably consistent relationship between the Cs-137/Sr-90 concentrations, and if these indicating radionuclides could be used to assign other radionuclide intakes such as FAP for dose reconstruction (DR) purposes. In addition, quantitative actinide data in relationship to Sr-90 and Cs-137 were analyzed when available. Data for this report included both monthly and annual radionuclide concentrations. A former SC&A report analyzed mostly annual radionuclide concentrations (SC&A 2016).

Cs-137/Sr-90

A total of 315 monthly and 77 annual matched pairs of measured Cs-137 and Sr-90 activities were located and analyzed, spanning the period 1961–1998 for a number of major facilities at the INL site. Of these matched data pairs, approximately 45% contained Cs-137/Sr-90 values centered on unity, within a factor of 2 (i.e., $\text{Cs-137/Sr-90} = 0.5$ to 2.0) for both monthly and annual data pairs (there were no outstanding differences between monthly and annual pair ratios). The remaining Cs-137/Sr-90 values were outside this interval, with values ranging from 0.03 to 159. Similar results were obtained for Sr-90/Cs-137 values, with the ratio values being the inverse of the Cs-137/Sr-90 ratio values. Cs-137 activities were generally slightly greater than Sr-90 activities.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 7 of 39
--------------------------------------	----------------------------------	--	----------------------------

Fission and Activation Product Ratios

A total of 280 monthly and 103 annual matched pairs of measured Sr-90 and FAP (cerium-144 [Ce-144], cobalt-60 [Co-60], and ruthenium-106 [Ru-106]) activity were located and analyzed, spanning the period 1961–1995 for various facilities at the INL site. The measured FAP/Sr values were compared to the FAP/Sr values recommended in Table 7-3a (with a one-year decay time) of ORAUT-OTIB-0054, *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses*, Revision 03, dated February 6, 2015. A comparison ratio value centered on 1.0 would indicate agreement between the measured and recommended values; however, ratio values not centered on unity would indicate insufficient or excessive FAP intake assignments during DR. Of these matched pairs, measured Ce-144/Sr-90 values generally fell below the recommended ratio of 12.5, measured Co-60/Sr-90 values almost always were greater than the recommended value of 1.8E-4, and measured Ru-106/Sr-90 values tended to be at or below the recommended ratio of approximately 1.0 (there were no outstanding differences between monthly and annual pair ratios for these FAPs). Similar results were obtained using FAP/Cs-137 data.

Actinide Ratios

A total of 56 monthly and 13 annual matched pairs of measured Sr-90 and an actinide (americium-241 [Am-241], plutonium-238 [Pu-238], and uranium-234 [U-234]) activity were located and analyzed, spanning the period 1974–1993 for various facilities at the INL site. The measured actinide/Sr values were compared to the actinide/Sr values recommended in Table 5-22 of ORAUT-TKBS-0007-5, *Idaho National Laboratory and Argonne National Laboratory – West – Occupational Internal Dose*, Revision 03, dated March 2, 2010. A comparison ratio value centered on 1.0 would indicate agreement between the measured and recommended values; however, ratio values not centered on unity would indicate insufficient or excessive actinide intake assignments during DR. Of these matched pairs, most of the measured actinide-to-Sr-90 ratios were greater, and a few were less, than the recommended value, with considerable scatter and relatively few values falling within the 0.5 to 2.0 ratio range. The measured Pu-238 (the most significant radionuclide for internal actinide doses)-to-Sr-90 ratio had the most recorded data points, compared to the other actinides, but with ratio values that had considerable scatter and fairly poor statistical correlation. Similar results were obtained using actinide/Cs-137 data.

Conclusions

These results indicate that at INL, the Cs-137/Sr-90 values may not be centered on unity as indicated in ORAUT-OTIB-0054, Revision 03, and some FAP/Sr and FAP/Cs measured values may be greater or less than recommended for assigning FAP intakes. Also, actinide/Sr or actinide/Cs values may not be sufficiently constant (or known) as required by ORAUT-TKBS-0007-5 (Tables 5-22 and 5-23) for assigning actinide intakes, even in situations where it can be assumed that the actinides are tied to an indicating radionuclide, such as Sr-90 or Cs-137.

Therefore, SC&A finds:

1. Cs-137/Sr-90 values are not sufficiently constant to assume a ratio of unity. Using the results of Cs-137 bioassays versus Sr-90 bioassays may lead to differences in FAP intake assignments for the radionuclides listed in Table 7-3 of ORAUT-OTIB-0054, Revision 03, for DR.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 8 of 39
--------------------------------------	----------------------------------	--	----------------------------

2. Measured Ce-144/Sr-90 or Ce-144/Cs-137 values indicate that the recommended values in Table 7-3 of ORAUT-OTIB-0054, Revision 03, are reasonable for DR purposes.
3. Measured Co-60/Sr-90 or Co-60/Cs-137 values are not in agreement with the recommended values in Table 7-3 of ORAUT-OTIB-0054, Revision 03. Using the recommended ratio values would usually lead to significant underestimation of Co-60 intake for the waste materials analyzed in this report.
4. Measured Ru-106/Sr-90 or Ru-106/Cs-137 values indicate that the recommended values in Table 7-3 of ORAUT-OTIB-0054, Revision 03, are generally reasonable for DR purposes, but there was noticeable scatter in the measured ratio values.
5. Measured FAP ratio values could only be obtained for relatively long-lived radionuclides (Cs-137, Sr-90, Ce-144, Co-60, and Ru-106). Therefore, the actual ratio values for the short-lived FAPs in Table 7-3 of ORAUT-OTIB-0054, Revision 03, are not known at this time for the material analyzed.
6. Measured actinide/Sr-90 or actinide/Cs-137 values are difficult to obtain because FAPs are generally not analyzed when actinide samples are taken. The relatively few data pairs obtained for this study indicate that for the three most dose-contributing radionuclides (Pu-238, Am-241, and U-234), the measured actinide/Sr-90 or actinide/Cs-137 values were generally greater than the recommended values in Tables 5-22 and 5-23 of ORAUT-TKBS-007-5, Revision 03, some by a significant amount.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 9 of 39
--------------------------------------	----------------------------------	--	----------------------------

1.0 INTRODUCTION AND BACKGROUND

The National Institute for Occupational Safety and Health (NIOSH) responded to the Idaho National Laboratory (INL) Special Exposure Cohort (SEC)-00219 in an evaluation report (ER) of March 12, 2015 (NIOSH 2015). In that ER, and also in the technical basis document for INL, ORAUT-TKBS-0007-5, Revision 03, *Idaho National Laboratory and Argonne National Laboratory – Occupational Internal Dose*, issued March 2010, NIOSH's basis for assigning internal intakes/doses for most years and locations at INL (except those actinides covered by NIOSH's proposed SEC class and the reserved areas and dates) relies on the important assumption that the cesium-137/strontium-90 (Cs-137/Sr-90) values are approximately unity, that fission activation products (FAPs) are directly tied by a known ratio to Sr-90 or Cs-137 (as per ORAUT-OTIB-0054, *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses* [also referred to as “OTIB-0054”]¹), and that actinides are directly tied by a known ratio to Sr-90 or Cs-137 (as per Tables 5-22 and 5-23 of ORAUT-TKBS-0007-5, Revision 03). However, SC&A found that apparently most of the radionuclide ratios recommended by NIOSH were generated by computer modeling, and that there was no indication that the computer-generated ratio values had been compared to ratio values derived from physical measurement.

In a previous evaluation using a relatively small sampling of bioassay and swipe data (SC&A 2015), SC&A found indications that the measured Cs-137/Sr-90 values were not always centered on unity, and that the measured FAP/Sr, FAP/Cs, actinide/Sr, and actinide/Cs values were not constant or necessarily representative of those recommended in ORAUT-TKBS-0007-5, Revision 03. Therefore, SC&A searched for further documentation of measured quantitative radionuclide analyses of Sr-90, Cs-137, FAP, and actinide activities. Searches for documentation of quantitative analyses of the fuel elements processed in the dissolver at the Idaho Chemical Processing Plant have not produced significant data to date.

In the process of searching the Site Research Database (SRDB), SC&A located INL waste management reports that contained quantitative Sr-90 and Cs-137 measurements performed on the same samples (some also contained FAP and actinide data). These reports span a relatively long time period (1961–1998) and cover a number of the major operational areas within INL. These data provide a good representation of the materials workers could have been exposed to, and the potential intake mixtures. SC&A analyzed annual and one-of-a-kind (i.e., point sample) radionuclide pairs in a report issued in June 2016 (SC&A 2016). In response to Work Group tasking at the joint INL/Argonne National Laboratory-West meeting held in August 2016, SC&A prepared this latest report. In this report, SC&A analyzed both monthly and corresponding annual data pairs to determine if the measured-to-recommended ratio values were useful for dose reconstruction (DR) purposes, and to determine if there is any difference in resulting ratios using monthly versus annual activity data. A list of the documents that contained useful radionuclide measures and some of the recorded data that were used in this evaluation are provided in this

¹ For this analysis, SC&A used ORAUT-OTIB-0054, Revision 03 of February 6, 2015. For Cases A and D, NIOSH used ORAUT-OTIB-0054, Revision 00 PC-1 of November 19, 2007; for Case B, NIOSH used Revision 02 of March 6, 2014; and for Case C, NIOSH used Revision 03 of February 6, 2015.

Figure 2. Cs-137/Sr-90 Values for Monthly and Annual Data Pairs from INL Waste Reports

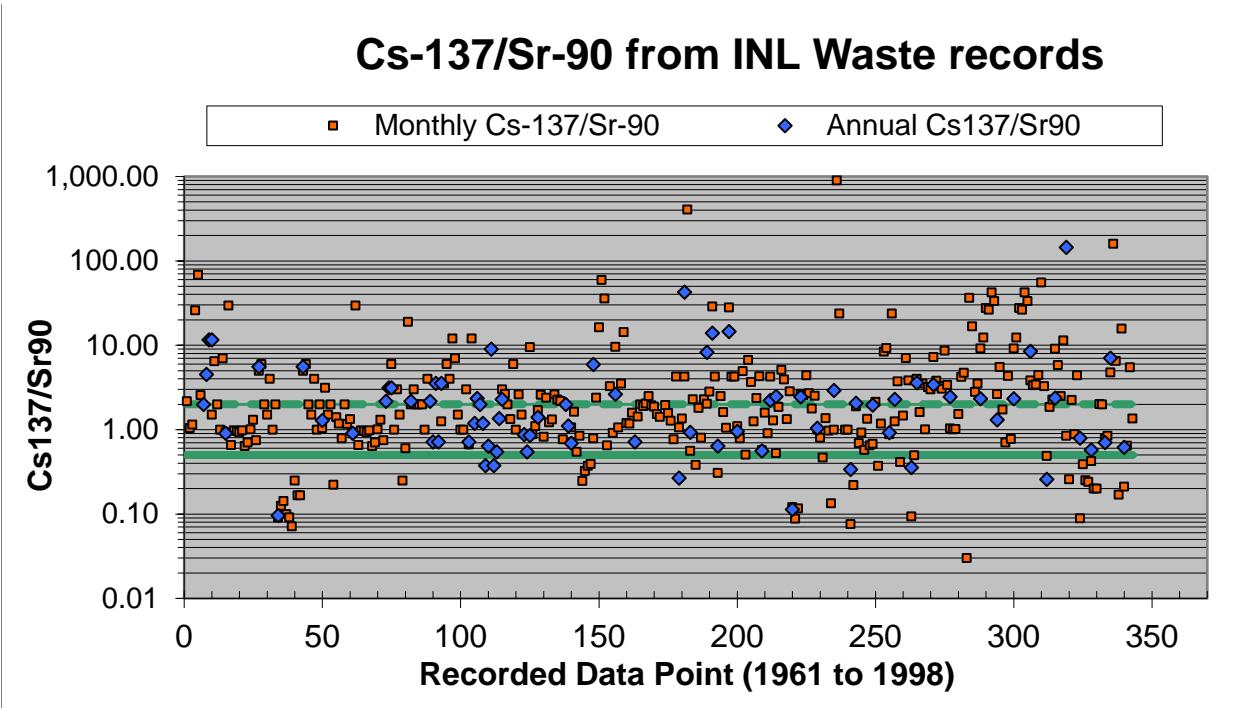
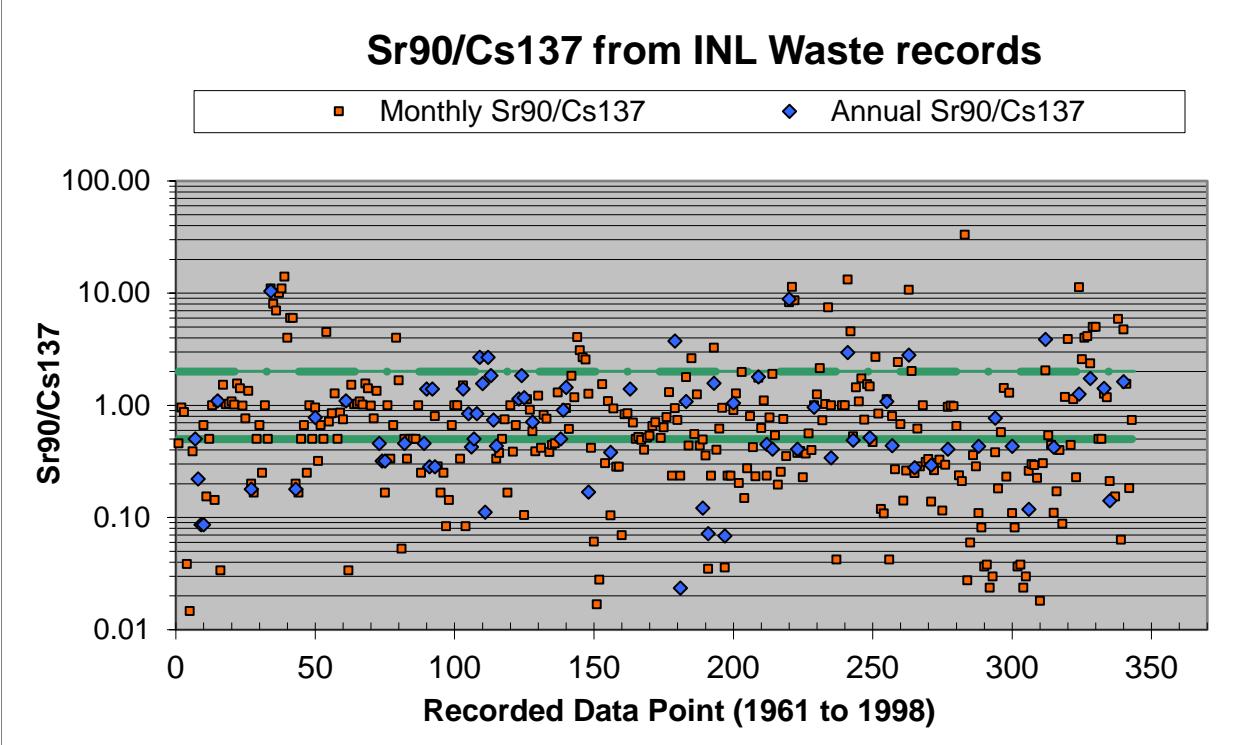


Figure 3. Sr-90/Cs-137 Values for Monthly and Annual Data Pairs from INL Waste Reports



Of the 315 monthly pairs, 45.7% contained Cs-137/Sr-90 or Sr-90/Cs-137 values centered on unity, within a factor of 2 (i.e., ratio = 0.5 to 2.0) of the recommended values; these are

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 12 of 39
-------------------------------	---------------------------	---	----------------------

represented by the data points between the solid and dashed thick green horizontal lines in Figures 2 and 3, and 59.7% falling within a factor of 3 (i.e., 0.33 to 3.0). The remaining ratio values were outside these intervals with values ranging from 0.03 to 159. Table 1 summarizes the distribution of the ratios that fell below, within, and above the recommended ratio by a factor of 2 (i.e., 0.5 to 2.0), and the corresponding data for a factor of 3 (i.e., 0.33 to 3.0) for both the monthly and annual data.

Table 1. Summary of Measured Cs-137/Sr-90 and Sr-90/Cs-137 Values Compared to Ratio Values Recommended in Table 7-3a of ORAUT-OTIB-0054

Radionuclides	Recommended NIOSH Ratio (OTIB-0054, Table 7-3a, at 1 year)	Number of Pairs	Type of Data	Percent of Pairs with Ratios <0.5	Percent of Pairs with Ratios 0.5–2.0	Percent of Pairs with Ratios >2.0	Percent of Pairs with Ratios <0.33	Percent of Pairs with Ratios 0.33–3.0	Percent of Pairs with Ratios >3.0
Cs-137/Sr-90	1	315	Monthly	12.7%	45.7%	41.6%	9.5%	59.7%	30.8%
Cs-137/Sr-90	1	77	Annual	10.4%	44.2%	45.5%	5.2%	68.8%	26.0%
Sr-90/Cs-137	1	315	Monthly	41.6%	45.7%	12.7%	30.8%	59.7%	9.5%
Sr-90/Cs-137	1	77	Annual	45.5%	44.2%	10.4%	26.0%	68.8%	5.2%

As can be seen from Table 1, there were no outstanding differences using monthly data as compared to using annual data to derive the Cs/Sr and Sr/Cs ratio values in these analyses. Also, the ratio values derived using monthly and annual activity data pairs in these analyses were similar to those obtained using only annual data in the June 2016 report (SC&A 2016), where 251 data pairs resulted in 33% of the ratio values falling in the 0.5 to 2.0 interval. The results obtained in these analyses, those contained in SC&A's June 2016 report (SC&A 2016), and those contained in SC&A's November 2015 report (SC&A 2015) were similar.

Figures 4 and 5 show the ratio values derived from the measured monthly and annual data compared to NIOSH's recommended ratios. The black bar inside the smaller shaded rectangle (gray) represents a central estimate of the value of the ratio from measured pairs (with the smaller shaded rectangle representing the 95% confidence interval³ of the central estimate of the value of the ratio from measured pairs). The center horizontal red line inside the larger shaded rectangle (pink) represents the NIOSH-recommended ratio value of approximately 1.0. The larger shaded rectangle represents the recommended ratio value, \pm a factor of 3.

³ The smaller shaded rectangle (gray) represents the 95% confidence interval of the center estimate of the ratio value derived from statistical analyses, which is different from the 95% confidence level of the original data in the data pairs.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 13 of 39
--------------------------------------	----------------------------------	--	-----------------------------

Figure 4. Comparison of Cs-137/Sr-90 Ratio of 1.0 Recommended by NIOSH with the Central Estimated Ratio and Its Confidence Interval from Measured Pairs

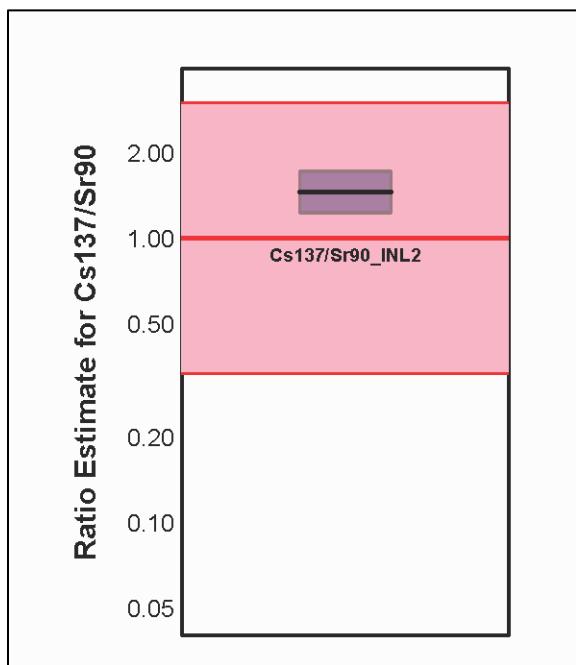
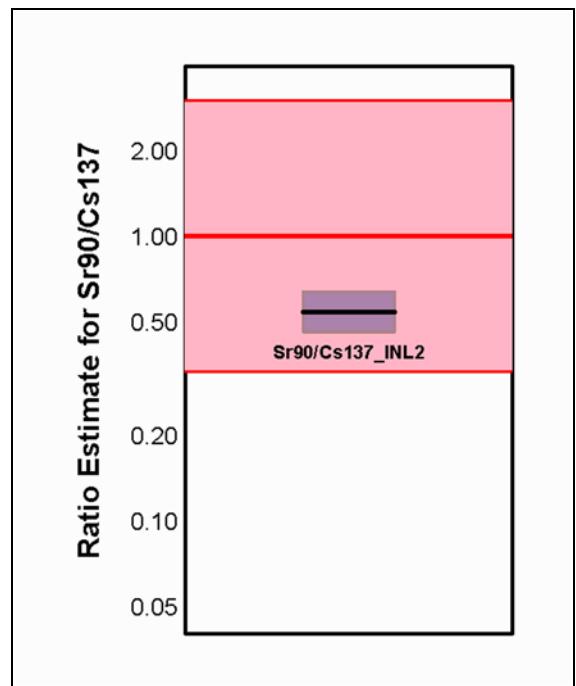


Figure 5. Comparison of Sr-90/Cs-137 Ratio Recommended by NIOSH with the Central Estimated Ratio and Its Confidence Interval from Measured Pairs



Although Figures 4 and 5 indicate that the central estimate of the ratio from measured pairs for Cs-137/Sr-90 and Sr-90/Cs-137 and its associated 95% confidence intervals generally fall within

Effective Date:	Revision No.	Document No./Description:	Page No.
01/11/2017	0 (Draft)	SCA-TR-2017-SEC001	14 of 39

a factor of 3, there was considerable scatter of the ratio values over a wide range, as shown in Figures 2 and 3.

3.0 FAP/Sr-90 VALUES

Although a large number of radionuclides are listed in Table 7-3 of ORAUT-OTIB-0054, Revision 03, many have relatively short half-lives (compared to the time from when the radionuclide was created until the time the waste sample was analyzed); therefore, when analyzing the INL waste reports, only those with relatively long half-lives (equal to or greater than approximately 1 year) were selected for analysis. A total of 280 monthly and 103 annual matched pairs of measured Sr-90 and FAP (cerium-144 [Ce-144], cobalt-60 [Co-60], and ruthenium-106 [Ru-106]) activities were located and analyzed, spanning the period 1961–1995 for various facilities at the INL site. The measured FAP/Sr values were compared to the FAP/Sr values recommended in Table 7-3a (with a one-year decay time) of ORAUT-OTIB-0054, Revision 03. A comparison ratio value centered on 1.0 would indicate agreement between the measured and recommended values; however, ratio values not centered on unity would indicate insufficient or excessive FAP intake assignments during DR. Figures 6 through 8 contain the results for the 280 monthly (represented by orange squares) and 103 annual (represented by blue diamonds) FAP INL data pairs analyzed.

Figure 6. Ce-144/Sr-90 Values for Monthly and Annual Data Pairs from INL Waste Reports

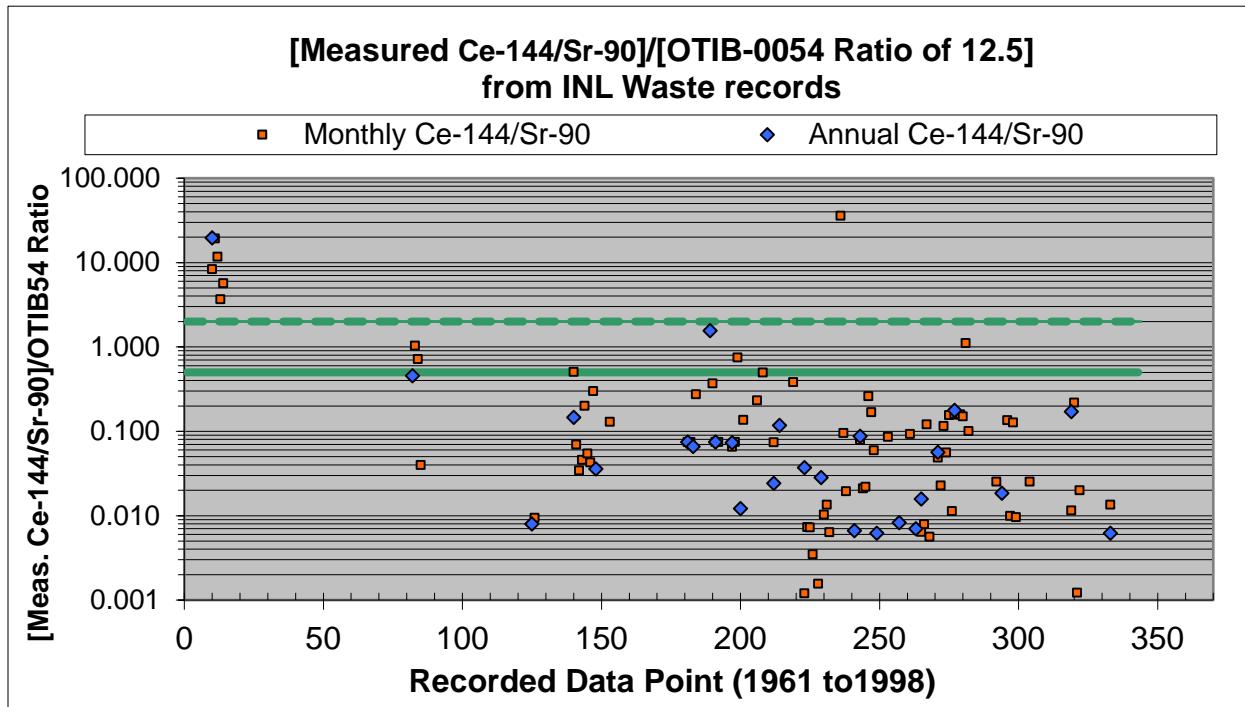


Figure 7. Co-60/Sr-90 Values for Monthly and Annual Data Pairs from INL Waste Reports

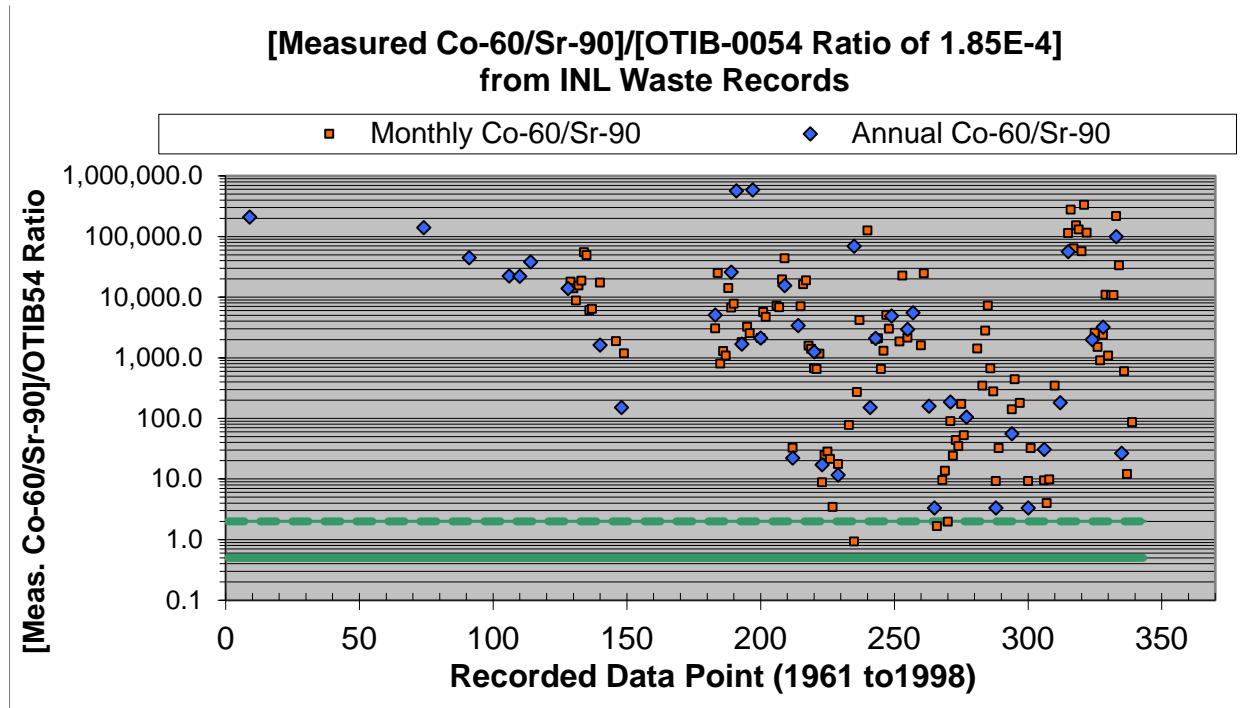
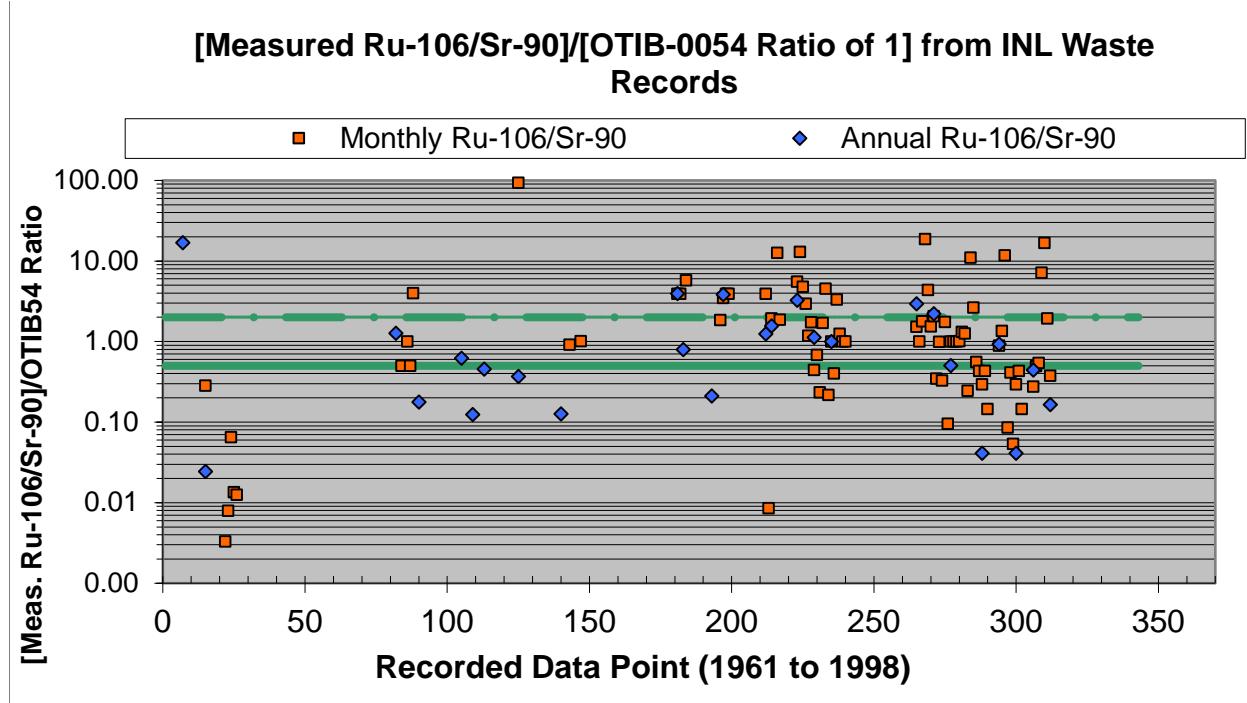


Figure 8. Ru-106/Sr-90 Values for Monthly and Annual Data Pairs from INL Waste Reports



Of the 280 monthly and 103 annual data pairs, some fell within a factor of 2 (i.e., ratio = 0.5 to 2.0) of the recommended value; these are represented by the data points between the solid and dashed thick green horizontal lines in Figures 6 through 8. The remaining ratio values were

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 16 of 39
--------------------------------------	----------------------------------	--	-----------------------------

outside this interval. Table 2 summarizes the distribution of the ratios that fell below, within, and above the recommended ratio by a factor of 2 (i.e., 0.5 to 2.0), and the corresponding data for a factor of 3 (i.e., 0.33 to 3.0).

Table 2. Summary of Measured FAP/Sr-90 and FAP/Cs-137 Values Compared to Ratio Values Recommended in Table 7-3a of ORAUT-OTIB-0054

Radionuclides	Recommended NIOSH Ratio (OTIB-0054, Table 7-3a, at 1 year)	Number of Pairs	Type of Data	Percent of Pairs with Ratios <0.5	Percent of pairs with ratios 0.5–2.0	Percent of Pairs with Ratios >2.0	Percent of Pairs with Ratios <0.33	Percent of Pairs with Ratios 0.33–3.0	Percent of Pairs with Ratios >3.0
Ce-144/Sr-90	12.5	81	Monthly	85.2%	7.4%	7.4%	82.7%	9.9%	7.4%
Ce-144/Sr-90	12.5	30	Annual	93.3%	3.3%	3.3%	90.0%	6.7%	3.3%
Ce-144/Cs-137	12.3	81	Monthly	90.1%	4.9%	4.9%	85.2%	9.9%	4.9%
Ce-144/Cs-137	12.3	30	Annual	96.7%	3.3%	0.0%	96.7%	3.3%	0.0%
Co-60/Sr-90	1.85E-04	114	Monthly	0.0%	2.6%	97.4%	0.0%	2.6%	97.4%
Co-60/Sr-90	1.85E-04	48	Annual	0.0%	0.0%	100.0%	0.0%	0.0%	100.0%
Co-60/Cs-137	1.83E-04	114	Monthly	0.9%	7.0%	92.1%	0.9%	10.5%	88.6%
Co-60/Cs-137	1.83E-04	48	Annual	0.0%	6.3%	93.8%	0.0%	6.3%	93.8%
Ru-106/Sr-90	1	85	Monthly	32.9%	38.8%	28.2%	22.4%	52.9%	24.7%
Ru-106/Sr-90	1	25	Annual	44.0%	36.0%	20.0%	32.0%	56.0%	12.0%
Ru-106/Cs-137	1	85	Monthly	50.6%	36.5%	12.9%	45.9%	45.9%	8.2%
Ru-106/Cs-137	1	25	Annual	52.0%	48.0%	0.0%	40.0%	60.0%	0.0%

As can be seen from Table 2, there were no outstanding differences using monthly data as compared to using annual data to derive the FAP/Sr and FAP/Cs ratio values in these analyses. Measured Ce-144/Sr-90 values generally fell below the recommended ratio of 12.3, measured Co-60/Sr-90 values were almost always greater than the recommended value of 1.8E-4, and measured Ru-106/Sr-90 values tended to be near or below the recommended ratio of 1.0. Similar results were obtained for FAP/Cs-137 values.

Figures 9 through 11 show the ratio values derived from the measured monthly and annual data compared to NIOSH's recommended ratios. The black bar inside the smaller shaded rectangle (gray) represents a central estimate of the value of the ratio from measured pairs (with the smaller shaded rectangle representing the 95% confidence interval of the central estimate of the value of the ratio from measured pairs). The center horizontal red line inside the larger shaded rectangle (pink) represents the NIOSH-recommended ratio value. The larger shaded rectangle represents the recommended ratio value, \pm a factor of 3.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 17 of 39
--------------------------------------	----------------------------------	--	-----------------------------

Figure 9. Comparison of Ce-144/Sr-90 Ratio of 12.5 Recommended by NIOSH with the Central Estimated Ratio and Its Confidence Interval from Measured Pairs

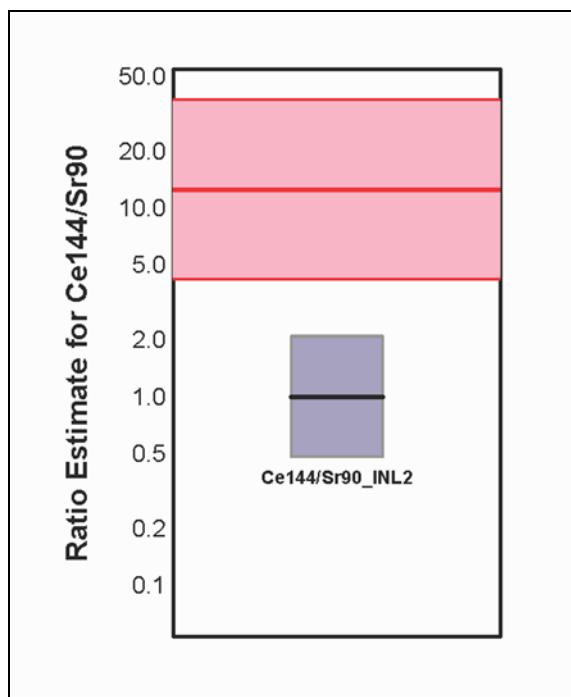
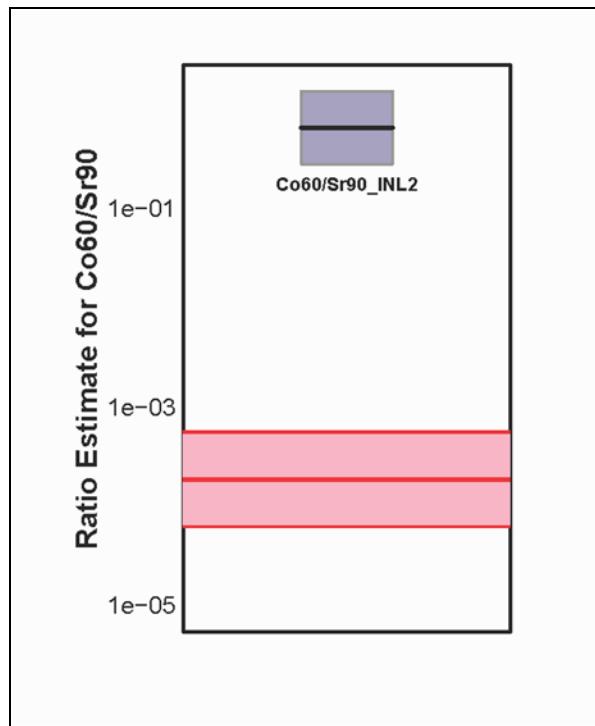


Figure 10. Comparison of Co-60/Sr-90 Ratio of 1.8E-4 Recommended by NIOSH with the Central Estimated Ratio and Its Confidence Interval from Measured Pairs



Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 18 of 39
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Figure 11. Comparison of Ru-106/Sr-90 Ratio of Approximately 1.0 Recommended by NIOSH with the Central Estimated Ratio and Its Confidence Interval from Measured Pairs

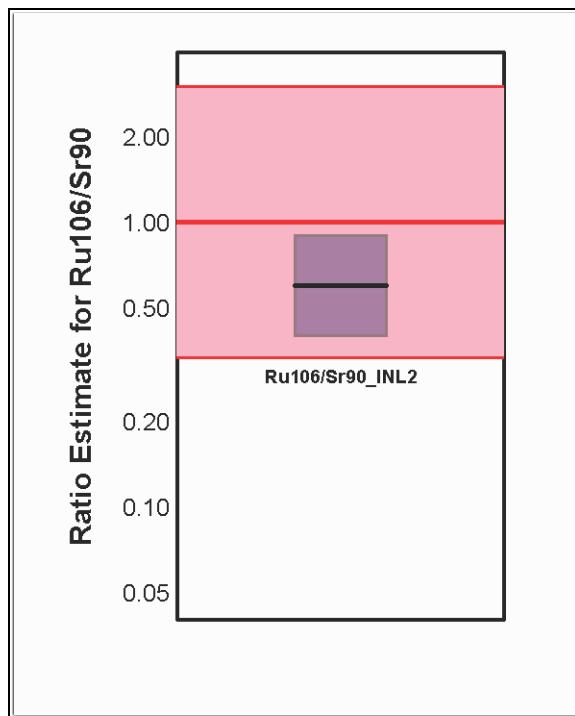


Figure 9 illustrates that the measured center estimated Ce-144/Sr-90 value, as well as the 95% confidence level area, fell below the recommended Ce-144/Sr-90 value (with a central estimate of the ratio from measured pairs of 1.0 compared to a recommended ratio of 12.5), with some scatter as shown in Figure 6.

Figure 10 illustrates that the measured center estimated Co-60/Sr-90 value, as wells as the entire 95% confidence level area, were well above the recommended Co-60/Sr-90 value (with a central estimate of the ratio from measured pairs of 0.64 compared to a recommended ratio of 1.8E-4), with considerable scatter as shown in Figure 7.

Figure 11 illustrates that the measured center estimated Ru-106/Sr-90 value, as well as the 95% confidence level area, fell slightly below the recommended value (with a central estimate of the ratio from measured pairs of 0.6 compared to a recommended ratio of approximately 1.0), with some scatter as shown in Figure 8.

Similar results were obtained for FAP/Cs-137 values.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 19 of 39
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4.0 FAP CONTRIBUTION TO INTERNAL DOSE

To determine the impact that FAP intake values and the resulting doses have on the total internal dose assignments, SC&A analyzed the FAP doses in four DR cases (A, B, C, and D⁴) obtained from the NIOSH OCAS Claims Tracking System (NOCTS). These cases were selected because they each had both FAP intakes assigned using ORAUT-OTIB-0054⁵ and actinide intakes assigned using ORAUT-TKBS-0007-5, Revision 03, Tables 5-22 and 5-23, and included major organs of interest. Table 3 summarizes this analysis.

Table 3. Summary of FAP Dose versus Total Internal Dose

Case	Indicating Radionuclide	Organ	Total Internal Dose Assigned (rem)	OTIB-0054 FAP Organ Dose (rem)	FAP % of Total Internal Dose (rem)
A	Cs-137	RBM	1.710	1.053	61.1%
B	Cs-137	RBM	1.182	1.063	89.9%
B	Cs-137	Lung	9.842	8.748	88.9%
C	Sr-90 & Cs-137	Lung	21.006	19.536	93.0%
C	Sr-90 & Cs-137	Testes	0.197	0.187	94.9%
D	Sr-90	Prostate	1.168	1.128	96.6%
D	Sr-90	Skin	0.616	0.570	92.5%
D	Sr-90	Kidney	0.827	0.700	84.6%

As can be seen from Table 3, FAP doses contribute significantly to the total internal dose, and differences in the FAP to Sr-90 or to Cs-137 values used could impact the total dose assigned and resulting probability of causation (POC) value. Therefore, there should be reasonable agreement among Sr-90 and Cs-137 ratios and among other FAP-to-Sr-90 or to-Cs-137 ratios.

Tables 4a and 4b provide the six largest contributors to these organ doses of the 17 FAPs listed in ORAUT-OTIB-0054, Revision 03. The top six contributors were selected for illustration because they constitute 80% to 95% of the total FAP internal dose assigned, depending on the organ.

⁴ A, B, C, and D are not the actual NOCTS claim numbers for these cases. They are placeholders assigned to these cases only for use in this analysis.

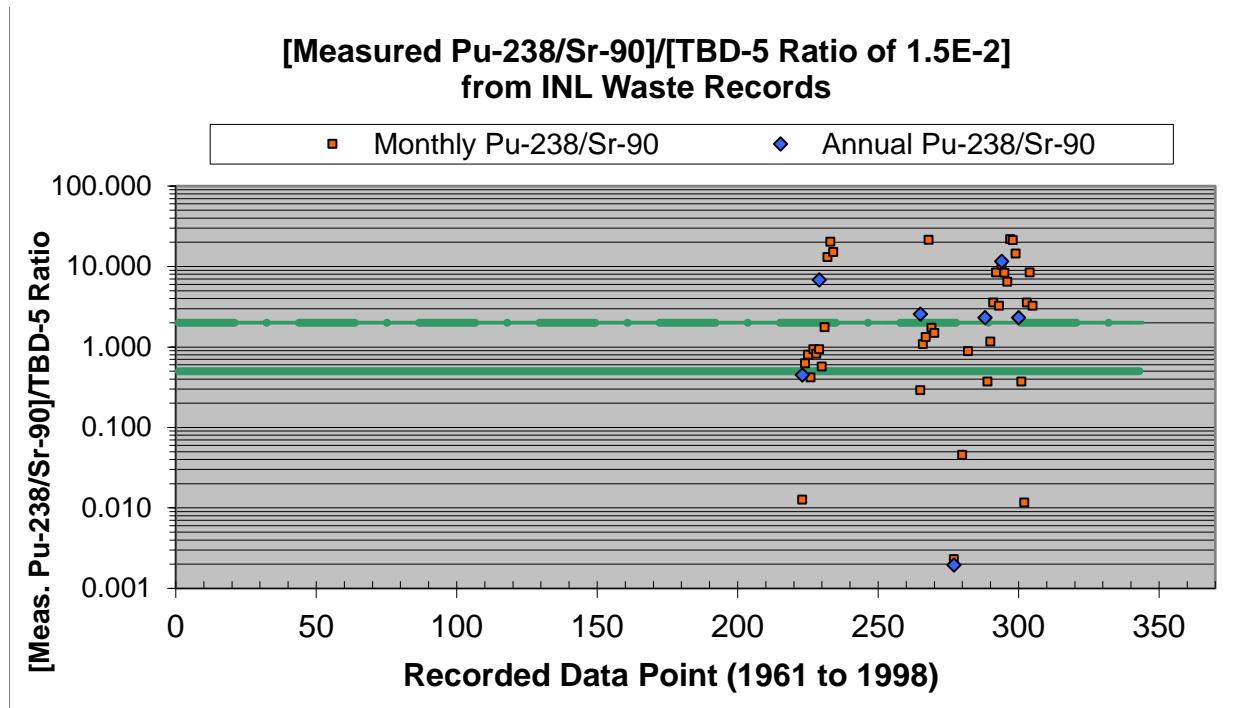
⁵ For Cases A and D, NIOSH used ORAUT-OTIB-0054, Revision 00 PC-1 of November 19, 2007; for Case B, NIOSH used Revision 02 of March 6, 2014; and for Case C, NIOSH used Revision 03 of February 6, 2015.

Effective Date:	Revision No.	Document No./Description:	Page No.
01/11/2017	0 (Draft)	SCA-TR-2017-SEC001	22 of 39

included in the waste reports even in the 1970s and afterwards. For the actinides that were available, SC&A determined the value of the measured actinide/Sr-90 (or actinide/Cs-137) and compared that value to the recommended value in Table 5-22 (or Table 5-23) to obtain a relative ratio. A relative ratio centered on unity indicates that the measured value is in agreement with the recommended value. A ratio >1.0 indicates that the measured value would assign an intake greater than that recommended in the tables, and a ratio <1.0 indicates that the measured value would assign an intake less than that recommended in the tables.

SC&A analyzed the INL waste report data for monthly and annual radionuclide activity concentrations. Only recorded and paired data points specifically listing an actinide and associated Sr-90 and Cs-137 in units of activity (μCi , Ci, etc.) on the same material were used in this analysis; this included air, liquid, and solid waste. Figure 14 contains the results for the Pu-238/Sr-90 data pairs analyzed.

Figure 14. Measured Pu-238/Sr-90 Values Compared to Ratio Values in Table 5-22 from ORAUT-TKBS-0007-5, Revision 03

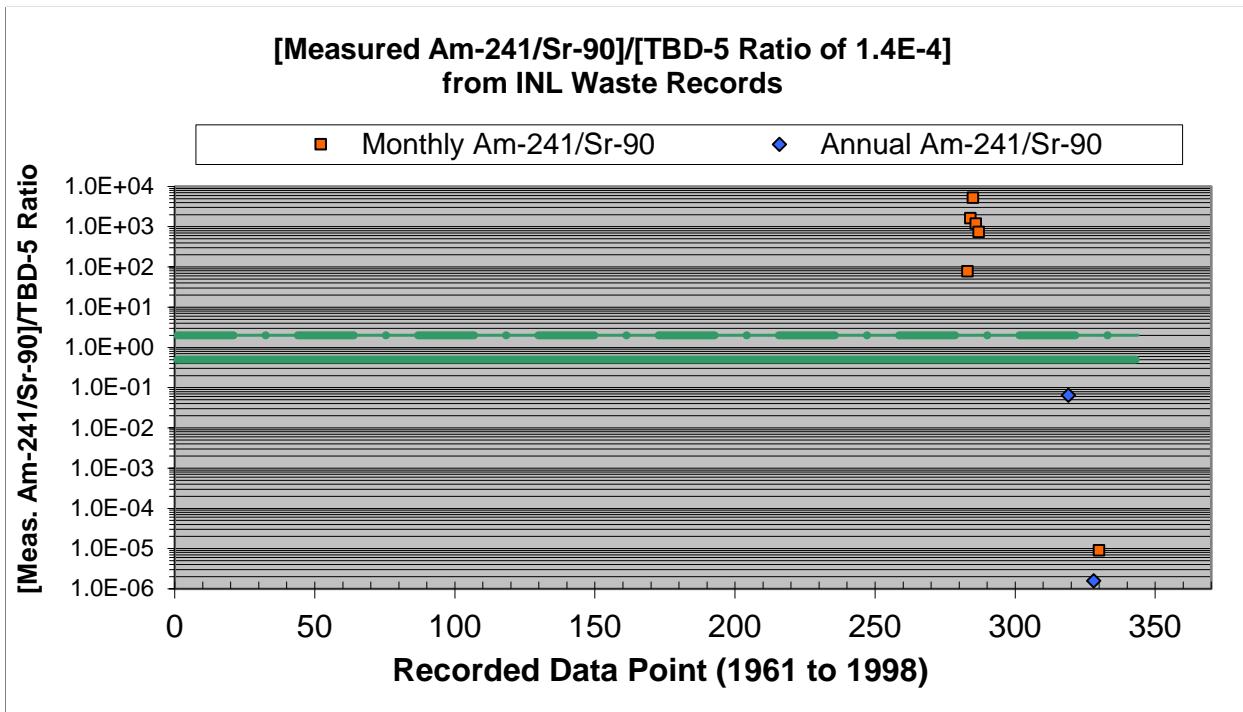


In this case, 32.5% of 40 monthly and 12.5% of eight annual Pu-238/Sr-90 data pairs fell within the 0.5 to 2.0 range; the other ratios had a wide range of values. Similar results were obtained for Pu-238/Cs-137 ratios.

Figure 15 contains the results for the Am-241/Sr-90 data pairs analyzed.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 23 of 39
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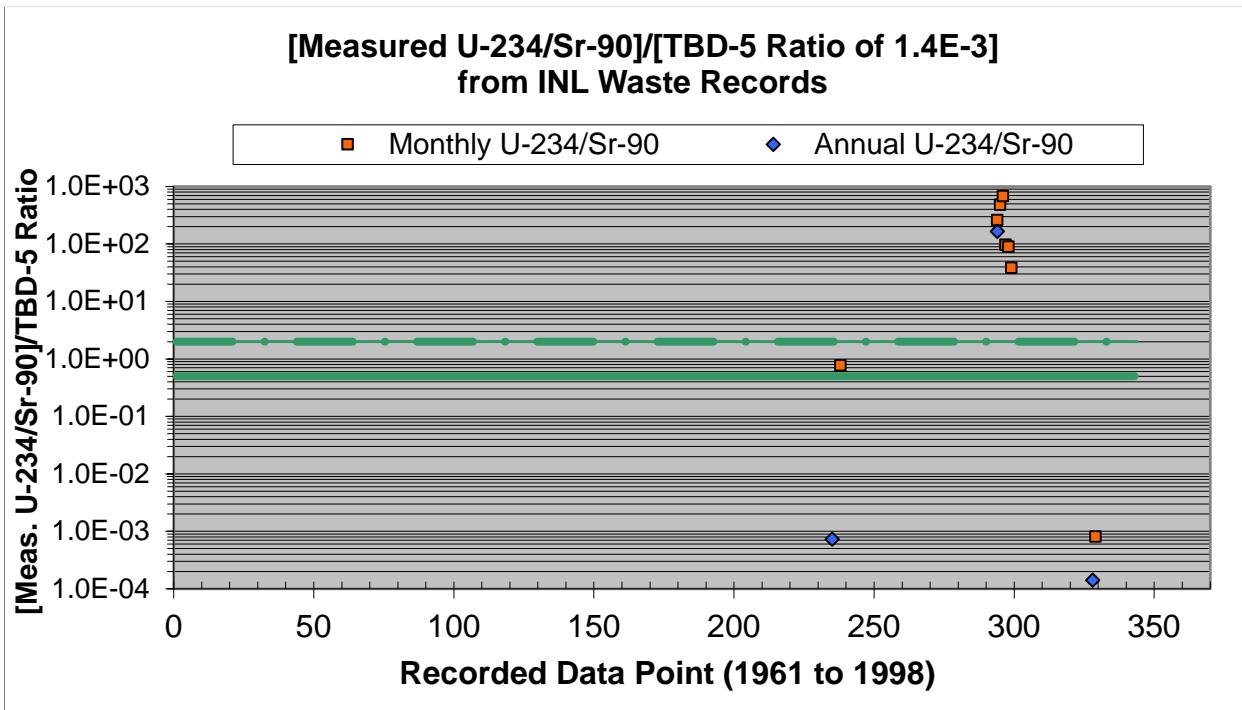
Figure 15. Measured Am-241/Sr-90 Values Compared to Ratio Values in Table 5-22 from ORAUT-TKBS-0007-5, Revision 03



In this case, none of the seven monthly and none of the two annual Am-241/Sr-90 data pairs fell within the 0.5 to 2.0 range; the other ratios had a wide range of values (one monthly Am-241/Sr-90 value is off this chart at 8.0E+08). Similar results were obtained for Am-241/Cs-137 ratios.

Figure 16 contains the results for the U-234/Sr-90 data pairs analyzed.

Figure 16. Measured U-234/Sr-90 Values Compared to Ratio Values in Table 5-22 from ORAUT-TKBS-0007-5, Revision 03



In this case, 11.1% of the nine monthly and none of the three annual U-234/Sr-90 data pairs fell within the 0.5 to 2.0 range; the other ratios had a wide range of values (one monthly U-234/Sr-90 value is off this chart at 2.2E-07). Similar results were obtained for U-234/Cs-137 ratios.

Table 5 summarizes the results using INL monthly and annual data pairs for the three most important actinide radionuclides evaluated.

Table 5. Summary of Measured Actinide/Sr-90 and Actinide/Cs-137 Values Compared to Ratio Values Recommended in Tables 5-22 and 5-23 of ORAUT-TKBS-0007-5, Revision 03

Radionuclides	Recommended NIOSH Ratio (TKBS-0007-5, Tables 5-22 & 5-23 at Max.)	Number of Pairs	Type of Data	Percent of Pairs with Ratios <0.5	Percent of Pairs with Ratios 0.5–2.0	Percent of Pairs with Ratios >2.0	Percent of Pairs with Ratios <0.33	Percent of Pairs with Ratios 0.33–3.0	Percent of Pairs with Ratios >3.0
Am-241/Sr-90	1.40E-04	7	Monthly	14.3%	0.0%	85.7%	14.3%	0.0%	85.7%
Am-241/Sr-90	1.40E-04	2	Annual	100.0%	0.0%	0.0%	100.0%	0.0%	0.0%
Am-241/Cs-137	1.30E-04	7	Monthly	14.3%	0.0%	85.7%	14.3%	0.0%	85.7%
Am-241/Cs-137	1.30E-04	2	Annual	100.0%	0.0%	0.0%	100.0%	0.0%	0.0%
Pu-238/Sr-90	1.50E-02	40	Monthly	25.0%	32.5%	42.5%	17.5%	40.0%	42.5%
Pu-238/Sr-90	1.50E-02	8	Annual	25.0%	12.5%	62.5%	25.0%	50.0%	25.0%
Pu-238/Cs-137	1.40E-02	40	Monthly	55.0%	17.5%	27.5%	50.0%	22.5%	27.5%
Pu-238/Cs-137	1.40E-02	8	Annual	37.5%	37.5%	25.0%	37.5%	37.5%	25.0%
U-234/Sr-90	1.40E-03	9	Monthly	22.2%	11.1%	66.7%	22.2%	11.1%	66.7%
U-234/Sr-90	1.40E-03	3	Annual	66.7%	0.0%	33.3%	66.7%	0.0%	33.3%
U-234/Cs-137	1.30E-03	9	Monthly	33.3%	0.0%	66.7%	33.3%	0.0%	66.7%
U234/Cs137	1.30E-03	3	Annual	66.7%	0.0%	33.3%	66.7%	0.0%	33.3%

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 25 of 39
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The most important actinide dose contributor of the radionuclides listed in ORAUT-TKBS-0007-5, Tables 5-22 and 5-23, is Pu-238 (see the next section for details). For the 40 monthly Pu-238 data pairs, the measured ratio compared to the recommended ratio fell within the interval of 0.5 to 2.0 32.5% of the time, the measured ratio was greater than the recommended ratio 42.5% of the time, and the measured ratio was less than the recommended ratio 25% of the time. There were fewer (eight) annual Pu-238 data pairs, but those showed the same trend. Similar results were obtained for actinide/Cs-137 ratios.

6.0 ACTINIDE CONTRIBUTION TO INTERNAL DOSE

To determine the effect actinide intake values, and the resulting doses, have on the total internal dose assignment, SC&A analyzed the actinide doses in the four DR cases (A, B, C, and D as previously discussed in this report). Table 6 summarizes this analysis.

Table 6. Summary of Actinide Dose Verse Total Internal Dose

Case	Indicating Radionuclide	Organ	Total Internal Dose Assigned (rem)	TBD-5 Actinide Organ Dose (rem)	Actinide % of Total Internal Dose (rem)
A	Cs-137	RBM	1.710	0.652	38.4%
B	Cs-137	RBM	1.182	0.119	10.1%
B	Cs-137	Lung	9.842	1.093	11.1%
C	Sr-90 & Cs-137	Lung	21.006	1.469	7.0%
C	Sr-90 & Cs-137	Testes	0.197	0.010	5.1%
D	Sr-90	Prostate	1.168	0.040	3.4%
D	Sr-90	Skin	0.616	0.046	7.5%
D	Sr-90	Kidney	0.827	0.127	15.4%

As can be seen from this table, actinide doses contribute significantly to the total internal dose in some cases. Therefore, differences in actinide-to-Sr-90 or to-Cs-137 ratio values used could impact the total dose assigned and the resulting POC value.

The largest contributor to organ dose of the eight actinides listed in Tables 5-22 and 5-23 of ORAUT-TKBS-0007 was Pu-238, followed by Am-241; the other six radionuclides accounted for the remaining actinide doses. Table 7 summarizes the percent of the individual radionuclide actinide dose compared to the total actinide dose assigned for each organ.

Table 7. Summary of Actinide Dose Contributors

Case	Organ	Pu-238 % of Total Actinide Dose	Am-241 % of Total Actinide Dose	Total of Other Six Actinides
A	RBM	98.7%	0.8%	0.5%
B	RBM	98.6%	0.8%	0.6%
B	Lung	92.6%	0.4%	7.0%
C	Testes	96.6%	2.5%	0.9%
C	Lung	97.6%	1.6%	0.8%
D	Prostate	97.8%	1.7%	0.5%
D	Skin	97.7%	0.3%	2.0%
D	Kidney	96.5%	2.2%	1.3%

These results indicate that plutonium and americium radionuclides are most important in assigning actinide internal doses when using ORAUT-TKBS-0007-5 for these organs, with Pu-238 contributing an average of 97% of the actinide internal dose for the organs in this analysis. Therefore, the Pu-238 and Am-241-to-Sr-90 or to-Cs-137 ratio values are important for actinide dose assignments, and there should be reasonable agreement among Sr-90 and Cs-137 ratios and among Pu-238- and Am-241-to-Sr-90 or to-Cs-137 ratios for adequate and consistent dose assignments.

7.0 SUMMARY AND CONCLUSIONS

The following list summarizes the results based on the measured data points analyzed to date and SC&A's conclusions about those results:

- **Cs-137/Sr-90** – Many of the Cs-137/Sr-90 values were not centered on unity; 45.7% of 315 monthly data points analyzed for Cs-137/Sr-90 from the 1961–1998 INL waste reports fell within a range of 0.5 to 2.0, and 59.7% fell within a range of 0.33 to 3.0, of the recommended ratio of 1.0. Some ratio values were several orders of magnitude above or below unity (similar results were obtained for annual data pairs). There were no outstanding differences in using monthly and annual data pairs for determining the Cs-137/Sr-90 ratios. Sr-90/Cs-137 results were similar. Therefore, SC&A finds that the measured Cs-137/Sr-90 values are not sufficiently constant to assume a ratio of unity. Differences in FAP intake assignments during DR for the radionuclides listed in Table 7-3 of ORAUT-OTIB-0054 could occur depending on whether Cs-137 bioassays or Sr-90 bioassays were performed.
- **FAP/Sr-90** – The analyses of a total of 280 monthly and 103 annual measured FAP/Sr-90 and FAP/Cs-137 data pairs provided the following results:
 - Ce-144: The majority (average of 95%) of the measured Ce-144/Sr-90 values were below a factor of 2 of the recommended ratio value of 12.5; Ce-144/Cs-137 results were similar. Therefore, SC&A finds that the use of the recommended Ce-144 ratios are reasonable for DR purposes.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 27 of 39
--------------------------------------	----------------------------------	--	-----------------------------

- Co-60: Almost all measured Co-60/Sr-90 and Co-60/Cs-137 values were greater than the recommended value of 1.8E-4, with many values orders of magnitude greater. Therefore, SC&A finds that the use of the recommended ratio of 1.8E-4 would result in significant under assignment of Co-60 intakes for much of the materials analyzed in this report.
- Ru-106: Many (average of 76%) of the Ru-106/Sr-90 values were near the recommended ratio value of approximately 1.0, within a factor of 2; Ru-106/Cs-137 results were similar. Therefore, SC&A finds that the use of the recommended Ru-106 ratios are generally reasonable for DR purposes, but there was noticeable scatter in the measured ratio values.
- Measured FAP ratio values could only be obtained for relatively long-lived radionuclides (Cs-137, Sr-90, Ce-144, Co-60, and Ru-106). Therefore, the actual ratio values for the shorter-lived FAPs in Table 7-3 are not known at this time for the material analyzed.
- **Actinide ratios** – Many of the measured actinide to Sr-90 or Cs-137 values were greater, and a few were less, than the recommended values in ORAUT-TKBS-0007-5, Tables 5-22 and 5-23. Some ratios were orders of magnitude above, and a few were orders of magnitude below, those recommended, with considerable scatter. Relatively few values fell within the 0.5 to 2.0 ratio (or the 0.33 to 3.0 ratio) range. For the most important (from a dose consideration) radionuclide, Pu-238, an average of 48% of the total 40 monthly and 8 annual measured Pu-238 to Sr-90 values compared to the recommended value of 1.5E-2 were equal to or below the recommended value within a factor of 2.0. Similar results were obtained for Am-241 and U-234, but there were fewer data pairs and much more scatter in the ratio values. Actinide/Cs-137 results were similar. SC&A found that measured actinide/Sr-90 or actinide/Cs-137 values are difficult to obtain, because FAPs are generally not analyzed when actinide samples are taken. Therefore, verification of the actinide ratios is not feasible from the data analyzed.
- **Use of only Sr-90 or Cs-137** – Evaluation of the many data pairs and radionuclide combinations indicates that the sole use of Sr-90 or Cs-137 does not offer an advantage in consistency when assigning FAP or actinide intakes. In addition, many bioassays only analyzed for Cs-137 or Sr-90, not both.
- **Monthly versus annual data** – Comparing the results obtained from using monthly and annual data in the waste reports to those obtained previously using bioassays, Brookhaven National Laboratory fuels, and smears (SC&A 2015) and those using mostly annual data (SC&A 2016) indicates similar results. There did not appear to be any outstanding differences in the resulting ratio values when using monthly compared to using annual data for the material analyzed for this report.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 28 of 39
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8.0 REFERENCES

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SC&A 2016. *SC&A’s Evaluation of Cs-137/Sr-90 Values and Actinides Using INL Waste Reports in Relationship to Assigning Intakes*, SCA-TR-2016-SEC007, Revision 0, SC&A, Inc., Vienna, Virginia, and Salliant, Inc., Jefferson, Maryland. June 24, 2016.

Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 37 of 39
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SRDB Ref #	Page #	Year	Area	Type of Waste	Type of Data	Measured Cs-137 Activity	Measured Sr-90 Activity	Measured Ru-106 Activity	[Ru-106/Sr-90] Divided by Table 5-22 Ratio of 1.0
140040	119	1975	CFA	Liquid	Monthly	5.09E-03	1.30E-03	2.42E-03	1.87E+00
140040	119	1975	CFA	Liquid	Monthly	4.73E-04	8.98E-04	1.73E-03	1.93E+00
140040	122	1975	CPP	Air	Monthly	2.01E-02	7.45E-03	2.20E-02	2.95E+00
140040	128-130	1975	CPP	Solid	Monthly	1.00E+01	4.20E-01	1.40E+00	3.33E+00
140040	125-126	1975	CPP	Liquid	Monthly	2.10E-02	2.16E-02	9.79E-02	4.54E+00
140040	122	1975	CPP	Air	Monthly	6.03E-02	1.37E-02	6.56E-02	4.79E+00
140040	122	1975	CPP	Air	Monthly	1.10E-01	4.13E-02	2.30E-01	5.57E+00
140040	119	1975	CFA	Liquid	Monthly	2.54E-03	4.98E-04	6.26E-03	1.26E+01
140040	122	1975	CPP	Air	Monthly	3.43E-02	1.34E-02	1.74E-01	1.30E+01
140040	117	1975	TAN	Solid	Annual	9.76E+00	4.37E+00	5.48E+00	1.25E+00
140040	128-130	1975	CPP	Solid	Annual	5.03E+02	1.71E+02	1.72E+02	1.01E+00
140040	125-126	1975	CPP	Liquid	Annual	4.78E-01	4.60E-01	5.19E-01	1.13E+00
140040	119	1975	CFA	Liquid	Annual	3.29E-02	1.33E-02	2.09E-02	1.57E+00
140040	122	1975	CPP	Air	Annual	5.97E-01	2.43E-01	7.94E-01	3.27E+00

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Effective Date: 01/11/2017	Revision No. 0 (Draft)	Document No./Description: SCA-TR-2017-SEC001	Page No. 39 of 39
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Table C.3. Documents and Data Used to Evaluate U-234/Sr-90 and U-234/Cs-137 Values

SRDB Ref #	Page #	Year	Area	Type of Waste	Type of Data	Measured Cs-137 Activity	Measured Sr-90 Activity	Measured U-234 Activity	U-234/Sr-90	[U-234/Sr-90] Divided by Table 5-22 Ratio of 1.4E-3
136493	288	1993	D&D	Solid	Monthly	2.46E-01	5.82E-01	1.78E-10	3.06E-10	2.19E-07
136493	288	1993	D&D	Solid	Monthly	2.88E-02	1.44E-01	1.64E-07	1.14E-06	8.12E-04
140040	128-130	1975	CPP	Solid	Monthly	3.19E+02	1.64E-01	1.76E-04	1.08E-03	7.69E-01
87191	157-159	1985	CPP	Liquid	Monthly	5.45E-03	7.04E-03	3.78E-04	5.37E-02	3.84E+01
87191	157-159	1985	CPP	Liquid	Monthly	4.28E-03	9.89E-04	1.24E-04	1.25E-01	8.96E+01
87191	157-159	1985	CPP	Liquid	Monthly	2.88E-03	4.09E-03	5.55E-04	1.36E-01	9.69E+01
87191	157-159	1985	CPP	Liquid	Monthly	3.10E-03	1.18E-03	4.26E-04	3.61E-01	2.58E+02
87191	157-159	1985	CPP	Liquid	Monthly	3.82E-03	6.90E-04	4.65E-04	6.74E-01	4.81E+02
87191	157-159	1985	CPP	Liquid	Monthly	2.12E-03	1.22E-03	1.16E-03	9.51E-01	6.79E+02
136493	288	1993	D&D	Solid	Annual	4.78E-01	8.27E-01	1.64E-07	1.98E-07	1.42E-04
140040	128-130	1975	CPP	Solid	Annual	5.03E+02	1.71E+02	1.76E-04	1.03E-06	7.33E-04
87191	157-159	1985	CPP	Liquid	Annual	2.82E-02	2.16E-02	4.99E-03	2.31E-01	1.65E+02

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