We put science to work.™

Savannah River
National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

A U.S. DEPARTMENT OF ENERGY NATIONAL LABORATORY • SAVANNAH RIVER SITE • AIKEN, SC

Annual Report, Spring 2015: Identifying Cost Effective Tank Waste Characterization Approaches

S. H. Reboul
R. H. Young
F. G. Smith
J. M. Pareizs
D. P. DiPrete
April 2015
SRNL-STI-2015-00144, Revision 0

SRNL.DOE.GOV

DISCLAIMER

Printed in the United States of America

Prepared for
U.S. Department of Energy

SRNL-STI-2015-00144
Revision 0

Keywords: *Waste, Characterization, Sludge, Salt, Residue*

Retention: *Permanent*

Annual Report, Spring 2015: Identifying Cost Effective Tank Waste Characterization Approaches

S. H. Reboul
R. H. Young
F. G. Smith
J. M. Pareizs
D. P. DiPrete

April 2015

Prepared in conjunction with work accomplished under contract number DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Savannah River National Laboratory™
OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

REVIEWS AND APPROVALS

AUTHORS:

S. H. Reboul, Advanced Characterization and Processing Date

R. H. Young, Analytical Development Date

F. G. Smith, Radiological Performance Assessment Date

J. M. Pareizs, Process Technology Programs Date

D. P. DiPrete, Analytical Development Date

TECHNICAL REVIEW:

M. S. Hay, Advanced Characterization and Processing, Reviewed per E7 2.60 Date

APPROVAL:

F. M. Pennebaker, Manager Date
Advanced Characterization and Processing

E. N. Hoffman, Manager Date
Engineering Process Development

S. L. Marra, Manager Date
Environmental & Chemical Process Technology Research Programs

G. R. Peterson, Headquarters Project Manager Date

ACKNOWLEDGEMENT

The authors wish to thank the individuals at DOE EM-21 for providing the funding to support this project.

EXECUTIVE SUMMARY

This annual report summarizes the status of ongoing work initiated in June 2014 – work performed to meet DOE-EM's objective of developing strategies and technologies to understand, optimize, scale, and speed up tank waste characterization. The end goal is to implement programmatic changes that accelerate tank waste processing and tank closure schedules, and at the same time reduce characterization costs, while maintaining data integrity.

To this end, the following five initial activities were performed at SRNL during the past year:
1) identification of the characterization activities driving cost and schedule; 2) investigation of potential streamlining of characterization requirements based on the relative constituent risks (with the goal of reducing characterization requirements for "low risk" and "negligible risk" constituents); 3) determination of the relative usefulness of various potential characterization bases, including laboratory analyses, waste receipt history, process knowledge, scaling factors, and historic trends; 4) utilization of the differences between sludge, salt, and post-cleaning residue to hone characterization needs as a function of waste type; and 5) investigation of alternative characterization methods holding promise for being less costly and/or less time consuming. These activities relied heavily on the use of baseline information drawn from SRNL's history of characterizing Savannah River Site (SRS) high level waste sludge, salt, and tank closure residue samples. The primary conclusions drawn from these activities are summarized below.

Of the various tank waste characterization programs, the program focusing on tank closure samples (post-cleaning residue) has clearly been the most resource-intensive and time-consuming, due to the complexity and variability of the sample matrices, and the low targeted minimum detection levels, which necessitate multiple sample preparations performed remotely in the Shielded Cells (which are considerably more labor intensive than sample preparations performed outside of the Shielded Cells), multiple cycles of radiochemical separations, and application of multi-step hybrid measurement techniques. For these reasons, the tank closure characterization program offers the greatest potential for reducing costs and schedule through application of alternative characterization approaches.

The second greatest potential for reducing cost and schedule is in the salt waste characterization program, due to the high frequency of the salt characterization campaigns (which include both salt batch qualification and quarterly waste acceptance criteria feed samples). In contrast, the sludge characterization program is a relatively small portion of the characterization scope, and therefore offers minimal potential for reducing cost and schedule at this point in time.

Of the approximately thirty analytical methods utilized during each characterization campaign, less than ten of the methods drive the costs, and a smaller number of the methods drive the schedule. The most costly methods include those for Cl-36, Ni-59/63, Tc-99, I-129, Ra-226, Th-229/230, Pa-231, and the Am/Cm isotopes. In contrast, the most time-consuming methods include those for Ra-226, Pa-231, and the Am/Cm isotopes. As such, improved methods for characterizing these constituents offer the greatest potential advantages with respect to reducing costs and schedule.

Environmental risk screening indicates that a relatively small number of waste constituents drives the future potential environmental dose. Based on waste-specific modeling and performance assessment modeling, the most significant constituents contributing potential dose from emptied, cleaned, and closed waste tanks include Tc-99, Ra-226, Pa-231, and Np-237 (and the applicable parent nuclides). In contrast, the most significant constituents contributing potential dose from Saltstone vaults containing treated/stabilized waste include Tc-99, I-129, Ra-226, and Np-237 (and the applicable parent nuclides). Clearly, these are some of the constituents for which highly accurate quantification is important.

Comparison of the quarterly salt feed characterization results with the Saltstone waste acceptance criteria (WAC) indicates that only five of the fifty-one radionuclides that are quantified have ever exceeded 10% of the WAC limits/targets. These five include Tc-99, Sb-125, Cs-135, Cs-137/Ba-137m, and Pu-238, three of which are now being removed via salt waste processing and therefore should now be less of an issue (the cesium is being removed using caustic side solvent extraction and the plutonium is being removed using monosodium titanate sorption). The very low incidence of constituent concentrations challenging the WAC limits/targets suggests an opportunity for reducing the monitoring frequency of many Saltstone constituents – specifically those with minimal likelihood of ever approaching or challenging the WAC limits/targets.

Regarding data quality, well-executed sampling and analysis typically provides the characterization data with the lowest relative uncertainties (typically ± 20%). In contrast, other available technical bases (such as tank receipt histories, process knowledge, scaling factors, and historic trends) often provide data with uncertainties in the 2X to 10X range. Despite the high relative uncertainties, such data may be adequate to meet the needs, particularly with constituents contributing low or negligible potential risks, or present at minimal concentrations that are known to be non-impactive. Such alternative characterization bases have been used effectively for safety and planning bases, and offer the benefits of being significantly less labor intensive than typical laboratory methods. As such, consideration of accuracy needs should feed the characterization requirements. While risk-driving constituents often need high accuracy quantification (in the ± 20% uncertainty range), conservative order of magnitude estimates may be adequate for "low risk" constituents. Alternatively, less frequent quantification may be an option for "low risk" constituents, and elimination from quantification may be an option for "negligible risk" constituents.

At present, the alternative laboratory approaches that would be most beneficial to SRNL's characterization programs include automation of radiochemical separation and waste removal processes, optimization of select radiochemical separation protocols, and implementation of new high sensitivity mass spectrometry instrumentation. Such approaches target quicker turnaround times and use of smaller sample aliquots, offering the potential for performing less sample preparations in the Shielded Cells.

To summarize, the primary potential approaches for increasing cost-effectiveness and timeliness of SRNL's characterization programs include: a) elimination or reduction of characterization requirements for "negligible risk" and "low risk" constituents; b) improved laboratory methods that reduce Shielded Cells processing requirements and/or standard "hands on" processing times; c) replacement of labor intensive methods with simpler methods, as applicable; d) utilization of non-laboratory methods for characterizing "low risk" constituents (this includes use of projections based on tank receipt histories, process knowledge, scaling factors, and/or historic trends); e) reduction of the characterization frequency for constituents with low potential risk or relatively consistent concentrations over time; and f) raising of highly stringent targeted minimum detection limits, as appropriate.

Over the next year, the proposed continuing scope to be performed by SRNL includes:

- Development of a technical basis and strategy for improving cost effectiveness and schedule of SRNL's tank closure characterization program
- Initiation of the design of hardware, plumbing, and software for automating select radiochemical separation and waste removal processes
- Development and feasibility testing for at least two improved radiochemical separation protocols (such as those for Ra-226, Pa-231, Tc-99/I-129, and/or Y/trivalent actinide separations)

Results of this project provide a starting point for developing more cost effective and practical characterization programs for application at the Savannah River Site, Office of River Protection, and other DOE sites involved in tank waste processing, tank waste disposition, and tank closure operations.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

AD	Analytical Development
AM	Atomic mass
ANN	Annulus
ARP	Actinide removal process
CA	Composite analysis
CST	Crystalline silicotitanate
CY	Calendar year
DOE	Department of Energy
DWPF	Defense Waste Processing Facility
E&CPT	Environmental and Chemical Process Technology
ELLWF	E-Area Low Level Waste Facility
FYSF	Fission yield scaling factor
HRR	Highly radioactive radionuclide
ICP-MS	Inductively coupled plasma mass spectrometry
INT	Interior
LIMS	Laboratory information management system
MC-ICP-MS	Multi-collector high resolution ICP-MS
MCU	Modular caustic side solvent extraction unit
MOP	Member of the public
MST	Monosodium titanate
NNDA	National Defense Authorization Act
OBIEE	Oracle Business Intelligence Enterprise Edition
PA	Performance assessment
PVC	Polyvinylchloride
RF	Radiofrequency
SA	Special analysis
SB	Sludge batch
SDC	Saltstone disposal cell
SDF	Saltstone Disposal Facility
SDU	Saltstone disposal unit
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
SRS	Savannah River Site
TACS	Time and attendance collection system
WAC	Waste acceptance criteria
WCS	Waste Characterization System
WAPS	Waste acceptance product specifications

1.0 Introduction

The U. S. Department of Energy (DOE) Office of Environmental Management (EM) has tasked the Savannah River National Laboratory (SRNL) with developing strategies and technologies to understand, optimize, scale, and speed up tank waste characterization.[a] The end goal is to implement programmatic changes that accelerate tank waste processing and tank closure schedules, and at the same time reduce characterization costs, while maintaining data integrity. This document is an annual report summarizing the status of ongoing work initiated in June 2014 – work being performed to meet DOE-EM's waste characterization program objectives.

Specifically, this document addresses the initial activities SRNL conducted to: a) gain a sound understanding of the relative costs, time requirements, and relevancy of current characterization activities/practices; b) assess potential alternative characterization methodologies; and c) identify opportunities for improving characterization practices in the context of reducing cost and schedule. This task relied heavily on baseline information drawn from SRNL's history of characterizing Savannah River Site (SRS) high level waste sludge, salt, and tank closure residue samples.

The task was divided into the following five primary activities:

- Identify characterization activities driving cost and schedule
- Investigate streamlining of characterization requirements based on the relative constituent risks (reduce characterization requirements for "low risk" constituents)
- Determine the relative usefulness of laboratory analyses, waste receipt history, process knowledge, scaling factors, and other potential characterization bases
- Utilize differences between sludge, salt, and post-cleaning residue to hone characterization needs as a function of waste type
- Investigate alternative characterization methods holding promise for being less costly and/or less time consuming

Details of each activity are presented in the sections below, by activity, with each section addressing the rationale, approach, results, conclusions, and references associated with a particular activity. Following these sections, overall conclusions based on the "sum of the findings" and a path forward are presented.

Results of this task provide a starting point for developing more cost effective and practical characterization programs for application at the Savannah River Site, Office of River Protection, and other DOE sites involved in tank waste processing, tank waste disposition, and tank closure operations.

Quality Assurance

This task was performed in accordance with the protocols identified in Task Technical and Quality Assurance Plan SRNL-RP-2014-00460 and Program Plan SRNL-RP-2014-00474. This report has been reviewed per the requirements established in Manual E7, Procedure 2.60. The extent and type of review has been identified using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

---

[a] Office of Tank Waste and Nuclear Materials (EM-20), **Work Authorization/Task Change Request**, Project HQTD1001, Topical Area WP-1.2.1 (Cost Effective Characterization Approaches), February 2004.

2.0 Identifying Characterization Activities Driving Cost and Schedule

This section focuses on the costs and durations for the analytical services provided by the SRNL Analytical Development (AD) staff to various tank waste characterization activities. While this represents only a portion of the overall services provided by SRNL groups [AD, shielded cells, Environmental and Chemical Processing (E&CPT), and statisticians], the AD effort typically has the longest duration of any of the services and consumes a significant fraction of the funding needed to complete tank characterization.

The cost and schedule duration for tank closure at Savannah River Site (SRS) has been of particular interest in the last five years, with Tanks 5, 6, 18, and 19 being characterized successfully to allow the desired end state of full-tank grouting. In addition, the primary and annular regions of Tank 16 have been sampled and characterized in preparation for grouting. Other characterization campaigns in the tank farms have also been included in this study because they represent a large analytical workload, provide additional cost and schedule metrics, and are integral to successful operations at important SRS facilities, such as the Defense Waste Processing Facility (DWPF), Actinide Removal Process/Modular Caustic-Side Extraction Unit (ARP/MCU), and Saltstone. Accordingly, the following four types of sample campaigns were examined for cost and schedule duration for the SRNL Analytical Development scope of work:

- Tank 50 quarterly supernate sample in support of Saltstone operation
- Tank 40 (typically) sludge sample in support of DWPF
- Tank 21 or 49 (typically) salt sample in support of ARP/MCU (excludes treatability studies on salt sample)
- Tank residual samples in support of permanent tank closure

2.1 Approach

To complete this study of the characterization activities driving cost and schedule for the scope of work completed by SRNL AD, data was mined from several available databases.

For the schedule duration data, the starting date for each of the methods in the analytical suite was determined by evaluating the travel copies contained in a Filemaker database, the AD Sample Management System. In addition to the date the sample was submitted to AD, this database provided the methods that were requested by the sample submitter and the cost code to which the work was to be charged. The cost code information was utilized to query other databases to determine the actual costs of labor and materials for each of the analytical campaigns.

The start date obtained from the AD Sample Management System does not reflect the date when samples were submitted to SRNL. That arrival date is earlier because the samples are received into the SRNL Shielded Cells for preliminary processing (physical testing, photographing, dissolution, aliquoting, initial radiochemical separations, etc.) and then transferred to AD on the start date.

More importantly, the start date does not represent the time at which AD starts each of the requested analytical methods. The AD staffing level is insufficient to support substantial parallel method preparations. With a task backlog of several man-months at any one time, the large analytical campaigns evaluated in this report are typically competing with other high-priority campaigns from the same or different sample submitters.

To determine the completion date for each of the requested analytical methods, the Oracle-based Laboratory Information Management System (LIMS) utilized by AD as the official sample data repository was queried. The total elapsed time for each method was calculated as the difference between the start date and the completion date. Because the analytical methods must be sequenced through the preparation and counting processes due to limited staffing, the actual hands-on time required for each method is typically significantly shorter than the total elapsed time. Each of the methods with 3 – 6 months of elapsed time could be easily completed in one month (or less) if they were prioritized to start earlier in the campaign and if equipment availability was prioritized to assure rapid turnaround of the characterization campaigns. For tank closure, the Highly Radioactive Radionuclides (HRR), such as Cs-137, Sr-90, Pu-238/239, and uranium isotopes, are typically prioritized for early completion and have significantly shorter elapsed time than the non-HRR analytes.

The completion date and total elapsed time documented in this report do not contain any of the time associated with efforts to process the samples in the Shielded Cells, to perform statistical evaluation of the data reported by AD, and to write reports compiling and evaluating such data.

To mine cost data, Oracle Business Intelligence Enterprise Edition (OBIEE) queries of the Peoplesoft business applications database were performed. The criteria used to customize the queries consisted of the start date from the AD Sample Management System, the method completion date from AD LIMS, and the cost code from the AD Sample Management system. In a few cases, the analytical work was completed prior to implementation of the Peoplesoft software, in which case an earlier database was queried to retrieve the pertinent data. The cost data was binned into two categories – the direct-charged costs (direct-charged labor and materials) and the LIMS-generated labor charges (distributed from the LIMS based on a pre-assigned number of hours per method). As an independent check of the LIMS charge category, the AD Sample Management System was used to generate a cost estimate for the specific methods that are recorded as completed in LIMS (in which the hours assigned per task are multiplied by the number of tasks and then multiplied by appropriate labor rates).

2.2 Evaluation of Durations for Analytical Methods

To obtain method-specific durations, the start date for each analytical method as obtained from the AD Sample Management System was subtracted from the completion date of the corresponding method as obtained from the AD LIMS. The durations are shown in Appendices A-D. A parameter designated as the grand average is calculated as the average of the method-specific averages as displayed in the right-most column in these appendices. This grand average indicates the approximate average duration for all the methods shown for a particular sample campaign.

In Appendix A, the durations for five Tank 50 batches (third quarter CY13 through third quarter CY14) have been calculated. The minimum, maximum, and average number of days required to complete the required methods are also displayed for each of the five batches. These statistics are calculated for the non-radiochemical and radiochemical methods separately to allow comparison of these two categories. In the right-most column, the average of the five batches is calculated on a method-specific basis. For the non-radiochemical methods, the range of durations for the five batches varied between 2 and 63 calendar days. The average duration of the methods for each batch varied from 21 to 36 days, and the grand average for all non-radiochemical methods during the five-batch evaluation was 27 days. For the radiochemical methods, the range of durations for the five batches varied between 2 and 76 calendar days. The average duration of the methods for each batch varied from 31 to 38 days, and the grand average for all radiochemical methods during the five-batch evaluation was 36 days.

In Appendix B, durations and the same statistical parameters are calculated for the three salt batches evaluated (Salt Batches 5, 6, and 7). For the non-radiochemical methods, the range of durations for the three batches varied between 2 and 45 calendar days. The average duration of the methods for each batch varied from 12 to 22 days, and the grand average for all non-radiochemical methods during the three-batch evaluation was 17 days. For the radiochemical methods, the range of durations for the three batches varied between 7 and 69 calendar days. The average duration of the methods for each batch varied from 28 to 40 days, and the grand average for all radiochemical methods during the three-batch evaluation was 34 days.

In Appendix C, durations and statistical parameters are calculated for the three sludge batches evaluated (Sludge Batches 7a, 7b, and 8). For the non-radiochemical methods, the range of durations for the three batches varied between 1 and 36 calendar days. The average duration of the methods for each batch varied from 8 to 23 days, and the grand average for all non-radiochemical methods during the three-batch evaluation was 16 days. For the radiochemical methods, the range of durations for the three batches varied between 3 and 192 calendar days. The average duration of the methods for each batch varied from 36 to 90 days, and the grand average for all radiochemical methods during the three-batch evaluation was 77 days. The grand average for the radiochemical methods are biased high by the last sludge batch (Batch 8) where a number of methods required in excess of 125 days to complete. Most of these methods were requested for the first time for Sludge Batch 8 and required special radiochemical separations starting in the Shielded Cells to meet the requested detection limits. Also, radiochemical methods for certain low-abundance radionuclides, such as Cl-36, Pa-231, Ra-226, and Th-229/230, frequently required multiple iterations to develop a preparation/analysis protocol that was successful.

In Appendix D, similar parameters are calculated for the four tank closure batches evaluated (Tanks 5, 6, 16 Primary, and 16 Annulus). For the non-radiochemical methods, the range of durations for the four batches varied between 1 and 118 calendar days. The average duration of the methods for each batch varied from 19 to 45 days, and the grand average for all non-radiochemical methods during the four-batch evaluation was 31 days. For the radiochemical methods, the range of durations for the four batches varied between 15 and 266 calendar days. The average duration of the methods for each batch varied from 80 to 173 days, and the grand average for all radiochemical methods during the four-batch evaluation was 129 days.

In Table 2-1, the key parameters which allow the most straightforward comparison of the overall durations for the various tank characterization matrices are tabulated – specifically, the maximum durations for each type of sample campaign (as opposed to the average durations, which are shorter and not reflective of the overall campaign durations). The table includes the number of non-radiochemical and radiochemical methods, respectively, requested by the sample submitter for each of the four types of waste characterization campaigns. The “best finish” and “worst finish” durations in the table provide an indication of the typical range of times needed to complete each campaign.

Table 2-1. Maximum Analysis Durations Over Multiple Batches for Selected Sample Campaigns

Sample Campaign	Type of Methods	# of Batches	# of Methods	Best Finish, in days	Worst Finish, in days
Tank 50	Non-radiochemical	5	16	42	63
	Radiochemical	5	19	62	76
Salt Batch	Non-radiochemical	3	12	21	45
	Radiochemical	3	15	48	69
Sludge Batch	Non-radiochemical	3	10	17	36
	Radiochemical	3	17	92	192
Tank Closure	Non-radiochemical	4	8	43	118
	Radiochemical	4	26	153	266

Using the maximum durations identified for all methods and for all batches within a sample campaign type, two other parameters are displayed in Table 2-1. The "best-finish" duration is determined by examining the maximum durations for the various batches in a sample campaign and then selecting the smallest maximum of this data set. For example, the salt batches in this report had maximum durations for non-radiochemical methods of 35 days for Salt Batch 5, 21 days for Salt Batch 6, and 45 days for Salt Batch 7. The "best-finish" duration is therefore 21 days, the shortest time that AD has demonstrated that it can finish all of the non-radiochemical methods for a salt batch. Similarly, the "worst-finish" duration for non-radiochemical methods is the batch maximum which is the largest. In the case of the three salt batches, the "worst-finish" duration is 45 days. AD has successfully analyzed all the batches in this campaign type within the "worst-finish" window.

Examination of the radiochemical method metrics in Table 2-1 indicates across-the-board higher values for all sample campaign types relative to the non-radiochemical methods, with the metrics for sludge batch and tank closure campaigns being significantly higher (more than 60 days). For every sample campaign type, the number of radiochemical methods requested is greater than the number of non-radiochemical methods. This difference is largest for tank closure samples, where radiochemical methods out-number non-radiochemical by 26 to 8. The "best-finish" duration for a sludge batch is 92 days, with the "worst case" being 192 days. For tank closure samples, the "best-case" scenario requires 153 days to complete the batch, while the "worst-case" requires 266.

For Tank 50 and salt batch campaigns, the starting matrix for both the non-radiochemical and radiochemical is supernatant liquid, which is far simpler to process analytically than the solid-phase sludge batch and tank closure matrices. In addition, for Tank 50 at least, a regulatory driver exists for publication of the sample results by a bilaterally agreed-upon milestone; accordingly, Tank 50 samples are given high priority for all methods, which minimizes the batch cycle (62 to 76 days).

For the sludge batch and tank closure campaigns, the starting matrix is a solid, which must be dissolved in the Shielded Cells and then transferred to AD. The non-radiochemical methods can proceed directly from this dissolution with minimal additional preparation steps (dilution, for example). Radiochemical methods for those analytes that are at high activity concentrations can also utilize the dissolution from the Shielded Cells, but the methods require significant separation efforts to segregate the analyte-of-interest from any interferences. For radionuclides at low abundance (thereby requiring low minimum detectable activities [MDAs]), a special dissolution must be completed in the Shielded Cells using a larger mass of the sample matrix. To decontaminate the special dissolution and allow safe handling in radiohoods, the special method dissolution undergoes one or more separation techniques in the cells. These cell activities add a minimum of 5 – 10 days to each special method for sludge batches and tank closure campaigns.

Once the dissolved sample aliquots reach AD from the Shielded Cells, customized preparation and counting techniques are applied to quantify the analytes for the radiochemical methods (not needed by the non-radiochemical methods). Because the tank closure sample matrices in particular are highly variable from tank to tank, they often require more than one customized preparation/analysis effort to achieve successful separation and quantitation for the radionuclides. In addition, the tank closure characterization program has much higher quality assurance requirements based on its Quality Assurance Project Plan[1] negotiated with South Carolina Department of Health and Environmental Control and the Environmental Protection Agency. Because of these quality requirements, the AD staff spends a substantially greater amount of time processing these batches, which increases duration and cost.

Besides sample preparation that is much more labor-intensive (typically 1 – 2 man days per method for up to 26 methods) than that for the non-radiochemical methods, the instrumental approaches are also much more complicated for the radiochemical methods. Typically, the radiochemical methods require two or more different instruments to measure the samples and quality control standards in each method batch (non-radiochemical methods require one). For example, the americium/curium method requires analysis using gamma spectrometry, alpha spectrometry, and inductively-coupled mass spectrometer. The samples must be scheduled to sequence through each of these instruments, and the data from the respective instruments must be processed by multiple personnel before it can be reported to the submitter. In summary, the radiochemical durations are extended (particularly for the solid matrices) relative to the non-radiochemical methods, where separations are not necessary and one instrument suffices for the quantification.

Finally, AD could theoretically complete any of its existing radiochemical methods in two months or less (regardless of matrix). This hypothesis assumes that the method has highest priority, that AD is provided much greater human resources, that the Shielded Cells facility can perform timely dissolutions and preliminary sample decontaminations, and that the applicable measurement instrumentation is fully available. Practically though, AD has more than 50 methods on its backlog at any given time and does not have the staff to achieve this level of rapid turnaround except in isolated cases. Specific workforce limitations include:

- One radiochemist/nuclear chemist to direct the customized methods and interpret data
- One radiochemist to direct the quality assurance program and interpret/report data
- Five chemists/technicians to prepare samples
- Two chemists to operate counting instruments and reduce data

AD is currently staffed such that methods associated with large sample campaigns (such as the four types discussed in this report) must be processed in parallel with other backlogged samples over a period of 1 – 6 months. To increase staff and instrumentation to accommodate less-than-one-month turnaround on all methods would not be a cost-effective approach for providing this highly specialized, labor-intensive analytical service because the AD workload can fluctuate substantially depending on site funding levels.

2.3 Evaluation of Cost for Analytical Methods

The same four types of sample campaigns were analyzed to determine the cost of the AD services required to analyze the non-radiochemical and radiochemical methods for the analytes requested by the sample submitter. The costs have been binned into two categories: 1) the labor charges distributed by application of a LIMS charging mechanism; and 2) the direct charges for the labor hours each individual worker enters into the site Time and Attendance Collection System (TACS) and for the cost of materials. To determine the costs distributed through the LIMS in the first category, the pre-assigned time to complete each method was multiplied by the number of submitted samples and then multiplied by the LIMS labor rate to obtain the charge for each method. This process was repeated for all methods for a sample type to get the total LIMS-generated charge.

Based on data mined from two Oracle databases, the LIMS-generated charges, the direct charges, the total charges, and the total charges/sample are shown in Appendix E. The Appendix also includes an estimate of the LIMS-generated charges that was obtained with a planning tool contained in the Filemaker Sample Management database. An example of the LIMS estimating worksheet is included as Appendix F.

A significant challenge in mining the AD cost data was defining the proper time interval for which charges should be examined. Many of the cost codes used for these sample campaigns were used to collect costs for multiple batches of samples of a particular sample type or for other scopes of work not related to the waste characterization examined in this report. Therefore, this cost evaluation is subject to a higher uncertainty level than would have been present if each sample batch had been assigned a unique cost code. However, the difference between the LIMS estimated and actual charges is less than 15% for all four sample types (for example, the LIMS estimate for the three Salt Batch campaigns was $47K while the actual LIMS-generated average charges was $40.9K, which is 13% less than the estimate). This agreement between the estimate and actual costs suggest that the data mined from the Oracle data bases was an adequate representation of the LIMS-generated cost element (Category 1).

On a per-sample basis, the actual LIMS-generated costs were approximately $20K per sample for the Tank 50, salt batch, and sludge batch sample types. The tank closure sample type was the only campaign that had a higher per-sample cost ($33.7K) for actual LIMS-generated costs.

The direct charges appear to be much more variable between the batches in each sample campaign. Despite this variability, the averages for each sample type provide a reasonable metric for comparing the costs. The Tank 50, salt batch and sludge batch campaigns again had the lowest direct charges for labor and materials at about $5.5K to $9K per sample. The direct charges for the tank closure campaigns at $31.5K were significantly higher than that for the other sample campaigns.

The same trend is evident for the total costs, where the per-sample cost for the Tank 50, salt batch, and sludge batch campaigns averaged $26.9K, 29.5K, and 26.8K, respectively. While the sludge batch preparations involved more complicated preparations, some of which start in the shielded cells, AD was required to analyze about 10% more methods for Tank 50 and salt batch campaigns. Also, the sludge batch had more samples in each batch (5 per batch), which typically resulted in lower direct charges because the preparation process was more efficient with larger batches.

The per-sample cost for tank closure campaigns averaged $65.2K. The tank closure costs were significantly higher because more of the sample preparations began in the shielded cells, which required AD to expend more labor and materials to launch the preparations performed by the shielded cell organization. Also, the tank closure batches were so large that the AD staff often had to create two sub-batches to process the 12 samples per batch. This approximately doubled the labor required to prepare the tank closure batches. Another significant increase in the labor arose because AD was required to meet the complex tank closure quality assurance program negotiated among SRNL, Savannah River Remediation, and state and federal regulators. These quality assurance protocols required significant effort during the counting, data reduction, and documentation phases of the batch processing, including preparation of case narratives and quality assurance checklists. None of the other sample campaigns involved this level of rigorous quality assurance.

2.4 Evaluation of Cost and Duration for Individual Radiochemical Methods for Tank Closure

To gain insight into the specific methods that drive the cost and duration for analytical studies, the radiochemical methods performed for the tank closure characterization were studied in more detail. The non-radiological methods for this sample type are all much less labor-intensive and therefore were not evaluated as drivers of either cost or schedule. To determine the costs of the radiochemical methods, the labor hours were estimated for the following phases of sample preparation and analysis: 1) direct charges by Shielded Cell operators for dissolution and initial sample decontamination steps; 2) LIMS per-method charges for Analytical Development labor; 3) direct charges by Analytical Development technicians for sample preparation; and 4) direct charges by chemists and task supervisors for counting operations, data reduction, data review, and preparation of quality assurance documentation. The labor estimates for Phases 1 and 4 were derived from discussion with the task supervisors involved with each task while those from Phases 2 and 3 were obtained from the LIMS hours-per-method list and the method-specific Research and Development directions, respectively. The labor hours in each phase were then converted into costs using the labor rates for the current fiscal year. The results of this evaluation are displayed in Appendix G.

A reasonable approach to identifying the most expensive radiochemical methods is to normalize the individual costs for each method to that for the most expensive method. This normalization is shown as the Relative Cost, % column at the right of Appendix G, with the Am/Cm method being used for normalization. If >60% of the cost of the Am/Cm method is used arbitrarily to filter for the most expensive methods, one observes that Am/Cm, Pa-231, Th-229/230, Cl-36, Ra-226, I-129, Tc-99, and Ni-59/63 (in decreasing order of cost) are the eight most costly methods. One also observes that all of these methods, except Ni-59/63, has a large cost contribution from the initial sample preparation steps (beyond the peroxide fusion or aqua regia digestions common for all of the less expensive methods) required in the Shielded Cells.

The durations for each method have been tabulated previously and displayed in Appendix D. By looking at the average durations for the four tank closure characterizations, one identifies Pt-193, K-40, Cl-36, Ra-226, Nb-94, Pa-231, Am/Cm, C-14, Th-229/230, Np-237, Cs-135, and Zr-93 (in decreasing order of duration) as the methods that have historically required the longest cycles for analysis. Of these methods, Pt-193, K-40, and Cl-36 have recently been deleted from the analyte list for future tank closure characterizations. The remaining nine methods represent the prime candidates for elimination if shorter schedule durations are needed for upcoming tank closures and if their deletion can be justified from a risk standpoint. An alternative approach would be to develop more rapid analytical methods and/or utilize other technical bases that support quicker characterization of these particular radionuclides. If both cost and schedule reductions are important considerations, then the Am/Cm, Pa-231, and Ra-226 methods are the ones that should be considered first as candidates for potential elimination or potential alternative characterization methods.

2.5 Frequency of Campaigns and Annualized Costs

If one considers the frequency with which SRNL Analytical Development receives samples from each of the campaign types evaluated in this report, one can estimate an annualized cost for each type by multiplying the frequency by the number of samples/campaign and then by the cost/sample. Using data shown in Appendix E, the annualized cost for each campaign type is shown in Table 2-2. For the salt matrix (taken as the sum of the Tank 50 and Salt Batch campaign types), the annualized cost is about $400K. For the sludge batch campaign, the annualized cost is much lower at $135K while, for the tank closure campaign, it is about $1050K.

On an annualized basis, the relative percentage of funds expended for sludge characterization is about 9%; for salt characterization, about 25%; and for tank closure characterization, about 66%. Therefore, a reasonable conclusion is that reductions in tank closure and, to a lesser degree, salt batch characterizations would offer the most potential for cost savings.

Table 2-2. Annualized Cost for Tank Characterization by Campaign Type

Sample Campaign	**Frequency**	**# of Samples/campaign**	**Cost/sample**	**Annualized Cost**
Tank 50	3 months	3	26.9K	323K
Salt Batch	9 months	2	29.4K	78.4K
Sludge Batch	12 months	5	26.8K	134K
Tank Closure	9 months	12	65.2K	1043K

2.6 Conclusions

Four types of sample campaigns were examined for cost and schedule duration for the SRNL Analytical Development (AD) scope of work:

- Tank 50 quarterly supernate sample in support of Saltstone operation
- Tank 40 (typically) sludge sample in support of DWPF
- Tank 21 or 49 (typically) salt sample in support of ARP/MCU (excludes treatability studies on salt sample)
- Tank residual samples in support of permanent tank closure

A number of factors drive the cost and schedule for these campaigns. The number of special sample decontamination preparations that must be completed in the Shielded Cells is higher for the sludge batch and much higher for tank closure characterization than for the salt campaigns. Another factor driving cost and schedule, particularly for tank closure, involves the number of analyses that require research and development and/or rework to obtain a reliable measurement. Other factors include the number of analyses that require multiple rounds of decontamination to achieve low minimum detection limits, the number that require integration of multiple measurement techniques, and the number that entail enhanced quality assurance protocols and documentation (all of these are greater for tank closure characterization).

This study revealed that the cost per sample is roughly \$30K for all sample campaign types, except tank closure where the cost is \$65K/sample. On an annualized basis, the relative percentage of funds expended for sludge characterization is about 9%; for salt characterization, about 25%; and for tank closure characterization, about 66%. Method durations averaged about 35 days for salt batches and Tank 50 salt feed, about 75 days for sludge batches, and about 130 days for tank closure.

An evaluation of the costs for the radiochemical methods, which dominated the total costs for tank closure, revealed that Am/Cm, Pa-231, Th-229/230, Cl-36, Ra-226, I-129, Tc-99, and Ni-59/63 were the most expensive methods. Of these, Am/Cm, Pa-231, and Ra-226 were also among the set of methods that were found to have the longest durations. If both cost and schedule reductions are important considerations, then these three methods are the ones that should be considered first as candidates for potential elimination or potential alternative characterization methods.

The greatest potential for reducing analytical cost and schedule durations clearly lies with tank closure characterizations. Both the salt programs (Tank 50 and salt batch) cumulatively consume more funding than the sludge batch characterization and are good candidates to investigate for potential savings, particularly if the frequency of batch qualifications is increased. In such cases, even small reductions in cost and turnaround times would be advantageous to the salt characterization campaigns. The sludge batch characterization has the least potential for cost savings although those methods that require sample decontamination in the Shielded Cells to attain low method detection limits could be evaluated.

2.7 Reference

[1] Pavletich, J. P., "*Liquid Waste Tank Residuals Sampling – Quality Assurance Program Plan*," SRR-CWDA-2011-00117, Rev. 1, July 2013.

3.0 Streamlining Characterization Requirements Based on Relative Constituent Risks

As part of this program to develop a more cost effective approach to waste characterization, it is proposed to consider a risk based approach that focuses the waste characterization on those radionuclides that pose a potential risk from dose to offsite members of the public. To establish some context for the current work, a Performance Assessment (PA) is required by DOE Order 435.1 to provide a technical basis to ensure that Department of Energy (DOE) low-level waste disposal facilities comply with radiological protection requirements established in the Order. A Composite Analysis (CA) is required by DOE Order 435.1 to provide a reasonable expectation that Department of Energy (DOE) low-level waste disposal, high-level waste tank closure, and transuranic waste disposal ensure radiological protection of the public. For a CA, the Order requires an accounting of all sources of DOE man-made radionuclides and DOE enhanced natural radionuclides that are projected to remain on the site after site operations have ceased. A Special Analysis (SA) is performed to ensure that unique waste disposal conditions deviating from those considered in the facility PA still comply with radiological protection requirements of DOE Order 435.1.

3.1 Approach

The approach taken in this work is a refinement of the screening process performed prior to PA, CA and SA calculations to reduce the number of radionuclides evaluated to a manageable number. The latest data compiled by the Savannah River National Laboratory (SRNL) includes 888 radionuclides that have ingestion dose coefficients and a total of 1252 radionuclides that would give some external or internal dose to an offsite member of the public. Typically, for facility PAs and the site CA the number of radionuclides considered in the analysis is reduced by first assuming an impossibly large starting inventory for each radionuclide (~1 million curies) performing a simple transport and decay calculation from the source to a receptor location and eliminating those radionuclides from further analysis that do not yield a significant dose to an offsite member of the public under these conditions (e.g. < 0.01 mrem/yr). This process typically reduces the number of radionuclides considered in PAs or the CA by an order of magnitude to less than 100.

The proposal evaluated in this work is to perform a more realistic screening calculation that is demonstrably conservative but is still at a level well below detailed PA modeling. It is anticipated that this approach will further reduce the number of radionuclides that must be considered in a characterization study by approximately another order of magnitude to on the order of 10 species. The cost associated with performing the risk analysis would be relatively small.

3.1.1 Risk Screening Model

GoldSim^{TM} is a graphically based programming environment that allows a modular approach to model development. The GoldSim^{TM} software[1] has been used to model radionuclide transport and perform dose calculations for several applications at SRS. GoldSim^{TM} is a convenient modeling tool for simple applications because it offers the capability of modeling one-dimensional radionuclide transport and decay and also includes the capability of performing probabilistic simulations for uncertainty analysis. GoldSim^{TM} models have typically been used to perform screening calculations such as those alluded to in the above discussion (e.g. Taylor SRNS-STI-2008-00117 [2]) and a GoldSim^{TM} model was used for all of the SRS CA modeling.[3] The risk based screening model used in this work was implemented in GoldSim^{TM} (Version 10.50 SP 3) and was to a large extent derived from the CA model.

While not all aspects of the risk screening model will be described here, a few parts of the model are shown in the figures below to provide a brief overview of how the risk screening calculations were performed. Figure 3-1 shows a screen capture from the GoldSim^{TM} model displaying a top level view of the radionuclide transport model. The closure cap placed over the waste disposal unit is modeled indirectly through specification of the infiltration flow into the waste zone. Infiltration representative of a geo-synthetic closure cap was used in the risk modeling. The waste zone is modeled as shown in Figure 3-2 by a series of 11 vertical cells. For purposes of modeling the waste tanks following closure, the first 10 cells were assumed to be clean grout and the contamination was assumed to be concentrated in the last cell which covered the bottom of the tank and was one foot thick. This is consistent with the approach used in the Tank Farm PAs. For Saltstone modeling, the contamination was assumed to be uniformly distributed throughout the waste zone. The concrete floor of the waste tank or Saltstone vault and supporting mats were modeled in the barrier container using a vertical series of five cells.

The unsaturated zone below the waste was modeled as 20 vertical cells having properties consistent with SRS vadose zone soils. At the bottom of the vadose zone, the flow is distributed to a set of ten horizontal cells representing discharge into the aquifer region below the waste. When the bottom of the waste disposal unit is below the water table, as indicated by a negative vadose zone length, the transport of radionuclides from the waste layer to the aquifer bypasses the vadose zone and directly enters the aquifer.

As shown in Figure 3-1, the aquifer is divided into two separate zones. These zones are further subdivided into regions as shown in Figure 3-3. The two zones can have different flow lengths which are defined through the model input. The number of cells in each zone is fixed at the values shown in Figure 3-3. The model of Zone 1 is shown in Figure 3-4. The first 10 cells of Zone 1 are used to distribute the incoming flow using distribution fractions defined through the model input. Transport from this region to the 100 m well is modeled using a GoldSim^{TM} pipe model equivalent to 10 cells. Cells in Zone 1 contain only sandy soil.

Transport beyond the well from Zone 1 to Zone 2 is represented by another pipe model with 80 cells. The first 50 cells of Zone 2 can have either sandy soil, clayey soil, or a mixture of soils to represent an aquitard region. The last 50 cells in Zone 2 are all sandy soil. The dose calculations presented in this report are based on the concentration of radionuclides in the aquifer at the 100 m well location. The part of the aquifer model beyond the well therefore has no impact on the current dose calculations but was retained in the model to allow calculation of a CA dose using stream or river radionuclide concentrations.

On a fast workstation, this simple model takes on the order of one to two minutes to run a transport and dose calculation. The model can be used to rapidly evaluate many scenarios and, in particular, is applicable to performing sensitivity and uncertainty analysis.

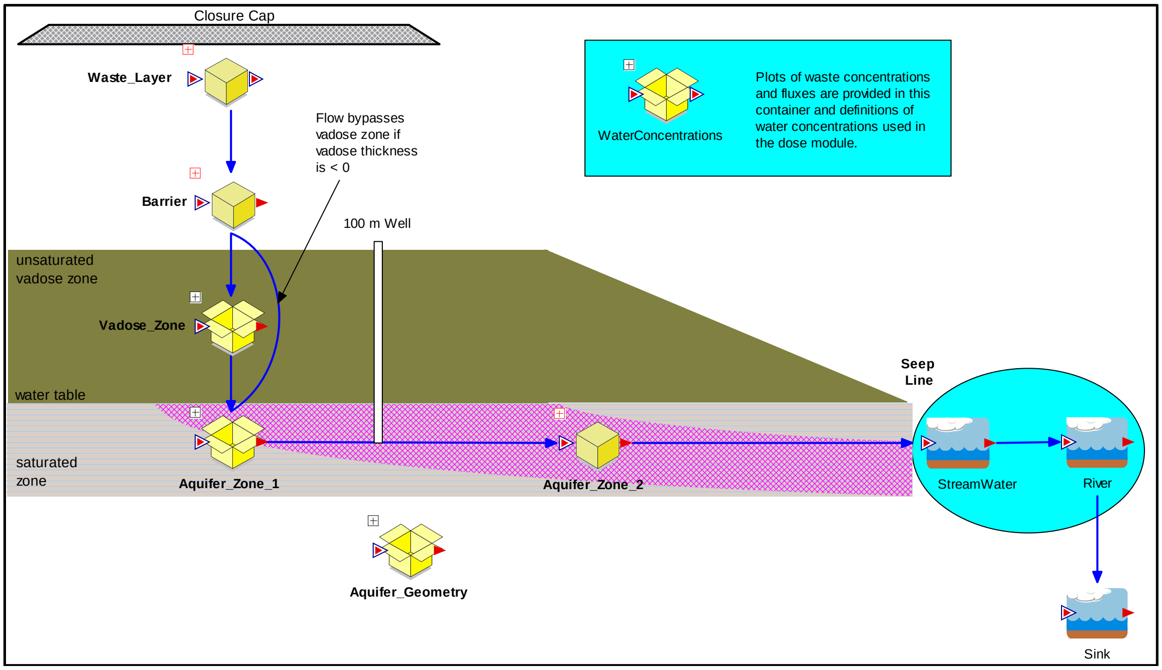
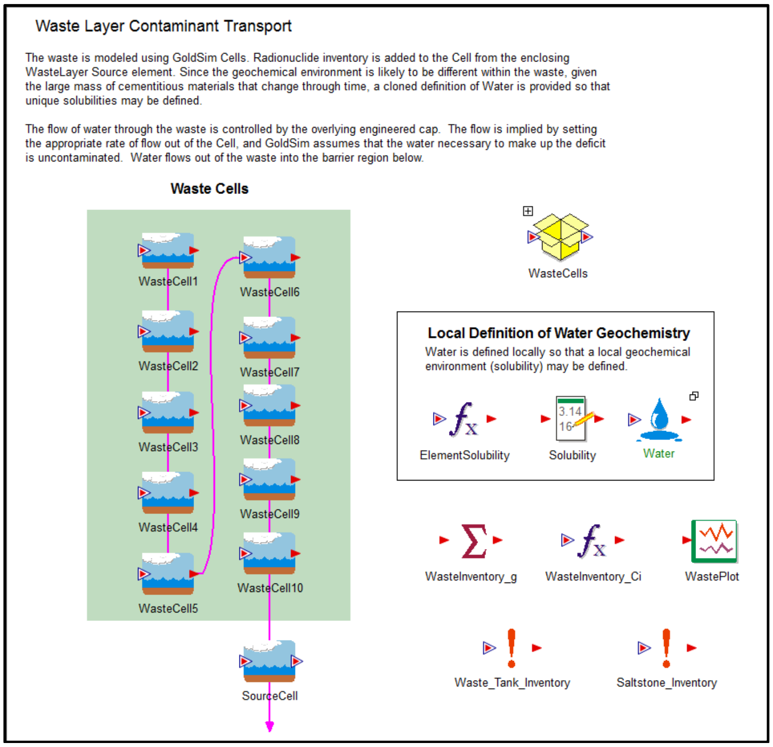


Figure 3-1. GoldSim Model of Radionuclide Transport in Waste Characterization Model



Waste Layer Contaminant Transport

The waste is modeled using GoldSim Cells. Radionuclide inventory is added to the Cell from the enclosing WasteLayer Source element. Since the geochemical environment is likely to be different within the waste, given the large mass of cementitious materials that change through time, a cloned definition of Water is provided so that unique solubilities may be defined.

The flow of water through the waste is controlled by the overlying engineered cap. The flow is implied by setting the appropriate rate of flow out of the Cell, and GoldSim assumes that the water necessary to make up the deficit is uncontaminated. Water flows out of the waste into the barrier region below.

Local Definition of Water Geochemistry

Water is defined locally so that a local geochemical environment (solubility) may be defined.

Figure 3-2. GoldSim Model of Waste Disposal Facility

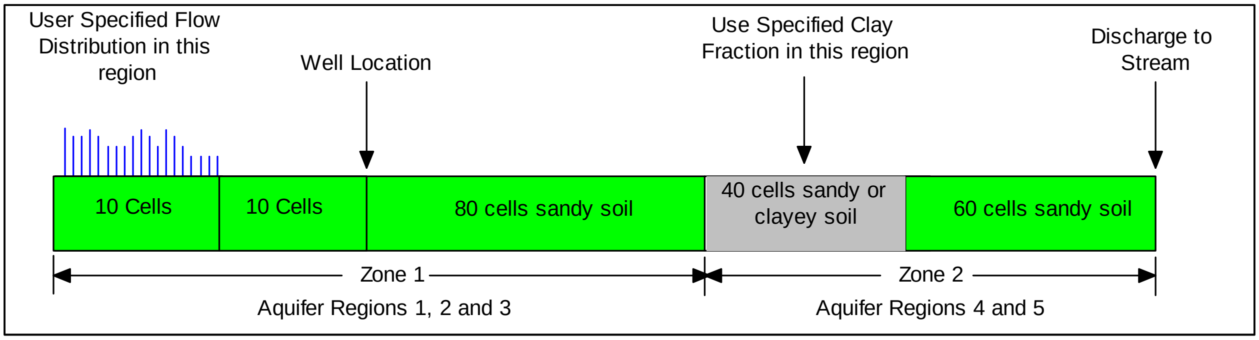
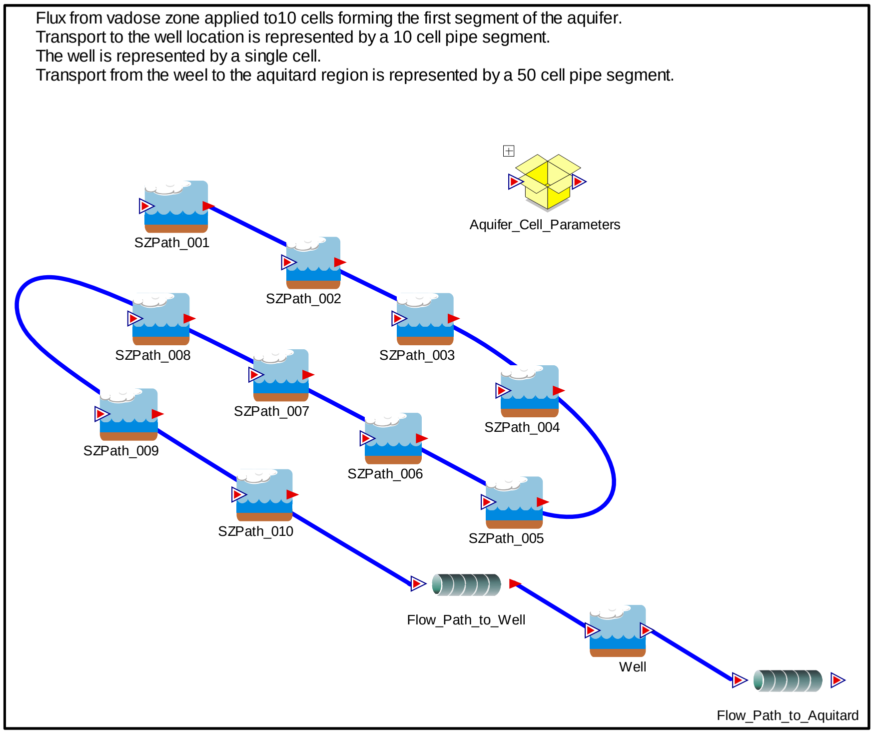


Figure 3-3. Schematic Representation of Aquifer Transport Model



Flux from vadose zone applied to10 cells forming the first segment of the aquifer.
Transport to the well location is represented by a 10 cell pipe segment.
The well is represented by a single cell.
Transport from the weel to the aquitard region is represented by a 50 cell pipe segment.

Figure 3-4. Representation of First Aquifer Transport Zone in GoldSim Model

3.1.2 Dose Calculations

For this preliminary scoping study, the dose to a member of the public from use of water obtained from a well located 100 m from the boundary of the waste disposal facility was assumed to be the all-pathways dose to a resident farmer. Doses to a typical member of the public considered in the analysis included:

1) **Ingestion Dose**
    a. Water
    b. Vegetables
    c. Meat
    d. Milk
    e. Garden Soil

2) **Inhalation Dose**
    a. Garden dust
    b. Irrigation water
    c. Shower water
3) **Dose from Direct Exposure**
    a. Garden soil

EPA groundwater protection limits for exposure to alpha radiation, beta-gamma radiation, radium and uranium were included in the model but were not considered in this preliminary risk analysis.

The dose calculations used updated dose parameters,[4,5] updated radionuclide data (ICRP-107), and employed somewhat revised dose equations from those used in prior analyses (e.g. Phifer et al. SRNL-STI-2009-00424 [6]). While this revised dose methodology has not yet been published, it should closely reflect the analysis that would be used in future SRNL PA, CA and SA calculations. The dose calculations performed for this study were based on the 63 radionuclides used in the SRS CA which are listed in Tables 3-1, 3-2 and 3-3 of the following Section.

3.1.3 Waste Tank Risk Assessment

3.1.3.1 Waste Tank Closure Inventories

To perform the proposed screening calculation it is necessary to assume a realistic but conservative source inventory. For the SRS waste tanks, attempts have already been made to estimate closure inventories for previous PA, CA and SA calculations. The current study compared the assumed inventories used in the Tank Farm PAs[7,8] and the SRS CA[9] to the actual residual inventory estimated from existing characterization studies. This preliminary study was limited to evaluating waste tanks 5-F, 6-F, 18-F, 19-F, 16-H and 12-H. Tanks 18-F, 19-F, 5-F, and 6-F have already been characterized and closed. Tank 16-H has been characterized and is in the process of being closed. Tank 12-H is currently being characterized and is the next tank scheduled for closure after 16-H. The inventory comparisons for the 63 radionuclides used in the SRS CA are shown in Tables 3-1, 3-2 and 3-3. Cells in the PA and CA inventory columns highlighted in yellow indicate assumed inventories that were lower than the value estimated from waste characterization data. The characterization data in these tables used 95% confidence limit values and upper bounds for non-detected species as best estimates (BE) of the residual inventory. Nominal estimates of residual volume and waste density were used to convert concentrations to total Curie inventory. The residual volumes and densities used to calculate total Curies for each tank are listed in Table 3-4.

Tanks 5-F and 6-F were the first tanks evaluated. Characterization data was obtained from Oji et al.,[10,11] Shine,[12] and SRR.[13] As shown in Table 3-1, these initial results were encouraging in that the CA estimated residual inventory appeared to fall relatively close to most characterization values while the PA inventory appeared to be in general very conservative. For a few radionuclides (Cf-249, Cm-247 and Cm-248) the CA residual inventory was significantly lower than the value based on sample characterization. However, it was thought that with perhaps some adjustment to the CA inventory it could serve as a reasonable basis for risk evaluation.

This trend did not continue for the other waste tanks evaluated. As shown in Tables 3-2 and 3-3, for many radionuclides, the residual inventory assumed in the CA falls below a reasonably conservative estimate based on waste characterization measurements. In addition, for Tanks 18-F, 19-F and 16-H, the inventory for many of the radionuclides assumed in the PA also fall below the measurement values. Characterization data for Tank 18-F was taken from Oji et al.,[14,15] Hay et al.,[16] Harris,[17] and SRR.[18]

Characterization data for Tank 19-F was obtained from Oji et al.,[19] Harris,[20] and SRR.[21] Characterization data for Tank 16-H was obtained from Oji et al.[22]

When increasing the PA inventories by a factor of 100, only I-129 and Pa-231 for Tank 16-H remain below the values calculated using characterization data. However, in many cases, these higher inventory values are substantially greater than corresponding characterization values. For example, the residual Pu-238 inventory in Tank 16-H (tank and annulus) is estimated to be 37.7 Curies from characterization data while the inventory used in the PA analysis is 290 Curies which is already a factor of 7.7 larger. Increasing the PA inventory by a factor of 100 gives an extremely conservative estimate of the residual Pu-238. On the other hand, increasing the CA inventories by a factor of 100 still left many values below that calculated from the characterization data. Performing uncertainty analysis using a distribution about the best estimate expected inventory would allow determination of inventory levels where the risk factor becomes insignificant.

From this analysis, it was concluded that no ideal method exists for estimating the residual inventory in the SRS waste tanks prior to characterization. Therefore, it was decided to use the characterization based inventory to determine which radionuclides contribute the most risk for dose to an offsite member of the public. This approach would appear to preclude applying the risk screening to an uncharacterized tank. However, as evidenced by the inventories assumed for the tank farm PAs, inventories in the SRS waste tanks are relatively consistent so using information from the five tanks that have had residual waste material characterized should provide a good indication of which radionuclides will in general impact the dose risk from other tanks. Since the residual waste in Tank 12-H has not yet been characterized this tank was analyzed using the PA inventory to determine what difference this assumption might make.

Table 3-1. Residual Radionuclide Inventory in Tanks 5-F and 6-F

	Tank 5-F			Tank 6-F		
Isotopes	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**
Ac-227	1.00E-03	2.63E-07	4.18E-06	1.00E-03	2.05E-07	6.78E-07
Ag-108m	0	0	0	0	0	0
Al-26	1.00E+00	9.51E-04	1.55E-04	1.00E+00	1.24E-03	0
Am-241	6.00E+02	4.91E+01	5.82E-01	6.00E+02	5.95E+01	1.29E+00
Am-242m	1.00E+00	5.81E-02	1.44E-03	1.00E+00	7.07E-02	1.88E-03
Am-243	1.40E+00	1.65E-04	4.45E-03	1.40E+00	1.36E-01	3.03E-02
Bi-210m	0	0	0	0	0	0
C-14	1.00E+00	1.38E-03	2.71E-05	1.00E+00	1.13E-03	3.12E-04
Ca-41	0	0	0	0	0	0
Cf-249	1.00E+00	3.55E-19	1.50E-04	1.00E+00	4.26E-19	1.47E-04
Cf-251	0	0	4.03E-04	0	0	2.23E-04
Cl-36	1.00E-03	0	6.72E-05	1.00E-03	0	0
Cm-243	1.00E+00	1.31E-03	1.12E-02	1.00E+00	1.71E-03	2.14E-02
Cm-244	1.20E+02	5.61E-03	2.59E-02	1.20E+02	5.20E+00	1.01E+00
Cm-245	1.00E+00	1.08E-08	1.20E-05	1.00E+00	1.28E-08	1.02E-04
Cm-246	0	0	2.57E-05	0	0	5.49E-04
Cm-247	1.00E-03	5.83E-17	4.32E-09	1.00E-03	6.94E-17	4.07E-09
Cm-248	1.00E-03	1.34E-17	4.57E-07	1.00E-03	1.60E-17	3.53E-07
Co-60	1.80E+01	9.76E-01	5.65E-02	1.80E+01	1.79E+00	1.31E-01
Cs-135	1.00E+00	4.40E-03	1.84E-05	1.00E+00	5.24E-03	4.17E-05
Cs-137	9.20E+03	7.07E+02	3.55E+00	9.20E+03	9.18E+02	8.19E+00
Eu-152	1.90E+01	1.30E+00	1.68E-03	1.90E+01	1.87E+00	2.82E-03
Eu-154	1.30E+02	8.53E+00	2.47E-01	1.30E+02	1.34E+01	3.25E-01
Eu-155	0	2.32E+00	4.68E-02	0	4.36E+00	6.04E-02
Gd-152	0	0	0	0	0	0
H-3	1.00E+00	2.72E-02	1.46E-04	1.00E+00	2.72E-02	3.20E-04
I-129	1.00E-03	3.13E-05	6.87E-06	1.00E-03	3.73E-05	3.02E-06
K-40	1.00E-03	0	4.10E-05	1.00E-03	0	0
Lu-174	0	0	0	0	0	0
Mo-93	0	0	0	0	0	0
Nb-93m	1.00E-03	0	0	1.00E-03	0	0
Nb-94	1.00E+00	2.84E-04	2.29E-05	1.00E+00	3.39E-04	3.17E-05
Ni-59	6.30E+00	5.43E-01	5.01E-02	6.30E+00	6.31E-01	7.47E-02
Ni-63	4.90E+02	4.09E+01	2.77E+00	4.90E+02	4.89E+01	5.53E+00
Np-237	2.30E-01	2.32E-02	2.19E-04	2.30E-01	5.86E-03	4.71E-04
Pa-231	1.00E-03	5.55E-07	2.82E-06	1.00E-03	4.32E-07	1.51E-05
Pb-210	0	0	0	0	0	0
Pd-107	1.00E-03	0	4.12E-05	1.00E-03	0	0
Pt-193	1.00E-03	0	1.25E-05	1.00E-03	0	0
Pu-238	1.40E+02	1.42E+01	2.20E-02	1.40E+02	7.17E-01	5.14E-02
Pu-239	3.20E+01	2.67E+00	7.09E-02	3.20E+01	1.73E+00	4.48E-02
Pu-240	7.20E+00	6.36E-01	1.64E-02	7.20E+00	6.00E-01	2.79E-02
Pu-241	3.20E+01	2.03E+00	1.09E-01	3.20E+01	3.21E+00	1.04E-01
Pu-242	1.00E+00	1.88E-04	3.27E-06	1.00E+00	1.18E-03	3.34E-05
Pu-244	1.00E-03	8.59E-07	9.42E-09	1.00E-03	5.41E-06	1.16E-08
Ra-226	1.00E-03	1.99E-02	7.81E-05	1.00E-03	2.45E-02	6.42E-05
Ra-228	0	0	0	0	0	0
Se-79	4.50E+00	3.80E-01	1.06E-04	4.50E+00	4.52E-01	2.99E-04
Sm-147	1.20E+04	0	6.49E+00	1.20E+04	0	3.10E+00
Sm-151	0	1.00E+03	0	0	1.23E+03	0
Sn-126	8.40E-01	7.06E-01	9.42E-03	8.40E-01	8.41E-01	1.18E-02
Sr-90	1.30E+05	9.82E+03	9.98E+01	1.30E+05	1.28E+04	2.45E+02
Tc-99	7.90E+01	6.57E+00	8.70E-05	7.90E+01	7.82E+00	1.73E-03
Th-229	2.40E-01	2.44E-02	2.45E-07	2.40E-01	6.17E-03	1.50E-06
Th-230	1.00E-03	1.98E-02	1.80E-05	1.00E-03	2.44E-02	1.47E-05
Th-232	0	0	0	0	0	0
U-232	1.00E+00	2.57E-04	2.77E-07	1.00E+00	3.15E-04	1.19E-06
U-233	2.30E-01	2.33E-02	6.05E-05	2.30E-01	5.88E-03	5.13E-05
U-234	1.70E-01	1.37E-02	3.76E-05	1.70E-01	1.69E-02	1.46E-04
U-235	5.80E-03	5.79E-04	1.69E-06	5.80E-03	4.52E-04	6.76E-06
U-236	1.00E+00	2.83E-04	2.07E-06	1.00E+00	3.46E-04	8.30E-06
U-238	1.70E-01	1.37E-02	3.92E-05	1.70E-01	1.69E-02	2.50E-04
Zr-93	1.00E-03	0	2.51E-02	1.00E-03	0	2.21E-02

Table 3-2. Residual Radionuclide Inventory in Tanks 18-F and 19-F

	Tank 18-F			Tank 19-F		
Isotopes	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**
Ac-227	1.00E-03	2.45E-07	1.79E-04	1.00E-03	4.10E-08	1.33E-05
Ag-108m	0	0	0	0	0	0
Al-26	1.00E+00	4.28E-03	1.56E-04	1.00E+00	1.71E-02	3.83E-05
Am-241	8.20E+01	1.86E+01	9.91E+01	2.30E+00	2.90E+00	2.58E+00
Am-242m	1.00E+00	1.87E-01	3.25E-02	1.00E+00	4.52E-01	2.66E-04
Am-243	1.00E-01	1.55E-06	1.95E+00	1.00E-01	4.45E-08	6.83E-03
Bi-210m	0	0	0	0	0	0
C-14	1.00E+00	1.41E-02	7.31E-01	1.00E+00	2.83E-02	4.08E+00
Ca-41	0	0	0	0	0	0
Cf-249	1.00E+00	3.40E-21	1.95E-03	1.00E+00	9.74E-23	5.33E-04
Cf-251	0	0	0	0	0	0
Cl-36	1.00E-03	0	2.27E-04	1.00E-03	0	9.16E-05
Cm-243	1.00E+00	1.63E-05	1.95E-02	1.00E+00	4.49E-07	2.17E-03
Cm-244	1.00E+02	2.28E+01	1.17E+02	1.00E+00	3.91E-04	4.08E-01
Cm-245	1.00E+00	5.48E-10	9.58E-03	1.00E+00	5.01E-10	1.58E-03
Cm-246	0	0	0	0	0	0
Cm-247	1.00E-03	5.48E-19	1.79E-06	1.00E-03	1.57E-20	1.33E-06
Cm-248	1.00E-03	1.27E-19	7.79E-05	1.00E-03	3.62E-21	5.83E-05
Co-60	1.00E+00	1.27E-01	5.36E-01	1.00E+00	1.75E-01	4.41E-02
Cs-135	1.00E+00	2.21E-04	6.17E+03	1.00E+00	2.02E-04	2.50E-02
Cs-137	9.70E+03	2.24E+03	6.50E-03	6.50E+03	8.30E+03	5.50E+03
Eu-152	1.00E+00	2.15E-02	3.90E-01	1.00E+00	5.70E-04	2.91E-04
Eu-154	3.20E+00	7.34E-01	0.00E+00	1.00E+00	9.11E-01	8.33E-03
Eu-155	0	6.15E-02	0	0	1.41E-03	0
Gd-152	0	0	1.17E-02	0	0	0
H-3	1.00E+00	6.15E-03	2.11E-04	1.00E+00	3.51E-02	4.50E-03
I-129	1.00E-03	1.62E-06	1.32E-02	1.00E-03	1.26E-05	2.25E-04
K-40	1.00E-03	0	0	1.00E-03	0	1.08E-03
Lu-174	0	0	0	0	0	0
Mo-93	0	0	6.17E-02	0	0	0
Nb-93m	1.00E-03	0	4.55E-04	1.00E-03	0	1.75E-02
Nb-94	1.00E-03	1.01E-05	2.60E-01	1.00E-03	9.24E-06	9.99E-05
Ni-59	1.00E+00	2.30E-01	1.38E+01	1.00E+00	2.11E-01	3.50E-04
Ni-63	8.20E+01	1.81E+01	1.14E-01	1.40E+01	1.72E+01	1.42E-02
Np-237	2.40E-01	3.81E-02	3.74E-02	2.20E-03	2.84E-03	1.42E-03
Pa-231	1.00E-03	1.15E-06	0	1.00E-03	2.01E-07	6.91E-05
Pb-210	0	0	1.01E-01	0	0	0
Pd-107	1.00E-03	0	3.41E-03	1.00E-03	0	2.00E-01
Pt-193	1.00E-03	0	7.79E+01	1.00E+00	0	1.67E-03
Pu-238	7.00E+01	1.56E+01	2.11E+02	4.40E+00	5.39E+00	3.50E+00
Pu-239	1.60E+02	3.72E+01	4.71E+01	6.40E+00	7.92E+00	3.91E+00
Pu-240	4.90E+01	2.15E+01	2.11E+02	2.30E+00	2.82E+00	9.99E-01
Pu-241	1.30E+02	2.89E+01	9.58E-03	4.60E+00	5.89E+00	6.25E+00
Pu-242	1.00E+00	2.60E-01	2.76E-06	1.00E+00	9.46E-02	1.67E-03
Pu-244	1.00E-03	8.56E-05	2.76E-03	1.00E-03	4.23E-04	4.58E-06
Ra-226	1.90E-03	1.23E-07	0	1.10E-03	2.68E-08	4.16E-03
Ra-228	0	0	3.90E-04	0	0	0
Se-79	1.00E+00	1.28E-02	0	1.00E+00	1.50E-02	4.58E-04
Sm-147	0	0	3.25E+01	0	0	0
Sm-151	4.60E+01	1.02E+01	1.06E-02	1.00E+00	2.89E-01	1.67E-01
Sn-126	1.60E-01	3.53E-02	9.91E+02	2.60E-02	3.24E-02	3.33E-03
Sr-90	1.10E+03	2.38E+02	7.31E-01	5.20E+00	6.60E+00	8.33E+00
Tc-99	1.00E+00	3.72E-01	7.15E-04	1.40E+00	1.72E+00	3.83E-01
Th-229	2.60E-03	5.72E-04	1.62E-03	1.00E-03	6.64E-04	2.00E-04
Th-230	1.90E-03	1.51E-05	0	1.10E-03	3.29E-06	1.17E-04
Th-232	0	0	6.17E-04	0	0	0
U-232	1.00E+00	4.69E-05	2.11E-02	1.00E+00	4.50E-05	1.08E-04
U-233	1.10E+00	3.85E-01	2.27E-01	1.90E-01	2.36E-01	4.08E-03
U-234	3.80E-01	1.72E-01	9.09E-03	1.10E-02	1.33E-02	4.75E-03
U-235	8.40E-03	1.93E-03	7.63E-03	2.60E-04	3.22E-04	1.67E-04
U-236	1.00E+00	3.10E-03	2.27E-01	1.00E+00	7.35E-04	2.50E-04
U-238	2.20E-01	4.92E-02	6.17E-02	8.70E-03	1.09E-02	5.41E-03
Zr-93	1.00E-03	0	1.79E-04	1.00E-03	0	1.75E-02

Table 3-3. Residual Radionuclide Inventory in Tanks 12-H and 16-H

	Tank 12-H			Tank 16-H		
Isotopes	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**	**PA (Ci)**	**CA (Ci)**	**95% BE (Ci)**
Ac-227	1.00E+00	6.20E-09	Not yet determined	1.00E+00	9.20E-09	0
Ag-108m	0	0	Not yet determined	0	0	0
Al-26	1.00E+00	4.20E-04	Not yet determined	1.00E+00	5.80E-04	0
Am-241	7.00E+02	1.50E+01	Not yet determined	8.10E+01	1.90E+01	8.69E+00
Am-242m	1.00E+00	1.20E-02	Not yet determined	1.00E+00	1.60E-02	8.48E-02
Am-243	3.00E+00	7.10E-02	Not yet determined	1.00E+00	9.90E-02	1.59E-01
Bi-210m	0	0	Not yet determined	0	0	0
C-14	1.00E+00	2.30E-04	Not yet determined	1.00E+00	3.30E-04	7.39E-03
Ca-41	0	0	Not yet determined	0	0	0
Cf-249	1.00E+00	3.00E-12	Not yet determined	1.00E+00	4.30E-12	1.96E-02
Cf-251	1.00E+00	1.10E-13	Not yet determined	1.00E+00	1.50E-13	5.37E-02
Cl-36	2.10E-03	5.40E-02	Not yet determined	5.30E-04	7.40E-02	6.15E-03
Cm-243	1.00E+00	1.60E-03	Not yet determined	1.00E+00	2.20E-03	6.37E-02
Cm-244	2.00E+01	1.50E-01	Not yet determined	2.40E+00	1.20E-01	1.00E+01
Cm-245	1.00E+00	7.40E-06	Not yet determined	1.00E+00	1.20E-05	1.67E-03
Cm-246	0	0	Not yet determined	0	0	0
Cm-247	1.00E+00	5.50E-13	Not yet determined	1.00E+00	7.70E-13	5.10E-07
Cm-248	1.00E+00	5.80E-13	Not yet determined	1.00E+00	8.10E-13	3.04E-05
Co-60	1.00E+00	2.30E-01	Not yet determined	1.00E+00	2.80E-01	2.67E-02
Cs-135	5.40E-03	1.60E-03	Not yet determined	9.90E-04	2.30E-03	1.98E-02
Cs-137	7.90E+02	2.10E+02	Not yet determined	1.30E+02	2.90E+02	5.66E+03
Eu-152	2.10E+01	4.00E-01	Not yet determined	1.00E+00	5.70E-01	0
Eu-154	2.90E+02	5.70E+00	Not yet determined	3.30E+01	7.70E+00	8.53E+00
Eu-155	0	4.40E-01	Not yet determined	0	6.10E-01	0
Gd-152	0	0	Not yet determined	0	0	0
H-3	1.00E+00	1.60E-02	Not yet determined	1.00E+00	2.00E-02	0
I-129	2.80E-04	8.60E-06	Not yet determined	5.30E-05	1.20E-05	1.18E-02
K-40	1.10E-03	6.80E-03	Not yet determined	2.60E-04	9.50E-03	1.48E-02
Lu-174	0	0	Not yet determined	0	0	0
Mo-93	0	0	Not yet determined	0	0	0
Nb-93m	0	1.60E+00	Not yet determined	0	1.90E+00	0
Nb-94	1.10E-01	4.40E-05	Not yet determined	2.60E-02	5.10E-05	2.14E-02
Ni-59	8.60E+00	2.50E-01	Not yet determined	1.00E+00	3.70E-01	3.22E+00
Ni-63	6.30E+02	1.80E+01	Not yet determined	1.10E+02	2.60E+01	2.50E+01
Np-237	2.10E-01	7.10E-03	Not yet determined	2.20E-02	5.20E-03	2.13E-02
Pa-231	2.10E-03	3.80E-08	Not yet determined	5.30E-04	5.70E-08	1.04E-01
Pb-210	0	0	Not yet determined	0	0	0
Pd-107	2.10E-01	3.90E-02	Not yet determined	5.30E-02	5.70E-02	0
Pt-193	2.10E-01	7.60E+00	Not yet determined	5.30E-02	1.10E+01	0
Pu-238	6.50E+03	1.00E+02	Not yet determined	2.90E+02	6.70E+01	3.77E+01
Pu-239	8.00E+01	2.10E+00	Not yet determined	7.70E+00	1.80E+00	4.80E+00
Pu-240	5.00E+01	1.10E+00	Not yet determined	3.70E+00	8.60E-01	2.18E+00
Pu-241	7.60E+02	7.70E+00	Not yet determined	2.00E+01	4.70E+00	1.48E+01
Pu-242	1.00E+00	1.70E-03	Not yet determined	1.00E+00	6.50E-04	9.56E-04
Pu-244	1.00E+00	7.60E-06	Not yet determined	1.00E+00	3.00E-06	2.49E-06
Ra-226	2.10E-02	7.20E-09	Not yet determined	5.30E-03	1.20E-08	1.46E-02
Ra-228	2.10E+00	5.90E-05	Not yet determined	5.30E-01	8.00E-05	0
Se-79	4.80E+00	1.40E-01	Not yet determined	1.00E+00	2.00E-01	0
Sm-147	0	0	Not yet determined	0	0	0
Sm-151	1.10E+04	2.50E+02	Not yet determined	1.80E+03	3.50E+02	0
Sn-126	4.60E+00	1.40E-01	Not yet determined	1.00E+00	1.90E-01	0
Sr-90	1.40E+04	3.60E+03	Not yet determined	2.20E+03	5.10E+03	2.58E+04
Tc-99	8.10E+00	2.40E+00	Not yet determined	1.50E+00	3.40E+00	3.08E+00
Th-229	2.10E-03	9.80E-05	Not yet determined	5.30E-04	5.80E-05	0
Th-230	2.10E-02	8.90E-07	Not yet determined	5.30E-03	1.50E-06	4.13E-03
Th-232	2.90E-02	1.20E-03	Not yet determined	5.30E-03	1.70E-03	0
U-232	2.10E-03	9.40E-06	Not yet determined	5.30E-04	1.40E-06	0
U-233	5.90E-01	3.40E-02	Not yet determined	8.70E-02	2.00E-02	4.51E-02
U-234	9.60E-02	3.20E-03	Not yet determined	2.40E-02	5.60E-03	2.07E-02
U-235	2.10E-02	6.10E-05	Not yet determined	5.30E-03	9.00E-05	1.76E-04
U-236	2.10E-02	2.60E-04	Not yet determined	5.30E-03	5.30E-04	4.63E-04
U-238	2.90E-02	6.10E-04	Not yet determined	5.30E-04	2.10E-06	7.89E-04
Zr-93	4.00E-01	1.60E+00	Not yet determined	6.30E-02	1.90E+00	1.38E+00

Table 3-4. Residual Waste Volume and Density used to Convert Concentrations to Total Curies

Tank	Residual Volume (gallons)	Density kg/L	Reference
5-F	1900	1.11	SRNL-STI-2012-00034 [10]
6-F	3000	1.30	SRR-LWE-2011-00245 [13] SRNL-STI-2012-00365 [11]
18-F	3900	1.10	SRR-CWDA-2010-00117 [18]
19-F	2000	1.10	SRR-CWDA-2010-00118 [21]
16-H	330/Tank 1900/Annulus	1.28/Tank 0.97/Annulus	U-ESR-H-00113 [23] SRNL-STI-2014-00321 [22]

3.1.3.2 Waste Tank Model Input

Transport parameters used in the GoldSim™ model for the waste tank analysis are listed in Table 3-5. For this scoping calculation, it was assumed that the residual inventory was placed in the waste in 2010 and the cap was placed over the grouted tanks in 2025. It was further assumed that the cap failed 500 years after placement. The Type 2 cap entered in Table 3-5 specifies a geo-synthetic material. The waste thickness is the height of the grout filled tank and the sand fraction of -1 for the waste identifies the waste material as concrete. As noted above, the radioactive contamination in waste tanks was placed in a one foot thick layer at the base of the waste zone. The barrier thickness is the thickness of the tank floor and base mat and setting the volume fraction of soil in the barrier to zero specifies concrete material properties.

The vadose zone thickness and distance from the tank to the 100 m well were obtained from the Tank Farm PAs.[7,8] The only aquifer parameter that influences the calculations reported in this work is the aquifer pore velocity which was obtained from the CA.[24] The source area was calculated as the tank cross-sectional area and the last 10 fractions are set to give a circular distribution of the contamination as it enters the aquifer as illustrated in Figure 3-5. Other parameters in the Table were taken from the SRS CA.[3]

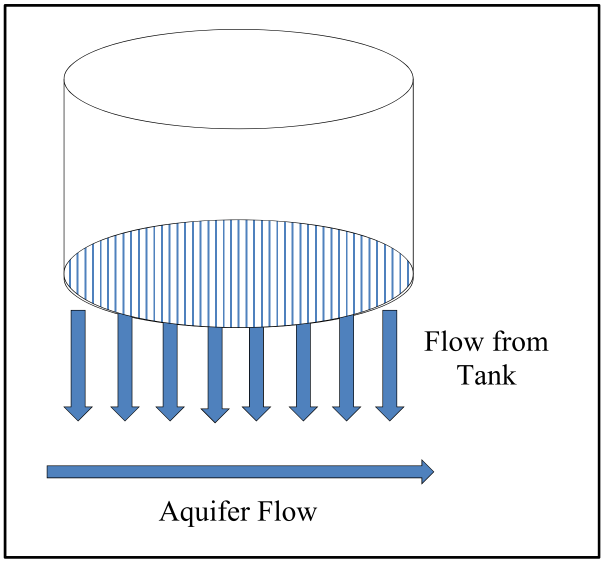


Figure 3-5. Illustration of Flow from Waste Tank or Vadose Zone to Aquifer

Table 3-5. GoldSim Model Parameters for Waste Tanks

	FTF Type I Tank 5	FTF Type I Tank 6	FTF Type IV Tank 18	FTF Type IV Tank 19	HTF Type I Tank 12	HTF Type II Tank 16
Cap Placement Year	2025	2025	2025	2025	2025	2025
Cap Time to Failure (yr)	500	500	500	500	500	500
Cap Type	2	2	2	2	2	2
Waste Thickness (m)	7.47	7.47	10.52	10.52	7.47	8.18
Fraction Sand in Waste	-1	-1	-1	-1	-1	-1
Barrier Thickness (m)	0.86	0.86	0.18	0.18	0.86	1.22
Volume Fraction Soil in Barrier	0	0	0	0	0	0
Vadose Zone Thickness (m)	3.23	3.11	0.64	0.52	-35.5	-6.60
Distance to Well (m)	264	300	132	127	284	574
Vadose Zone Clay (m)	0	0	0	0	0	0
Total Aquifer Path Length (m)	1765.8	1765.8	1627.3	1627.3	2879.7	3300.9
Length of First Aquifer Zone (m)	1212.6	1212.6	1156.2	1156.2	2031.3	2302.1
Length of Aquitard Zone (m)	189.5	189.5	124.3	124.3	239.0	308.2
Aquifer Pore Velocity (m/yr)	3.98	3.98	2.98	2.98	6.53	10.90
Aquitard Pore Velocity (m/yr)	2.96	2.96	2.06	2.06	3.58	5.87
Stream ID	1	1	1	1	1	1
Release Mechanism	1	1	1	1	1	1
Source Release Rate (1/yr)	0	0	0	0	0	0
Source Area (m^2)	410.43	410.43	527.18	527.18	410.43	527.18
Number of Aquitard Clay Cells	34	34	26	26	28	30
Footprint Fraction (1)	0.0520	0.0520	0.0520	0.0520	0.0520	0.0520
Footprint Fraction (2)	0.0903	0.0903	0.0903	0.0903	0.0903	0.0903
Footprint Fraction (3)	0.1099	0.1099	0.1099	0.1099	0.1099	0.1099
Footprint Fraction (4)	0.1212	0.1212	0.1212	0.1212	0.1212	0.1212
Footprint Fraction (5)	0.1265	0.1265	0.1265	0.1265	0.1265	0.1265
Footprint Fraction (6)	0.1265	0.1265	0.1265	0.1265	0.1265	0.1265
Footprint Fraction (7)	0.1212	0.1212	0.1212	0.1212	0.1212	0.1212
Footprint Fraction (8)	0.1099	0.1099	0.1099	0.1099	0.1099	0.1099
Footprint Fraction (9)	0.0903	0.0903	0.0903	0.0903	0.0903	0.0903
Footprint Fraction (10)	0.0520	0.0520	0.0520	0.0520	0.0520	0.0520

3.1.4 Saltstone Risk Assessment

The risk screening model was applied to evaluate the Saltstone Disposal Facility (SDF) in a similar manner to that described above for the waste tanks.

3.1.4.1 Saltstone Closure Inventories

A diagram of the Saltstone disposal facility is shown on the left hand side of Figure 3-6 at the end of this section. Saltstone disposal began with rectangular Vaults 1 and 4 and progressed to include six small Saltstone Disposal Cells (SDC) designated as SDC-2A, SDC-2B, SDC-3A, SDC-3B, SDC-5A and SDC-5B. While it was initially planned to construct 64 SDCs, it is currently planned to complete Saltstone disposal using six larger Saltstone Disposal Units (SDU) designated as SDU-6 through SDU-12 as shown in Figure 3-6. The SDUs are designed to hold approximately ten times the waste material contained in a single SDC. Saltstone disposal in Vault 1 and Vault 4 is complete.

The right hand side of Figure 3-6 shows aquifer flow paths from the SDF disposal sites across the 100 m perimeter. As this diagram shows, in general, radionuclide transport from the upper group of disposal units splits into two paths while transport from the lower group of units follows the same general path. It is expected that the lower group of disposal sites will create the highest dose to an offsite MOP. The maximum dose at the 100 m boundary will be the cumulative dose from radionuclide concentrations in ground water originating from all of the units in the lower group. However, for the purposes of this study, the four different types of Saltstone disposal units were evaluated separately. SDC-2B and SDU-11 were chosen for this analysis because they appear to have the shortest flow paths to the 100 m perimeter.

The inventories for the disposal vaults and cells used in the Saltstone PA and the SRS CA were obtained from Dean[25] as reported in the Saltstone PA. Table 3-6 lists the estimated inventories for Vaults 1 and 4 and for SDC-2B and SDU-11. In the Saltstone PA all of the SDCs and SDUs are assumed to have the same inventory on a unit volume basis. Vaults 1 and 4 have significantly different inventories from each other and from the other disposal units presumably because they are based on estimates (or measurements) of the actual waste material disposed in these units. For example, Vault 4 contains a significant amount of Sr-90, Cs-135, Cs-137, and plutonium isotopes, whereas very small amounts of these isotopes are present in the other disposal units, because they are now being stripped from salt waste (via sorption and extraction processing treatments) and sent to high-level waste disposal at the Defense Waste Processing Facility.

Table 3-6. Radionuclide Inventory in Saltstone Disposal Facilities

Isotopes	Vault 1	Vault 4	SDC-2B	SDU-11
Ac-227	0	1.60E-05	1.70E-07	1.64E-06
Ag-108m	0	0	0	0
Al-26	0	3.40E-01	1.90E-01	1.84E+00
Am-241	4.70E-04	5.60E+00	1.40E+00	1.35E+01
Am-242m	0	2.80E-03	5.90E-04	5.70E-03
Am-243	0	1.60E-02	3.70E-02	3.58E-01
Bi-210m	0	0	0	0
C-14	1.30E+00	2.70E+01	2.00E+00	1.93E+01
Ca-41	0	0	0	0
Cf-249	0	6.50E-13	6.70E-13	6.48E-12
Cf-251	0	0	2.30E-14	2.22E-13
Cl-36	7.60E-04	3.00E-03	4.20E-04	4.06E-03
Cm-243	0	0	2.10E-04	2.03E-03
Cm-244	0	2.80E+00	9.50E-01	9.18E+00
Cm-245	0	4.20E-03	2.40E-04	2.32E-03
Cm-246	0	0	0	0
Cm-247	0	1.20E-13	7.10E-14	6.86E-13
Cm-248	0	1.20E-13	7.40E-14	7.15E-13
Co-60	0	0	5.40E-02	5.22E-01
Cs-135	0	5.40E+00	1.30E-04	1.26E-03
Cs-137	4.30E+00	3.00E+05	2.30E+01	2.22E+02
Eu-152	0	0	9.80E-02	9.47E-01
Eu-154	0	0	1.80E+00	1.74E+01
Eu-155	0	0	1.30E-01	1.26E+00
Gd-152	0	0	0	0
H-3	6.10E+00	2.60E+02	3.00E+01	2.90E+02
I-129	1.10E-01	2.80E-01	3.80E-01	3.67E+00
K-40	7.60E-04	3.00E-03	4.20E-04	4.06E-03
Lu-174	0	0	0	0
Mo-93	0	0	0	0
Nb-93m	2.50E-01	1.30E+00	3.70E-01	3.58E+00
Nb-94	2.50E-03	1.30E-02	3.80E-03	3.67E-02
Ni-59	3.50E-02	5.20E-02	8.40E-02	8.12E-01
Ni-63	7.80E-01	2.20E+01	2.40E+00	2.32E+01
Np-237	4.50E-03	6.10E-01	5.00E-02	4.83E-01
Pa-231	0	9.30E-05	9.80E-07	9.47E-06
Pb-210	0	0	0	0
Pd-107	1.90E-03	5.00E-02	5.60E-03	5.41E-02
Pt-193	3.70E-01	1.00E+01	1.10E+00	1.06E+01
Pu-238	7.80E-03	9.10E+03	1.70E+02	1.64E+03
Pu-239	1.20E-02	3.80E+02	1.50E+01	1.45E+02
Pu-240	1.20E-02	1.20E+02	4.10E+00	3.96E+01
Pu-241	9.80E-03	2.40E+03	4.20E+01	4.06E+02
Pu-242	9.00E-04	8.10E-01	3.90E-03	3.77E-02
Pu-244	0	3.60E-03	1.60E-05	1.55E-04
Ra-226	6.40E-07	3.60E-06	7.80E-07	7.54E-06
Ra-228	0	1.60E-06	8.70E-05	8.41E-04
Se-79	3.00E-01	1.40E+00	1.40E+00	1.35E+01
Sm-147	0	0	0	0
Sm-151	0	0	5.90E+01	5.70E+02
Sn-126	0	0	8.20E+00	7.93E+01
Sr-90	6.90E-03	4.60E+04	3.70E+01	3.58E+02
Tc-99	1.10E+02	5.80E+02	5.40E+02	5.22E+03
Th-229	3.00E-01	4.80E+00	3.90E-02	3.77E-01
Th-230	4.10E-01	7.40E+00	1.90E-01	1.84E+00
Th-232	0	2.70E-05	1.40E-03	1.35E-02
U-232	0	0	3.10E-04	3.00E-03
U-233	2.80E-01	4.60E+00	3.70E-02	3.58E-01
U-234	2.80E-01	5.10E+00	1.30E-01	1.26E+00
U-235	3.20E-03	2.10E-01	3.00E-03	2.90E-02
U-236	3.20E-03	1.70E-01	1.60E-02	1.55E-01
U-238	7.40E-03	5.90E-01	1.00E-01	9.67E-01
Zr-93	2.50E-01	1.30E+00	3.70E-01	3.58E+00

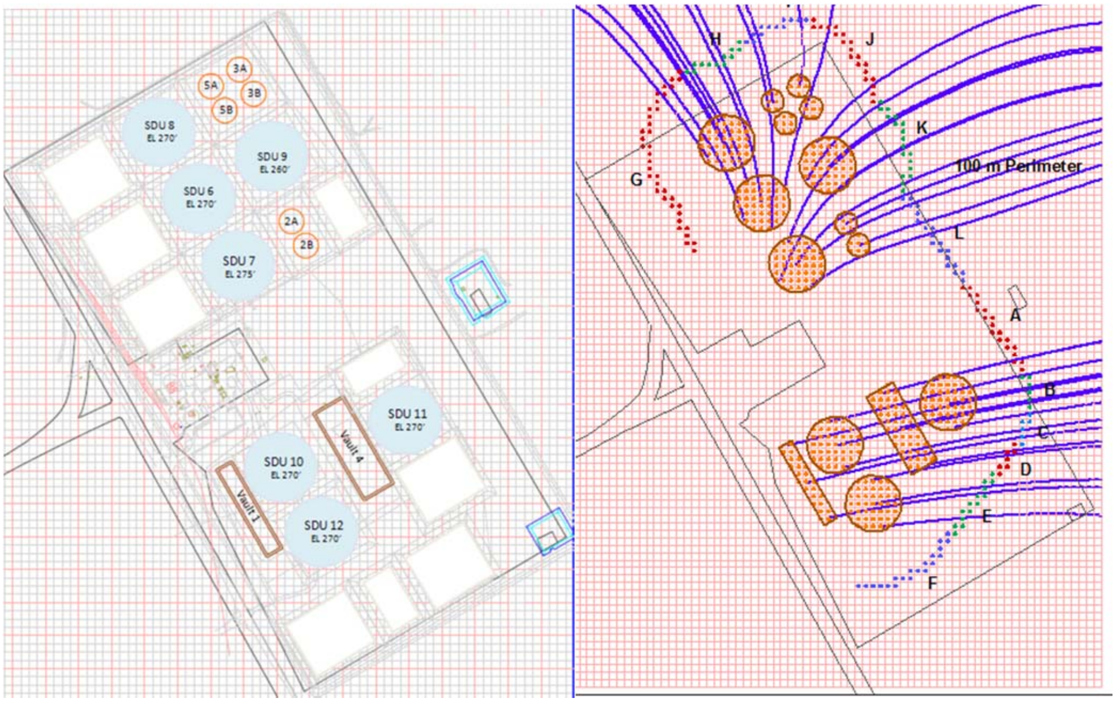


Figure 3-6. Diagrams Showing Location of Saltstone Disposal Facilities and Aquifer Flow Streamlines

3.1.4.2 Saltstone Model Input

Transport parameters used in the GoldSim^{TM} model for the Saltstone analysis are listed in Table 3-7. Parameters applied in the Saltstone analysis are similar to those used for the waste tanks. For this scoping calculation, it was assumed that the residual inventory was placed in the waste zone in 2010 and the cap was placed over the facility in 2025. It was further assumed that the cap failed 500 years after placement. The Type 2 cap entered in Table 3-7 specifies a geo-synthetic material. The waste thickness is the height of the Saltstone and the sand fraction of -1 for the waste identifies the waste material as concrete. The barrier thickness is the thickness of the facility floor and base mat and setting the volume fraction of soil in the barrier to zero specifies concrete material properties.

The vadose zone thickness was obtained from the Saltstone PA.[26] The distance from each disposal facility to the 100 m perimeter was estimated from Figure 4-1. The SDC and SDU facilities with the shortest distances to the 100 m perimeter (SDC-2B and SDU-11) were used in the analysis. The only aquifer parameter that influences the calculations in this work is the aquifer pore velocity which was obtained from the CA.[24] The source area was calculated as the disposal unit cross-sectional area and the last 10 fractions in the model input are set to give an even distribution into the aquifer for the rectangular vaults and a circular distribution for the SDC and SDU facilities. Other parameters in the Table were taken from the SRS CA.[3]

Table 3-7. GoldSim Model Parameters for Saltstone Disposal Facilities

	Vault 1	Vault 4	SDC-2B	SDU-11
Cap Placement Year	2025	2025	2025	2025
Cap Time to Failure (yr)	500	500	500	500
Cap Type	2	2	2	2
Waste Thickness (m)	8.23	9.14	6.71	13.11
Fraction Sand in Waste	-1	-1	-1	-1
Barrier Thickness (m)	0.61	0.61	0.2	0.3048
Volume Fraction Soil in Barrier	1	1	1	1
Vadose Zone Thickness (m)	14.63	11.58	12.19	12.19
Distance to Well (m)	438	273	150	172
Vadose Zone Clay (m)	0	0	0	0
Total Aquifer Path Length (m)	3188.4	2525.4	3188.4	2525.4
Length of First Aquifer Zone (m)	2164.5	1180.6	2164.5	1180.6
Length of Aquitard Zone (m)	374.6	601.0	374.6	601.0
Aquifer Pore Velocity (m/yr)	109.3	174.9	109.3	174.9
Aquitard Pore Velocity (m/yr)	17.6	17.0	17.6	17.0
Stream ID	1	1	1	1
Release Mechanism	1	1	1	1
Source Release Rate (1/yr)	0	0	0	0
Source Area (m^2)	5574.2	11148.4	1641.7	10260.8
Number of Aquitard Clay Cells	1	1	1	1
Footprint Fraction (1)	0.10	0.10	0.0520	0.0520
Footprint Fraction (2)	0.10	0.10	0.0903	0.0903
Footprint Fraction (3)	0.10	0.10	0.1099	0.1099
Footprint Fraction (4)	0.10	0.10	0.1212	0.1212
Footprint Fraction (5)	0.10	0.10	0.1265	0.1265
Footprint Fraction (6)	0.10	0.10	0.1265	0.1265
Footprint Fraction (7)	0.10	0.10	0.1212	0.1212
Footprint Fraction (8)	0.10	0.10	0.1099	0.1099
Footprint Fraction (9)	0.10	0.10	0.0903	0.0903
Footprint Fraction (10)	0.10	0.10	0.0520	0.0520

3.2 Results and Discussion

3.2.1 Waste Tank Results and Discussion

The GoldSim™ model described in Section 2.0 was run for a 10,000 year evaluation period to determine the all-pathways dose at a 100 m well for each radionuclide. As noted in the previous section, Tank 12-H was evaluated using the PA inventory while analysis of the other tanks used an initial radionuclide inventory determined from results of residual waste characterization studies. Results of the model calculations are shown in Figures 3-7 through 3-12 and summarized in Tables 3-8 and 3-9. Figures 3-7 through 3-12 show the total all-pathways dose and the dose from the 14 radionuclides yielding the highest doses. Radionuclides are listed in the plot legends ranked from highest to lowest in order of maximum dose. The plots show doses greater than 1.0E-6 mrem/yr (1.0 nrem/yr). In some cases a radionuclide listed in the legend may not appear on the plot if the maximum dose is less than 1.0E-06 mrem/yr.

The results in Table 3-8 list maximum doses greater than 1.0 nrem/yr over the 10,000 year period of assessment. Highlighted values in Table 3-8 indicate that the maximum dose was obtained at the end of the 10,000 year period of analysis and may still be increasing. The maximum doses are sorted from largest to smallest in Table 3-9 and categorized into three levels. The results in Table 3-9 show that only 10 radionuclides yield doses greater than 0.01 mrem/yr. Nb-94 was included in this group because its maximum dose rounds off to 0.01 mrem/yr. The tables also show that the results were very consistent among the six tanks evaluated and that substituting the assumed PA inventory in place of characterization data for Tank 12-H did not significantly change the results.

Radioactive decay chains complicate interpretation of the results. Five of the ten radionuclides (including the top three dose contributors) yielding doses greater than 0.01 mrem/year are daughters in decay chains. Parent radionuclides for these radionuclides are listed in Table 3-10. The parents add another 17 radionuclides that may also need to be included in waste characterization analysis. It is necessary to have characterization information on parent radionuclides to account for daughter ingrowth during the evaluation period. While it is likely that not all of these parents contribute significantly to the final dose (e.g. parents with long half-lives), this effect was not evaluated as part of this preliminary study.

Conversely, the E-Area Low Level Waste Facility (ELLWF) PA[27] found that Bi-214 was a major contributor to dose even though it did not exist in the facility's initial inventory.[28-30] This demonstrates that short-lived daughter radionuclides may contribute to dose risk while not appearing in the initial inventory. It is possible that parent radionuclides could be screened out of the analysis if they move slowly and do not reach the point of assessment or if they have a low dose contribution while daughter products may contribute to dose.

3.2.2 Comparison with Tank Farm PAs

Brief excerpts from the F-Area Tank Farm PA and H-Area Tank Farm PA that summarize results from the dose calculations performed in the PAs have been copied into Appendices A and B, respectively, for quick reference.

The F-Area Tank Farm PA concludes that over a 10,000 year period of assessment, the radionuclides having the largest contribution to dose to a member of the public (MOP) at the 100 m boundary are Np-237 and Ra-226 in agreement with the results from this study. However, the PA finds that other radionuclides contributing > 5% to the peak dose are Cs-137, U-233 and U-234. As shown in PA Table 5.5-4 reproduced in Appendix H, the Cs-137 dose appears to come from ingestion of fish. Fish ingestion was not included as a dose pathway in the present study because, as was done in the SRS CA, consumption of contaminated fish was assumed to occur from recreational use of stream or river water not from residential use of well water. The appearance of U-233 and U-234 as significant dose contributors in the PA is at odds with the results of this study which shows U-233 appearing as only a small contributor to dose from Tank 6-F near 10,000 years.

The F-Area Tank Farm PA included ancillary equipment and transfer lines in its analysis. The PA found that the ancillary equipment accounted for dose peaks in early years. The PA summary notes significant doses from I-129 and C-14 from Type IV tanks which is again in general agreement with the results obtained in this study. The F-Area Tank Farm PA also concludes that the six parent radionuclides Am-241 (for Np-237), Am-243 (for Pu-239), Cm-244 (for Pu-240), Pu-238 (for Ra-226), Th-230 (for Ra-226), and U-235 (for Pa-231) must be included in the analysis. All of these parent radionuclides were included in the present analysis although an assessment of which parent radionuclides are important to dose was not made.

The H-Area Tank Farm PA concludes that over a 10,000 year period of assessment, peak doses to a MOP at the 100 m boundary are primarily from Tc-99, Pa-231 and Ra-226. Based on characterization data, the current study found doses from Tank 18-H are primarily from Pa-231, I-129, Ra-226, Np-237 and Nb-93m. The dose from Tc-99 was found to be relatively insignificant. Using the PA inventory for Tank 12-H, Ra-226, Np-237 and Pa-231 gave the highest doses while the dose from Tc-99 was again small. Table 5.5-2, copied from the H-Area Tank Farm PA in Appendix I, shows that the peak dose at 8,790 years in Sector A is 96% from Tc-99 with smaller contributions from Ra-226, Pa-231, Np-237 and Nb-94. Table 5.5-3 from the PA, also reproduced in Appendix I, indicates that 97% of the dose in Sector A at 8,760 years is from Tanks 9-H – 12-H. PA Table 5.5-4, reproduced in Appendix I, indicates that Tc-99 dominates the dose in Sectors A (Tanks 9-H – 12-H), E and F, but is less important in Sectors B, C and D. The footnote to Table 5.5-3 indicates that only Type I Tanks 9-H – 12 H, Type II Tanks 13-H – 16-H and Type IV Tanks 21-H – 24-H contribute to dose over the 10,000 year period of assessment.

The appearance of Tc-99 as the primary dose contributor for H-Area tanks in the Tank Farm PA is significantly different from the results obtained with the risk screening model. The H-Area Tank Farm PA assumes an initial inventory of 8.2 Ci Tc-99 in Tank 12-H. The F-Area Tank Farm PA assumes an initial inventory of 79.0 Ci Tc-99 in Tanks 5-F and 6-F, which is almost ten times greater. Nevertheless, Tc-99 does not dominate F-Area Tank Farm doses as it does those for the H-Area Tank Farm. The source of this difference is likely associated with differences in the respective modeling assumptions and/or in the application of the dose model. Further investigation of this difference should be pursued and the source of the difference reconciled if the screening method is adopted for routine use.

The H-Area Tank Farm PA also includes ancillary equipment and transfer lines in its analysis and attributes dose peaks in early years to these sources. The H-Area Tank Farm PA concludes that the six parent radionuclides Am-241 (for Np-237), Pu-238 (for Ra-226), Pu-239 (for Pa-231), Th-230 (for Pb-210 and Ra-226), U-234 (for Pb-210 and Ra-226), and U-235 (for Pa-231) must be included in the analysis. All of these radionuclides were included in the present analysis. It has not been determined if all of these parents produce sufficient decay products to necessitate their inclusion in waste characterization measurements.

Table 3-8. Maximum Doses (in mrem/yr) Greater than 1.0 nrem/yr for Waste Tanks

	Waste Tank					
Nuclide	**5-F**	**6-F**	**18-F**	**19-F**	**12-H***	**16-H**
Ac-227			6.3E-04	1.2E-05	5.8E-05	9.9E-05
C-14		1.5E-06	6.7E-02	3.9E-01	4.0E-03	2.8E-06
Cl-36	1.3E-04		7.9E-04	3.2E-04	1.3E-03	1.5E-03
Cs-135			3.0E-03	3.2E-03	2.0E-04	2.8E-04
I-129	1.1E-03	5.0E-04	4.5E-02	4.8E-02	1.5E-02	1.4E-01
K-40	5.4E-05		2.7E-02	2.3E-03	6.6E-04	3.4E-03
Nb-93m	2.8E-03	1.8E-03	1.7E-02	5.1E-03	3.7E-02	4.3E-02
Nb-94	4.6E-06	6.4E-06	3.3E-04	7.2E-05	7.2E-03	4.6E-04
Ni-59	2.5E-04	3.6E-04	3.8E-03	5.3E-06	1.4E-02	1.8E-03
Np-237	7.8E-03	1.7E-02	8.8E+00	1.3E-01	2.7E+00	6.0E-02
Pa-231	2.8E-04	1.5E-03	1.1E+00	2.1E-02	9.9E-02	1.7E-01
Pb-210	1.4E-05	9.7E-06	4.0E-03	1.7E-03	1.1E-02	4.6E-04
Pd-107			9.3E-04	1.9E-03	2.3E-04	
Ra-226	2.4E-03	1.7E-03	6.8E-01	2.9E-01	1.9E+00	7.8E-02
Sr-90					3.8E-06	
Tc-99	4.7E-05	3.8E-04	3.3E-04	3.3E-04	1.3E-04	7.9E-05
Th-229			4.3E-05		3.0E-05	
U-233			3.8E-05		2.9E-05	

*Analysis of Tank 12-H used the PA inventory while the analysis of the other tanks used an inventory determined from residual waste characterization data. Shaded cells indicate that the maximum dose was reached at 10,000 years.

Table 3-9. Maximum Doses (in mrem/yr) Greater than 1.0 nrem/yr Ranked in Order of Maximum Dose for Waste Tanks

		Waste Tank						
	Nuclide	**5-F**	**6-F**	**18-F**	**19-F**	**12-H***	**16-H**	**Maximum**
> 0.01 mrem/yr	Np-237	7.8E-03	1.7E-02	8.8E+00	1.3E-01	2.7E+00	6.0E-02	8.8E+00
	Ra-226	2.4E-03	1.7E-03	6.8E-01	2.9E-01	1.9E+00	7.8E-02	1.9E+00
	Pa-231	2.8E-04	1.5E-03	1.1E+00	2.1E-02	9.9E-02	1.7E-01	1.1E+00
	C-14		1.5E-06	6.7E-02	3.9E-01	4.0E-03	2.8E-06	3.9E-01
	I-129	1.1E-03	5.0E-04	4.5E-02	4.8E-02	1.5E-02	1.4E-01	1.4E-01
	Nb-93m	2.8E-03	1.8E-03	1.7E-02	5.1E-03	3.7E-02	4.3E-02	4.3E-02
	K-40	5.4E-05		2.7E-02	2.3E-03	6.6E-04	3.4E-03	2.7E-02
	Ni-59	2.5E-04	3.6E-04	3.8E-03	5.3E-06	1.4E-02	1.8E-03	1.4E-02
	Pb-210	1.4E-05	9.7E-06	4.0E-03	1.7E-03	1.1E-02	4.6E-04	1.1E-02
	Nb-94	4.6E-06	6.4E-06	3.3E-04	7.2E-05	7.2E-03	4.6E-04	7.2E-03
< 0.01 > 0.001 mrem/yr	Cs-135			3.0E-03	3.2E-03	2.0E-04	2.8E-04	3.2E-03
	Pd-107			9.3E-04	1.9E-03	2.3E-04		1.9E-03
	Cl-36	1.3E-04		7.9E-04	3.2E-04	1.3E-03	1.5E-03	1.5E-03
	Ac-227			6.3E-04	1.2E-05	5.8E-05	9.9E-05	6.3E-04
< 0.001 > 1.0E-06 mrem/yr	Tc-99	4.7E-05	3.8E-04	3.3E-04	3.3E-04	1.3E-04	7.9E-05	3.8E-04
	Th-229			4.3E-05		3.0E-05		4.3E-05
	U-233			3.8E-05		2.9E-05		3.8E-05
	Sr-90					3.8E-06		3.8E-06

*Analysis of Tank 12-H used the PA inventory while the analysis of the other tanks used an inventory determined from residual waste characterization data.

Table 3-10. Parent Radionuclides*

Radionuclide	**Parents (Half-life in Years)**	
Np-237	Am-241 (4.32E+02) Pu-241 (1.43E+01)	Cm-245 (8.50E+03) Cf-249 (3.51E+02)
Ra-226	Th-230 (7.54E+04) U-234 (2.46E+05) U-238 (4.47E+09)	Pu-238 (8.77E+01) Pu-242 (3.75E+05) Am-242m (1.41E+02)
Pa-231	U-235 (7.04E+08) Cm-243 (2.91E+01) Am-243 (7.37E+03)	Pu-239 (2.41E+04) Cm-247 (1.56E+07)
Nb-93m	Mo-93 (4.00E+03)	Zr-93 (1.53E+06)
Pb-210	Ra-226 (1.60E+03) Th-230 (7.54E+04) U-234 (2.46E+05)	U-238 (4.47E+09) Pu-238 (8.77E+01)

*Radionuclides with half-lives less than 1000 years are highlighted.

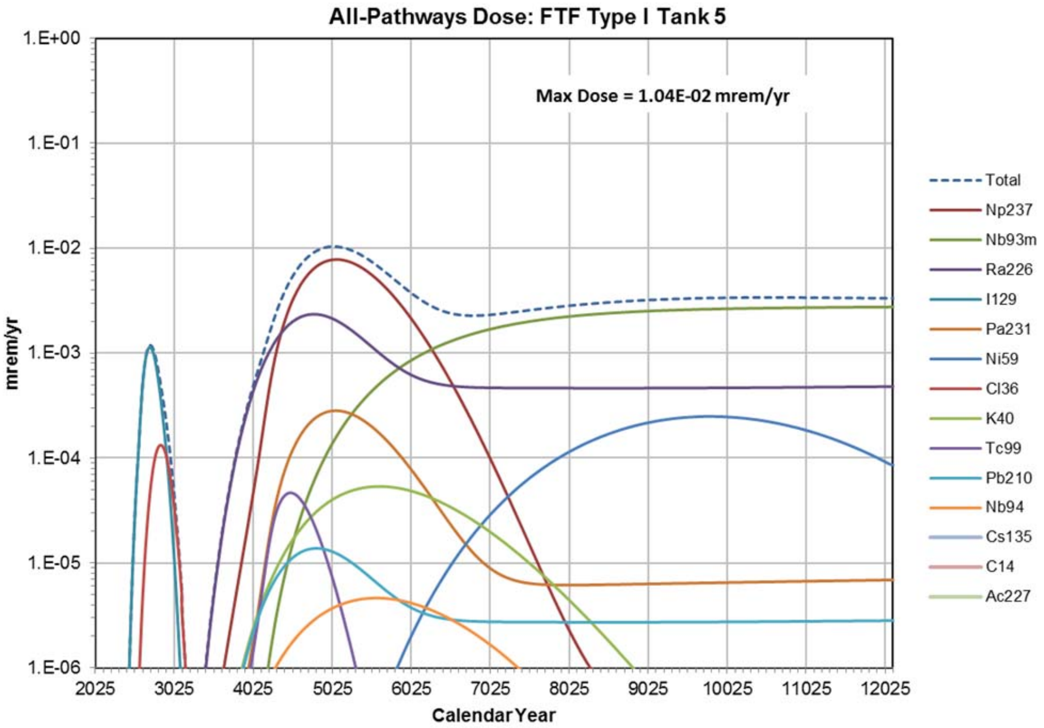


Figure 3-7. All-Pathways Dose from Tank 5-F Based on Characterization

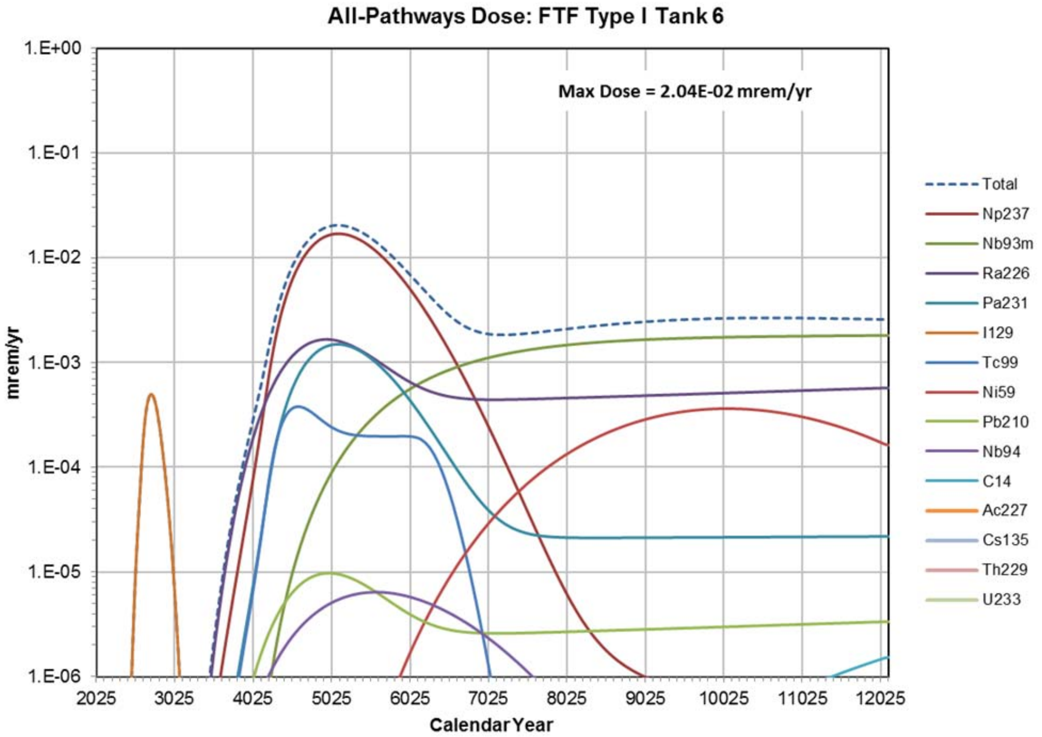


Figure 3-8. All-Pathways Dose from Tank 6-F Based on Characterization

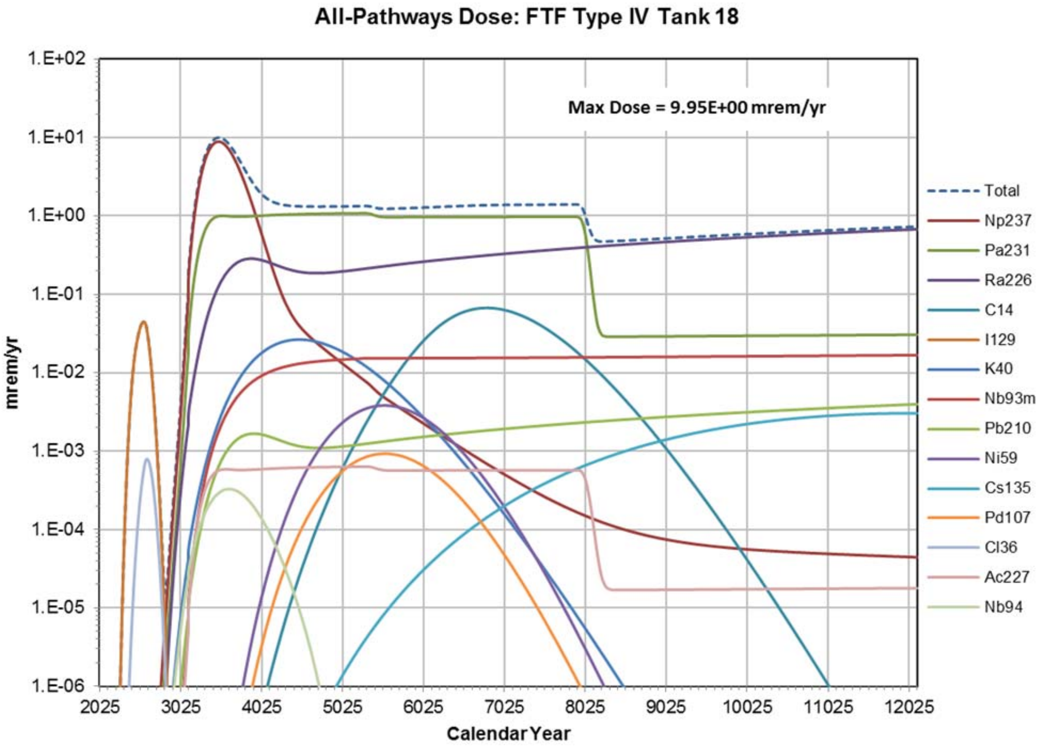


Figure 3-9. All-Pathways Dose from Tank 18-F Based on Characterization

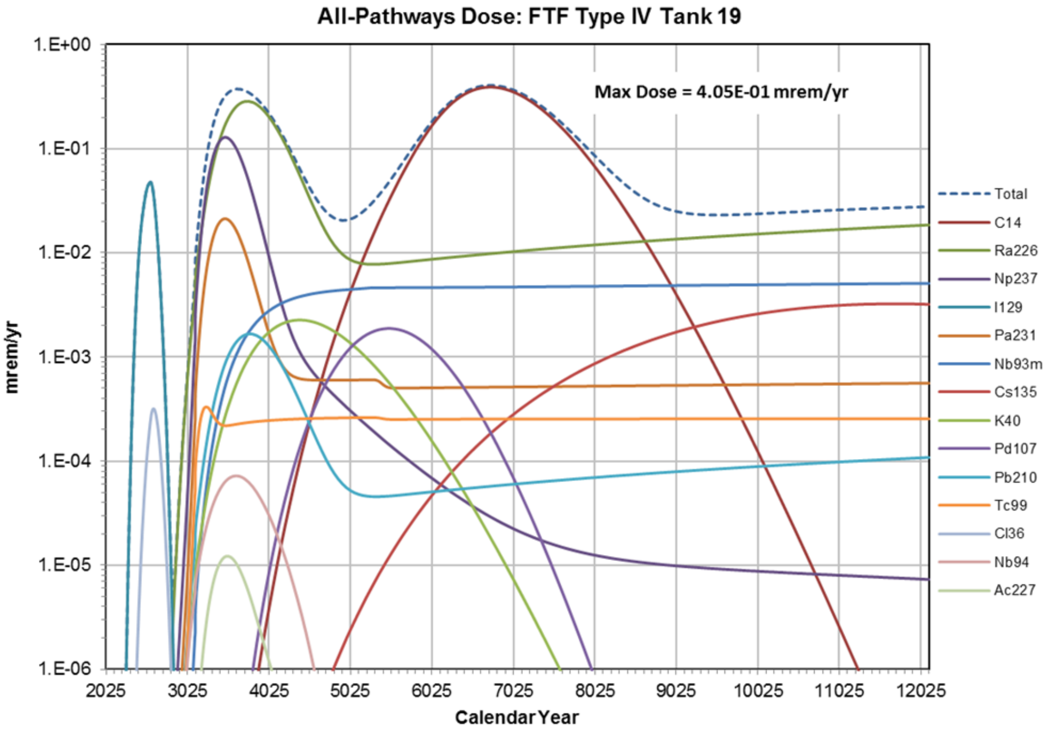


Figure 3-10. All-Pathways Dose from Tank 19-F Based on Characterization

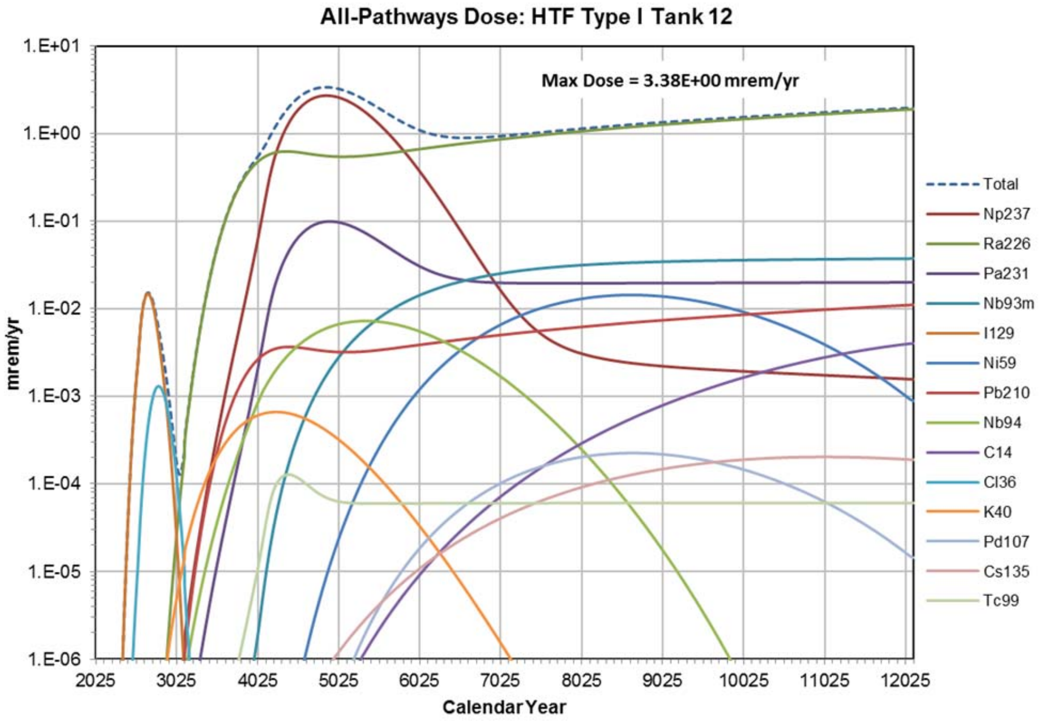


Figure 3-11. All-Pathways Dose from Tank 12-H Based on PA Inventory

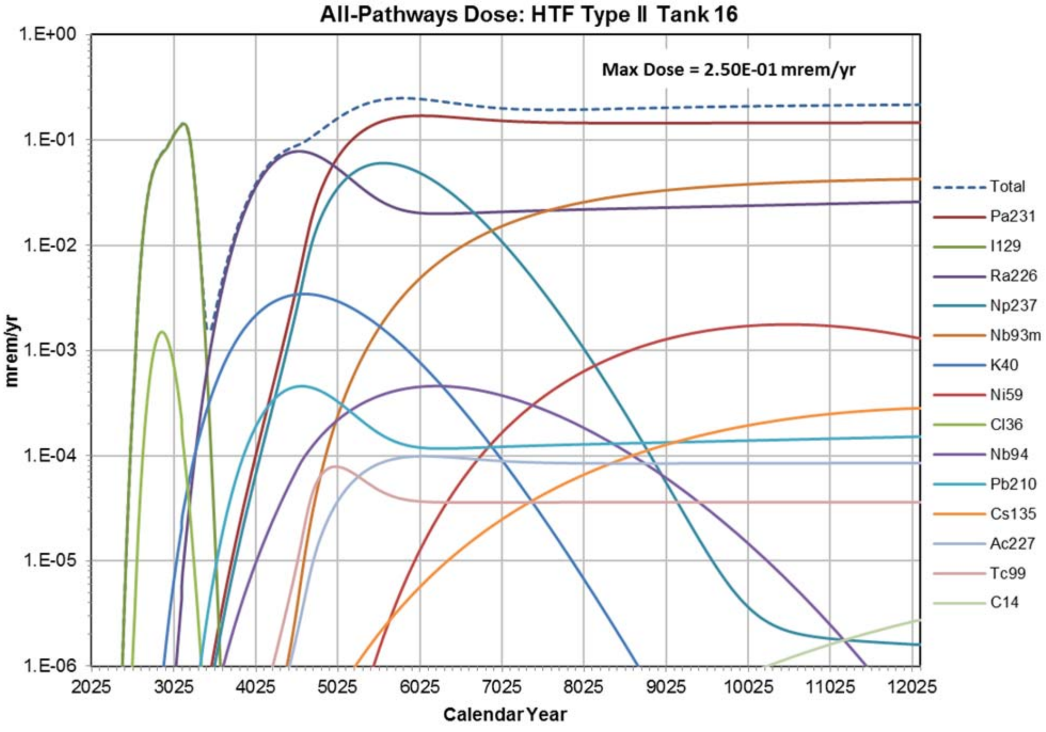


Figure 3-12. All-Pathways Dose from Tank 16-H Based on Characterization

3.2.3 Saltstone Results and Discussion

Results of the model calculations for Saltstone disposal are shown in Figures 3-13 through 3-16 and summarized in Table 3-11. Figures 3-13 through 3-16 show the total all-pathways dose and the dose from the 14 radionuclides yielding the highest doses. Radionuclides are listed in the plot legends ranked from highest to lowest in order of maximum dose. The plots show doses greater than 1.0E-06 mrem/yr (1.0 nrem/yr). In some cases a radionuclide listed in the legend may not appear on the plot if the maximum dose is less than 1.0E-06 mrem/yr. The plots show that after approximately 8,000 years total doses from the SDC and SDU units have peaked and remain nearly constant.

The results in Table 3-11 list maximum doses greater than 1.0 nrem/yr over the 10,000 year period of assessment. Maximum doses are sorted from largest to smallest in Table 3-11 and categorized into three levels. The results in Table 3-11 show that only three radionuclides (Ra-226, I-129 and Np-237) yield doses greater than 0.01 mrem/yr. Extending the risk level to include doses greater than 0.001 mrem/yr (1.0 µrem/yr) adds another five parent radionuclides. There is a significant fall-off in dose below 1.0 µrem/yr with the next highest dose from Nb-94 a factor of 20 lower than the dose from Pb-210. The table also shows that results are consistent among the four types of disposal unit evaluated. As expected Cs-135 contributes significantly to the dose from Vault 4 but is not a factor for the other units and would not need to be included in future Saltstone waste characterization. The seven radionuclides other than Cs-135 that contribute significantly to Saltstone dose risk are included in the ten radionuclides that contribute significantly to dose risk from the waste tanks. As for the waste tanks, radioactive decay chains complicate interpretation of the results. Five of the eight radionuclides (including two of the top three dose contributors) yielding doses greater than 0.001 mrem/year are daughters in decay chains. The parent radionuclides for these daughters are listed in Table 3-10 and have been discussed in Section 3.2.1.

The model used in this analysis is very simple compared to the models used for more extensive PA and SA calculations. The risk assessment model is intended to conservatively predict the relative importance of radionuclides in determining dose to an off-site MOP and may not provide accurate calculations of absolute doses. Nevertheless, model results should reflect the relative importance of radionuclides to dose and it is expected that the top eight radionuclides identified by the Saltstone risk analysis are the species of most significance.

3.2.4 Comparison with Saltstone PA

A brief excerpt from the Saltstone PA that summarizes results from the dose calculations performed in the PA has been copied into Appendix J for quick reference.

The Saltstone PA identified a set of key radionuclides identified as any radionuclide with a peak all-pathways dose greater than 0.05 mrem/yr over a 20,000 year period of assessment. The key radionuclides identified in the PA are: Tc-99, I-129, Ra-226, Np-237 and Pa-231. The risk screening analysis would conservatively include Nb-93m, Pb-210, C-14 and Cs-135 (for Vault 4) and remove Tc-99. The PA also identifies the parent radionuclides U-235 (for Pa-231), Th-230 (for Ra-226), U-234 (for Ra-226), and Pu-238 (for Ra-226) as significant contributors to the total dose. Results from the PA are in general agreement with the results obtained from the risk assessment. Again some discrepancy appears to exist for Tc-99 with the PA dose model finding a significant dose from Tc-99 while risk screening determines that the dose from Tc-99 is negligible. Tc-99 release and transport from Saltstone was modeled very differently in the PA including effects such as Tc-99 solubility, a moving reduction front, and flow through fractured media which were not considered in the simple risk screening model. Both calculations agree that I-129 and Ra-226 are the radionuclides that primarily contribute to dose.

Table 3-11. Maximum Doses (in mrem/yr) Greater than 1.0 nrem/yr Ranked in order of Maximum Dose for Saltstone Disposal Facilities

		Saltstone Disposal Facility				
	Nuclide	**Vault 1**	**Vault 4**	**SDC-2B**	**SDU-11**	**Maximum**
> 0.01 mrem/yr	Ra-226	4.32E-02	2.72E-01	1.07E-01	1.10E-01	2.72E-01
	I-129	1.42E-02	1.09E-02	4.01E-02	2.52E-02	4.01E-02
	Np-237	7.80E-05	3.53E-03	3.08E-02	3.82E-02	3.82E-02
< 0.01 > 0.001 mrem/yr	C-14	2.71E-05	3.61E-04	3.37E-03	1.78E-03	3.37E-03
	Nb-93m	6.46E-04	1.50E-03	3.29E-03	3.00E-03	3.29E-03
	Cs-135		1.75E-03			1.75E-03
	Pa-231	6.89E-05	1.68E-03	1.06E-03	1.32E-03	1.68E-03
	Pb-210	2.53E-04	1.60E-03	6.27E-04	6.39E-04	1.60E-03
< 0.001 > 1.0E-06 mrem/yr	Nb-94	1.30E-06	2.88E-06	7.11E-05	4.56E-05	7.11E-05
	Ni-59	1.91E-06	1.05E-06	3.29E-05	2.23E-05	3.29E-05
	K-40	6.34E-06	7.72E-06	1.75E-05	1.15E-05	1.75E-05
	Cl-36	1.54E-06	1.90E-06	1.11E-05	7.05E-06	1.11E-05
	Tc-99	6.93E-06	4.33E-06	6.93E-06	4.33E-06	6.93E-06
	Pd-107			1.38E-06		1.38E-06

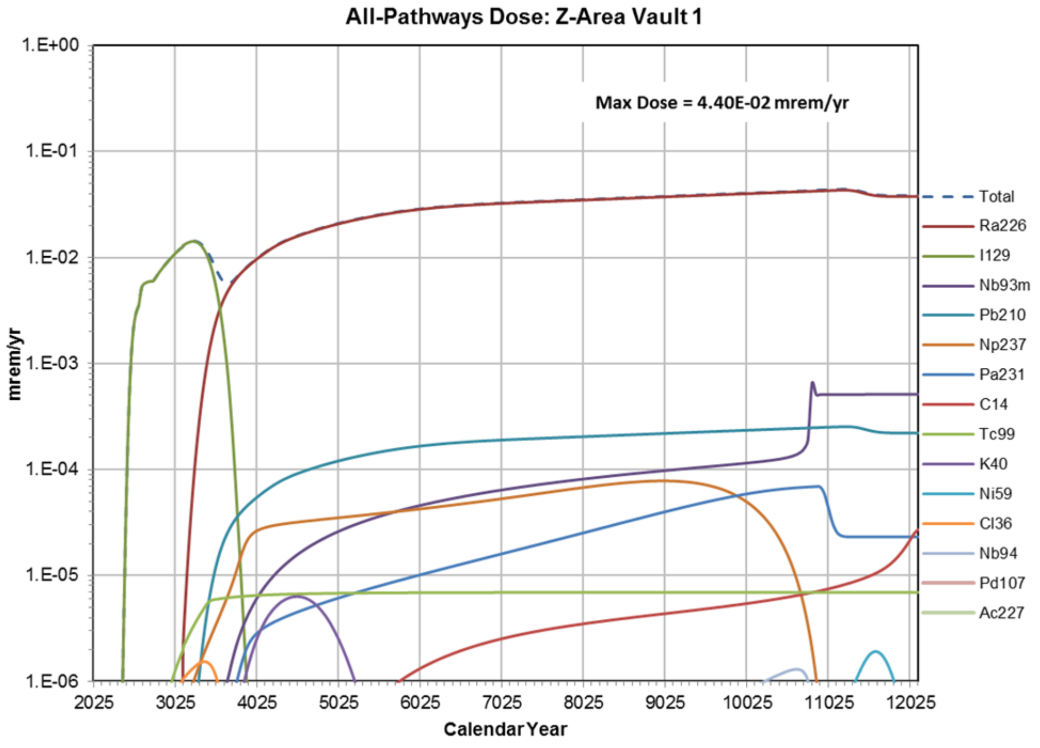


Figure 3-13. All-Pathways Dose from Saltstone Vault 1 Based on PA Inventory

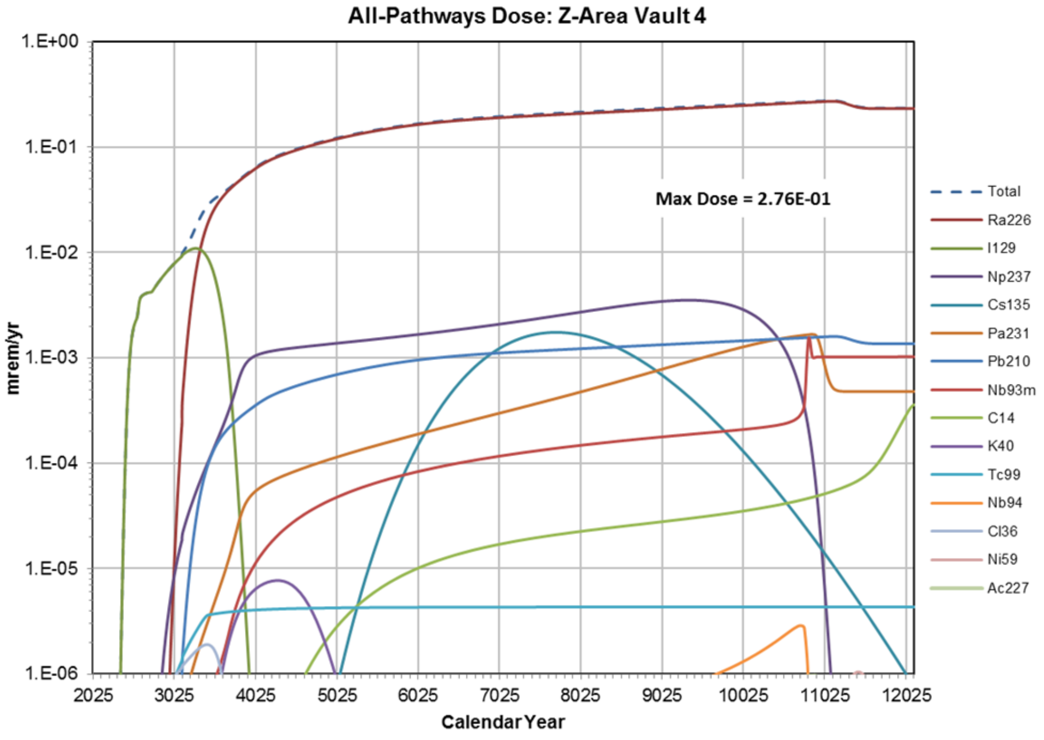


Figure 3-14. All-Pathways Dose from Saltstone Vault 4 Based on PA Inventory

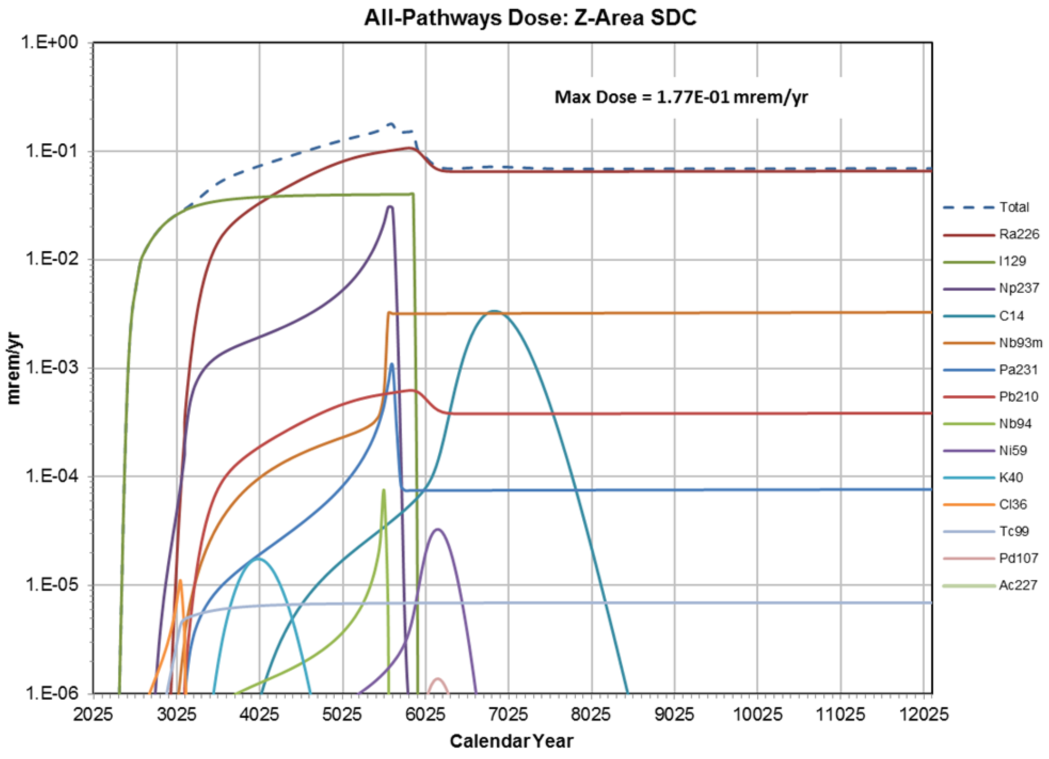


Figure 3-15. All-Pathways Dose from SDC 2B Based on PA Inventory

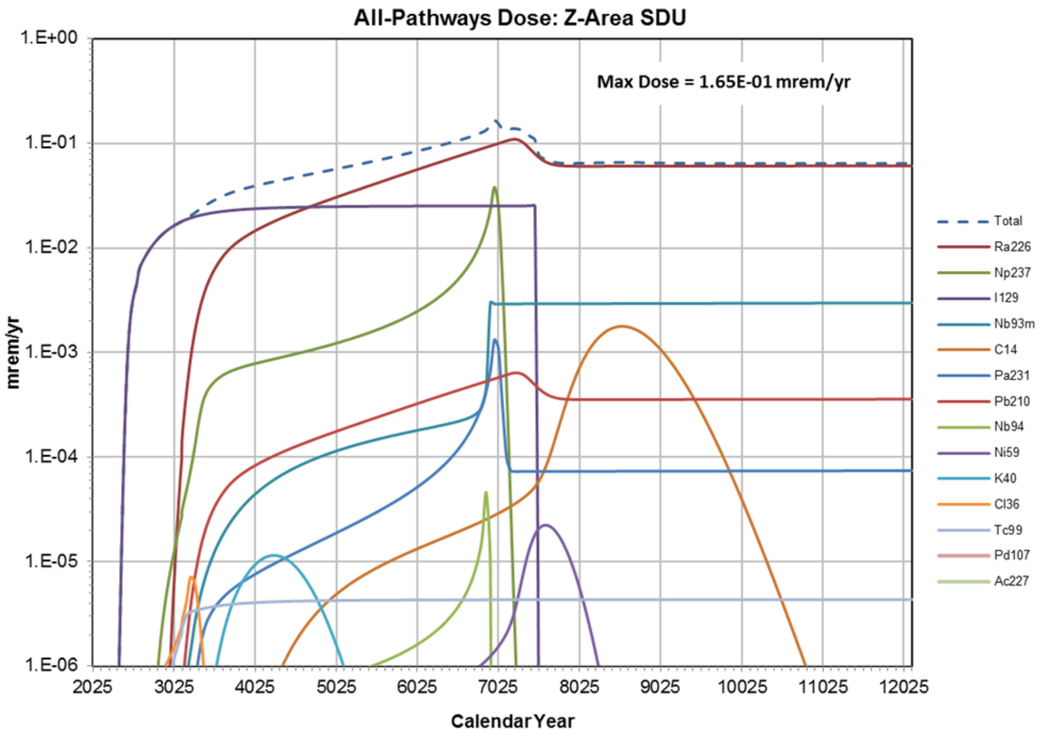


Figure 3-16. All-Pathways Dose from SDU 11 Based on PA Inventory

3.3 Conclusions

Results from this study evaluating risk screening as a method of reducing waste characterization requirements are promising. Results for the six waste tanks considered in this analysis gave consistent results indicating that ten radionuclides contribute significantly to dose. However, the levels of as many as 17 parent radionuclides may need to be determined as well adding a further degree of complexity to the required characterization. Similarly, results for the four Saltstone disposal units considered in this analysis gave consistent results indicating that eight radionuclides contribute significantly to the dose risk. In addition, the two sets of results are very similar. With the exception of Cs-135, which is only significant for Saltstone Vault 4, the ten radionuclides identified as important for waste tank dose include the eight radionuclides identified as important for Saltstone dose.

Neither the residual waste tank inventories assumed in the Tank Farm PAs or in the SRS CA represented particularly accurate estimates of the actual residual material present at tank closure as determined from waste characterization analyses. Inventories for individual radionuclides assumed in the Tank Farm PAs were both significantly larger and significantly lower than values determined from characterization data. Even though radionuclide inventories assumed in the CA were in general lower than values based on characterization data, in many cases absolute differences were small. The CA estimate of waste tank closure inventory was based on preliminary estimates made before any characterization of residual waste had been performed. The CA concluded that doses from the closed waste tanks were small and the differences between the preliminary inventory estimates and characterization based values found in this study are unlikely to change this conclusion.

The proposed methodology is also applicable to solid waste disposal in E-Area. The E-Area PA[27] uses probabilistic modeling with GoldSim^{TM} to evaluate variability and uncertainty in PA model input parameters. The result is a distribution of likely dose or risk to a member of the public. E-Area disposal includes a wide variety of contaminated materials and no attempt was made to identify the most important radionuclides for solid waste disposal in this preliminary study.

3.4 Recommendations, Path Forward or Future Work

SRNL and Savannah River Remediation (SRR) have developed GoldSim^{TM} models used in the Saltstone, Tank Farm, and E-Area PAs that are more comprehensive than the simple risk assessment model described in Section 3.1.1 which was derived from the model used for the SRS CA. If the risk screening approach is to be applied, it may be preferential to employ these more detailed models. Alternatively, the model developed in this study could be enhanced to better emulate PA modeling results. The model needs to be validated against PA dose results before acceptance for risk screening. In particular, the discrepancy between the importance of Tc-99 to doses predicted in the H-Area Tank Farm and Saltstone PAs and the relative unimportance of this radionuclide in the risk screening model results needs to be resolved. The SRNL/SRR models were used to conduct uncertainty analysis for the PAs and including uncertainty in the risk screening analysis provides a measure of the reliability of conclusions reached by applying the model.

Some additional work is also required to determine whether all of the 17 parent radionuclides identified in Table 3-10 contribute significantly to the dose from daughter radionuclides. The Saltstone and Tank Farm PA analyses would suggest that about half of these radionuclides (Am-241, Pu-238, Pu-239, Th-230, U-234 and U-235) with the possible addition of Mo-93 and Zr-93 actually need to be considered. This evaluation could be accomplished relatively easily by running the screening model for each parent and daughter individually to see how much each radionuclide alone contributes to total dose.

3.5 References

[1] GTG (GoldSim^{TM} Technology Group) 2010, *"GOLDSIM User's Guide"*, Version 10.6, GoldSim Technology Group LLC, Issaquah, WA, January 2010.

[2] Taylor, G.A., L. McDowell-Boyer, P.L. Lee and E.L. Wilhite, *"Radionuclide Screening Model for the Savannah River Site's Composite Analysis"*, SRNS-STI-2008-00117, September 30, 2008.

[3] SRNL, *"Savannah River Site Composite Analysis"*, Savannah River National Laboratory, Aiken, SC, SRNL-STI-2009-00512, Rev. 0, June 10, 2010.

[4] Jannik, G.T., D.J. Karapatikis and P.L. Lee, *"Land and Water Use Characteristics and Human Health Input Parameters for Use in Environmental Dosimetry and Risk Assessments at the Savannah River Site"*, SRNL-STI-2010-00447, Rev. 0, August 6, 2010.

[5] Stone, D.K. and G.T. Jannik, *"Site Specific Reference Person Parameters and Derived Concentration Standards for the Savannah River Site"*, SRNL-STI-2013-00115, March 2013.

[6] Phifer, M.A., L. McDowell-Boyer, K.E. Young and J.R. Cook, *"Savannah River Site Composite Analysis: Dose Module"*, SRNL-STI-2009-00424, Rev. 0, August 2009.

[7] SRS, *"Performance Assessment for the F-Tank Farm at the Savannah River Site"*, Savannah River Site, SRS-REG-2007-00002, Rev. 1, January 2007.

[8] SRR, *"Performance Assessment for the H-Area Tank Farm at the Savannah River Site"*, Savannah River Remediation, SRR-CWDA-2010-00128, Rev. 1, November 2012.

[9] Hiergesell, R.A., M.A. Phifer, J.R. Cook, K.E. Young, M.B. Birk and W.B. Dean, "*Inventory of Residual Radioactive Material at the Projected Savannah River Site End State"*, SRNL-STI-2008-00380, Rev. 0, October 2008.

[10] Oji, L.N., D. Diprete and C.J. Coleman, *"Analysis of the Tank 5 Final Characterization Samples-2011 (U)"*, SRNL-STI-2012-00034, Rev. 2, September 27, 2012.

[11] Oji, L.N., D. Diprete, C.J. Coleman, M.S. Hay and E.P. Shine, *"Analysis of the Tank 6F Final Characterization Samples-2012 (U)"*, SRNL-STI-2012-00365, Rev. 2, January 2013.

[12] Shine, E.P., *"Statistical Analysis of Tank 5 Floor Sample Results"*, SRNL-STI-2011-00613, Rev. 2, January 2013.

[13] SRR, *"Tank 6 Final Volume Determination and Uncertainty Estimate"*, Savannah River Remediation, SRR-LWE-2011-00245, Rev. 2, June 2012.

[14] Oji, L.N., D. Diprete and D.R. Clark, *"Characterization of the Tank 18F Samples"*, SRNL-STI-2009-00625, Rev. 0, December 17, 2009.

[15] Oji, L.N., D. Diprete and C.J. Coleman, *"Characterization of the Additional Tank 18F Samples"*, SRNL-STI-2010-00386, Rev. 0, September 2, 2010.

[16] Hay, M.S., D.R. Click, D.P. Diprete and C.C. Diprete, *"Characterization of Tank 18F Wall and Scale Samples"*, SRNL-STI-2009-00802, Rev. 0, March 2010.

[17] Harris, S.P, *"Statistical Analysis of Tank 18F Floor Sample Results"*, SRNL-STI-2010-00401, September 2, 2010.

18 SRR, *"Tank 18 Residual Characterization Report"*, Savannah River Remediation, SRR-CWDA-2010-00117, Rev. 1, October 26, 2011.

19 Oji, L.N., D. Diprete, C.J. Coleman and M.S. Hay, *"Characterization of the Additional Tank 19F Floor Samples"*, SRNL-STI-2010-00439, Rev. 0, August 31, 2010.

20 Harris, S.P, *"Statistical Analysis of Tank 19F Floor Sample Results"*, SRNL-STI-2010-00400, September 2, 2010.

21 SRR, *"Tank 19 Residual Characterization Report"*, Savannah River Remediation, SRR-CWDA-2010-00118, Rev. 1, October 26, 2011.

22 Oji, L.N., D. Diprete, C.J. Coleman, M.S. Hay and E.P. Shine, *"Tank 16H Residual Sample Analysis Report (U)*", SRNL-STI-2014-00321, Rev. 0, August 2014.

23 SRR, *"Tank 16 Final Residual Solids Determination and Uncertainty Estimate"*, Savannah River Remediation, U-ESR-H-00113, Rev. 0, November 2013.

24 Hamm, L.L., F.G. Smith and M.A. Phifer, *"Savannah River Site Composite Analysis: Aquifer Flow Path Parameters"*, SRNL-STI-2009-00438, Rev. 0, July 2009.

25 Dean, W. B., "*Estimated Closure Inventory for the Saltstone Disposal Facility"*, Savannah River Site, SRNS-J2100-2008-00004, Rev. 2, June 22, 2009.

26 SRR, *"Performance Assessment for the Saltstone Disposal Facility at the Savannah River Site*", Savannah River Remediation, SRR-CWDA-2009-00017, Rev. 0, October 2009.

27 WSRC, *"E-Area Low-Level Waste Facility DOE 435.1 Performance Assessment"*, Westinghouse Savannah River Company, WSRC-STI-2007-00306, July 2008.

28 Taylor, G.A., *"A Proposed Method for Performing Sensitivity Analyses for Performance Assessment and the Curious Case of ^{214}Bi"*, SRNL-STI-2010-00762, Rev. 0, October 2010.

29 Taylor, G.A., "*^{214}Bi Contribution to Garden Dose: Fact or Fiction"*, SRNL-STI-2011-00488, Rev. 0, August 2011.

30 Taylor, G.A. and R.A. Hiergesell, *"Benchmarking Exercises to Validate the Updated ELLWF Goldsim Trench Models"*, SRNL-STI-2010-00737, Rev. 1, November 2013.

4.0 Determining Relevancy of Available Technical Bases

This section addresses an investigation undertaken to determine the relative usefulness of various potential sources of tank characterization data, including: a) laboratory analysis of tank waste samples; b) tank waste receipt histories; c) tank waste process knowledge; d) theoretical scaling factors; and e) historic data trends. Although sample-specific data generated through laboratory analysis is typically utilized on its own (in the absence of other waste considerations), there is the possibility of utilizing alternative technical bases, where justified, either as a complement to the analytical data or as a potential standalone source of data. Such alternative approaches offer particular advantages in cases where the existing laboratory characterization method is unwieldy, costly and/or time consuming. Additionally, in cases where the constituent being characterized has minimal or no practical impact on the disposition decisions/requirements, a non-laboratory method may provide sufficient quantification accuracy without the high costs and time requirements of laboratory analyses.

4.1 Approach

Discussions of the merits and limitations of the various available technical bases are provided in this section, along with comparisons providing a measure of data consistency and relative data uncertainty. This information serves as a basis for developing a path forward for integrating appropriate alternative characterization approaches into SRNL's existing tank waste characterization program. It also serves as a basis for developing "lessons learned," which can benefit other DOE sites faced with tank waste characterization challenges.

4.2 Laboratory Analysis of Tank Waste Samples

Laboratory analysis of tank waste samples provides a direct means of acquiring waste-specific characterization data. Although sampling and analysis is extremely resource-intensive, it is often the preferred characterization approach due to reasons of data defensibility, sample specificity, and analyte specificity. However, sampling and analysis is not without problems and clearly requires a high level of expertise and extensive facility capabilities.

Radioanalytical methods are typically time-consuming and costly, due to the special handling requirements associated with potential radiation exposures and the large amount of sample preparation/treatment needed to remove interfering radionuclides and stable constituents. Most radioanalytical methods incorporate a succession of the following primary steps: 1) sample dissolution and/or dilution; 2) removal of interfering constituents via chemical separations; 3) purification of analytes via radiochemical separations; 4) measurement of analytes; and 5) quality assurance measures demonstrating analysis effectiveness. Given that chemical/radiochemical separation techniques are often matrix-dependent, the methods for effectively performing radionuclide analyses typically require some amount of R&D. Despite the challenges, sampling and analysis remains the primary source of data used to characterize DOE tank waste in preparation for disposition or supporting tank closure.

At the Savannah River Site (SRS), sampling and analysis has been successfully utilized for characterizing: a) sludge waste in preparation for being transferred to the Defense Waste Processing Facility (DWPF); b) salt batch qualification demonstrating processability; c) processed salt waste in preparation for being transferred to the Saltstone Production Facility/Saltstone Disposal Facility (SPF/SDF); and d) post-cleaning residue in preparation for being stabilized in grout prior to operational tank closure. In each of these cases, characterization of the waste includes quantification of a wide set of radionuclides and stable constituents, with analysis costs typically on the order of hundreds of thousands of dollars and analysis durations on the order of months.

Thus far, nine SRS sludge batches have undergone extensive radionuclide characterization to quantify "reportable" radionuclides per the requirements of the Waste Acceptance Product Specifications (WAPS). In these characterizations, up to 66 different radionuclides were quantified, with the number of "reportable" nuclides being on the order of 30.[1-9] Per the WAPS, radionuclides contributing greater than or equal to 0.01 percent of the total curie content over the years 2015 to 3115 are required to be "reportable."[10]

In contrast, thus far, eight SRS salt batches have been qualified for the purpose of demonstrating effective treatment, and two dozen quarterly salt feed samples have been characterized for the purpose of determining Waste Acceptance Criteria (WAC) compliance. In the salt feed characterizations, 60+ different radionuclides were quantified, with almost all the nuclides found to be present at concentrations significantly below the WAC limits and targets.[11-34]

Waste characterizations at SRS have also included post-cleaning residue from a total of seven tanks, six of which are now operationally closed, and one of which is currently in the process of being closed. This includes one H-area waste tank (tank 16H, currently in the process of being closed)[35] and six F-area waste tanks (tanks 6F, 5F, 19F, 18F, 20F, and 17F, all of which have been closed).[36-41] Note that characterization of post-cleaning residue from a second H-area waste tank (tank 12H) is now in progress.[42] Also note that for Tank 16H, the annulus of the tank was characterized in addition to the tank interior, due to tank leakage which introduced waste material into the annulus.

In the recent tank closure characterizations (those completed in 2010 or after 2010), up to 59 radionuclides were quantified for the purpose of demonstrating compliance with the requirements of Section 3116 of the Ronald W. Reagan National Defense Authorization Act (NDAA) for Fiscal Year 2005. In contrast, in the characterizations supporting closure of the first two tanks (tanks 20F and 17F, which were closed in 1997), significantly fewer radionuclides were quantified via sampling and analysis, due to the less stringent reporting requirements of the time (prior to issue of the NDAA).

The availability of sample analysis data for multiple sludge, salt, and tank closure campaigns provides a frame of reference for evaluation of potential alternative characterization bases and for identifying potential concentration trends and/or magnitudes of normal concentration variations. However, for such comparisons to be meaningful, there must be confidence that the quality of the sample analysis data is high and that the magnitudes of the analytical uncertainties are known. For the vast majority of the SRNL sample analyses, the analytical uncertainties are reasonably low, typically on the order of twenty percent based on QA sample results and inter-method comparisons.

4.3 Tank Receipt Histories

Records of the types and quantities of waste received into a given waste tank over time provide a potential basis for understanding the variability of waste compositions in a given tank, as well as the expected ranges of constituent concentrations and the expected spatial distributions of constituents. Heterogeneity of waste constituents is particularly applicable to solid-phase waste, including sludge and post-cleaning residue. Because waste receipts typically occur in the absence of mixing, there is clearly the potential for early receipts into a tank to dominate the character of waste solids that are present at the very bottom of a tank, with later receipts dominating the character of the waste solids in the upper layer of solids. Similarly, intermediate receipts tend to dominate the character of the mid-layer solids. Of course, the magnitudes of the potential impacts are also a function of the relative quantities of waste that are received over time.

Although the distribution of liquid-phase constituents in a tank tends to be significantly more homogeneous than that of solid-phase constituents (due to the higher diffusion/equilibration rates), there is still utility in understanding the ranges of liquid-phase constituent concentrations carried with a given waste. Such understanding can provide a basis for quantifying liquid-solid phase constituent partitioning and it also can provide a basis for projecting liquid-phase compositions, such as those associated with dissolved salt waste.

At SRS, the quantities and compositions of sludge received into each tank over time have been recorded to address: a) the date of each waste receipt; b) the origin of each waste that was received; c) the type of each waste that was received; d) the age of each waste; e) the volume of each waste; and f) the composition of each waste. These records capture information applicable to nine primary types of waste, identifying the quantities of radionuclides and elemental compounds contained in each receipt. Specifically, thirty-seven radionuclides are quantified, including fission products, activation products, and actinides. Also quantified are thirty-seven elemental compounds, including compounds of iron, aluminum, uranium, thorium, calcium, manganese, nickel, mercury, silicon, and sixteen other elements. Quantification of the constituents contained in each waste receipt is based on an understanding of the compositions of each waste type.[43-47] In addition to receipts, the tank history records also address removals, as waste is transferred to alternate tanks and/or removed for dispositioning.

In the past, the SRS tank waste receipt history has been used for three primary purposes: 1) for demonstration of safe tank storage; 2) for planning of sludge processing at DWPF; and 3) for planning of salt processing and disposition at Saltstone.[48] In addition, the waste receipt history has also been used for tank closure planning purposes. More recently, the waste receipt records were used to identify general chemical and radioisotopic differences between sludge solids at the F and H area Tank Farms.[49] For each of these purposes, information from the waste receipt history has been utilized to quantify the average concentrations of waste constituents in a given tank, as well as the total inventories of constituents in a given tank. While such use of the information has been effective and continues to meet project needs, there does seem to be the potential to take advantage of the knowledge of the temporal distribution of the receipts, to better understand the waste variations, uncertainties, and impacts, and to hone the characterization expectations as a function of the waste's spatial location within the tank.

To date, utilization of the time-dependent differences in a tank's waste receipts has been very limited when characterizing a particular waste. However, there have been multiple occasions at SRS when the usefulness of such a characterization approach has been demonstrated. Two applicable examples are provided below.

Example 1: Key Constituents in Tank 12 Sludge

As previously mentioned, SRS Tank 12 has already undergone waste removal and chemical cleaning, and is currently being prepared for closure. Fortunately, samples collected prior to, during, and after bulk waste removal provide a source of data for comparison with expectations based on the waste receipt history. Brief descriptions of the samples that were collected and analyzed, the sampling timeline, and the waste characteristics identified in the tank receipt history are given below.

In 2008, prior to waste removal, solids from the top layer of accumulated sludge in Tank 12 were sampled and submitted to SRNL for characterization in support of aluminum dissolution processing.[50] These solids provide a basis for estimating the composition of the later waste receipts. Subsequently, as the Tank 12 waste removal operations began and progressed, a series of additional Tank 12 solids samples were collected and submitted to SRNL for measurements needed to support effective pumping/transfer operations. Unused portions of these samples were combined and mixed, and subsequently utilized in 2011 as a test medium for performing enhanced chemical cleaning experiments.[51] These solids provide a basis for estimating the composition of the intermediate waste receipts. In 2013, following completion of aluminum dissolution and bulk waste removal (but prior to oxalic acid cleaning), solids were collected from the floor of Tank 12, from an area where residual solids were found. These solids were submitted to SRNL, along with the subsequent tank cleaning solutions, for the purpose of determining the mass balance of the chemical cleaning operations.[52] These solids provide a basis for estimating the composition of the early waste receipts.

As identified in SRR's tank receipt history database,[53] the earliest material received into Tank 12 was ~41,000 kg of depleted uranium waste solids, received over the time period from October 1956 to November 1957. The U-235 isotopic enrichment of this early material was identified as being 0.62% (by weight), the Pu-238:Pu-239 ratio was identified as being 7.0 (by activity, after adjusting for radiological decay occurring since receipt), and the Al:Fe ratio was identified as being 0.13 (by weight). In contrast, the vast majority of waste received into Tank 12 after November 1957 was highly enriched uranium waste solids, with typical U-235 isotopic enrichments ranging from 40-80% (by weight), an average Pu-238:Pu-239 ratio of 44 (by activity, after adjusting for radiological decay), and an average Al:Fe ratio of 3.3 (by weight). Specifically, ~150,000 kg of this waste was identified as being received into Tank 12 over the time period from October 1963 to May 1973.

Given this history, one would expect that in the absence of mixing, solids in the bottom layer of sludge (at the tank floor) would have a U-235 isotopic enrichment of about 0.6%, a Pu-238:Pu-239 activity ratio of about 7, and an Al:Fe mass ratio of about 0.1. Similarly, in the absence of mixing, one would expect that solids in the top layer sludge (away from the floor) would have a U-235 enrichment of at least 40%, a Pu-238:Pu-239 activity ratio of about 40, and an Al:Fe mass ratio of about 3. (Note that U-235 enrichment, Pu-238:Pu-239 ratio, and Al:Fe ratio were chosen for this example because of their high variability in the Tank 12 waste and the fact that they are key constituents from the standpoints of material accountability, processing, and criticality).

Table 4-1 provides a comparison of the various values based upon the waste receipt history and the sample analysis data referenced above. As previously described, three general types of Tank 12 samples are addressed, those collected from the top layer of sludge (from the aluminum dissolution study),[50] those collected from the middle layer of sludge (from the waste removal activities),[51] and those collected from the bottom layer (from the investigation targeting mass balance of the chemical cleaning activities).[52] As shown in the table, expectations based on the receipt records are given solely for the top layer (late receipts) and the bottom layer (early receipts), with expectations for the middle layer omitted, but expected to fall somewhere between those for the top and bottom layers.

Table 4-1. Comparison of Tank 12 Analytical Data with Expectations Based Upon Receipt Records

Waste Descriptor	U-235 Enrichment (Mass %)		Pu-238:Pu-239 Activity Ratio		Al:Fe Mass Ratio	
	Receipt Records	Sample Analysis	Receipt Records	Sample Analysis	Receipt Records	Sample Analysis
Top layer (late receipts)	≥40	11	44	36	3.3	10
Middle layer (intermediate receipts)		1.8		26		5.8
Bottom layer (early receipts)	0.62	0.73	7.0	13	0.13	0.34

In all cases, there are differences between the values determined by sampling and analysis and the values based upon the receipt records. However, the trends of the sample data are clearly consistent with the trends of the waste receipt history – namely, that U-235 enrichments increase from the bottom to the top. For example, the receipt records suggest that the U-235 enrichment in the top layer should be at least 40%, while the sample data indicate it is significantly lower at 11%. In contrast, the receipt records suggest that the U-235 enrichment in the bottom layer should be about 0.6%, while the sample data indicate it is slightly higher at about 0.7%. Additional sample data indicate that the U-235 enrichment in the intermediate layer is between those of the top and bottom layers (at about 2%), which makes sense, given the expected upward trend as one moves from the bottom layer to the top. Of course, knowing that mixing occurred when the solids were slurried during the sampling and waste removals operations, there is clearly the expectation that some "averaging" of the waste occurred. Such "averaging" may be responsible for some of the differences observed.

In the case of the Pu-238:Pu-239 ratios, the differences between the measured values and the values based on receipt history were smaller, with values in the top layer varying by about 20% (36 versus 44) and values in the bottom layer varying by a factor of approximately two (13 versus 7). As in the case of the U-235 enrichment, the Pu-238:Pu-239 ratio for the middle layer was intermediate between that of the top and bottom layers (26 for the middle layer), which makes sense, given the expected upward trend as one moves from the bottom layer to the top. The smaller relative difference observed for the Pu-238:Pu-239 ratio case versus the U-235 enrichment case is likely due to the smaller relative variability of Pu-238:Pu-239 ratios.

In the case of the Al:Fe ratios, the measured values were about three times the expected values based on the receipt histories, both for the top layer and the bottom layer. Specifically, the measured ratios for the top and bottom layers were 10 and 0.34, respectively, as opposed to 3.3 and 0.13 based on the receipt records. As in the cases of U-235 enrichment and Pu-238:Pu-239 ratio, the Al:Fe ratio measured for the middle layer (5.8) was intermediate between that of the top and bottom layer, which is consistent with expectations. The fact that the measured Al:Fe ratios for both top and bottom layers were consistently higher than the values indicated by the receipt records may be reflective of: a) higher concentrations of aluminum in the waste than anticipated; b) lower concentrations of iron in the waste than anticipated; and/or c) physical partitioning of aluminum compounds and iron compounds. Regardless of the reason, it is clear that deviations between sample results and expectations will occur and must be considered when applying a characterization strategy.

Example #2: Key Constituents in Tank 18 Post-Cleaning Floor Residue and Wall Material

As Tank 18 was being prepared for closure (following bulk removal and cleaning), multiple residue samples were collected from the floor and wall of the tank, and subsequently submitted to SRNL for extensive characterization. This includes six floor residue samples, two wall corrosion product samples, and one wall scale sample. The analytical results were reported in two separate documents, one for the floor samples[39] and one for the wall samples.[54]

In a data quality assessment performed by SRR,[55] a portion of the analytical data for the Tank 18 floor samples was compared to expectations based on the tank receipt histories. Specifically, the comparison addressed constituent concentrations for Al, Fe, Hg, Mn, U, Tc-99, U-235, U-238, Pu-239, and Pu-240, and radioisotope ratios for U-235:U-238 and Pu-240:Pu-239. A summary of the compared values is given below in Tables 4-2 and 4-3, both tables taken from SRR's data quality assessment report.[55]

Table 4-2. Comparison of Constituent Concentrations Based on Tank 18 Receipt History and Sample Analysis Results [55]

Constituent	Units	Receipt History Concentrations		Measured Concentrations in Samples				
		Range	Mean	18-1	18-2	18-3	18-4	18-5
Al	wt%	0 to 3.3E+01	6.7E+00	1.1E+01	1.4E+01	1.7E+01	1.7E+01	1.6E+01
Fe	wt%	0 to 2.8E+01	2.7E+01	9.8E+00	7.0E+00	7.3E+00	7.5E+00	8.8E+00
Hg	wt%	0 to 7.2E-02	6.8E-02	7.2E-02	3.5E-02	2.5E-02	5.0E-02	6.3E-02
Mn	wt%	0 to 1.3E+00	1.2E+00	1.0E+00	8.3E-01	7.9E-01	9.7E-01	1.1E+00
U	wt%	1.4E+00 to 7.8E+01	3.2E+00	6.3E+00	3.7E+00	1.6E+00	1.2E+00	8.3E-01
Tc-99	µCi/g	0 to 2.7E-01	1.1E-01	2.7E-02	3.3E-02	3.2E-02	4.5E-02	4.5E-02
U-235	µCi/g	4.5E-05 to 1.0E-02	2.2E-04	8.0E-04	4.4E-04	1.7E-04	1.3E-04	8.4E-05
U-238	µCi/g	4.8E-03 to 2.6E-01	1.1E-02	2.0E-02	1.1E-02	4.3E-03	3.4E-03	2.1E-03
Pu-239	µCi/g	3.3E+00 to 9.3E+02	6.6E+00	1.6E+01	9.0E+00	6.6E+00	6.8E+00	7.6E+00
Pu-240	µCi/g	7.3E-01 to 2.1E+02	1.7E+00	3.6E+00	2.1E+00	1.5E+00	1.6E+00	1.8E+00

Table 4-3. Comparison of Constituent Ratios Based on Tank 18 Receipt History and Sample Analysis Results [55]

Constituent Ratio (Ci/Ci)	Receipt History Ratio		Ratio Based on Sample Measurements					
	Range	Mean	18-1	18-2	18-3	18-4	18-5	18-8
U-235/U-238	7.7E-03 to 4.9E-02	2.0E-02	4.0E-02	4.0E-02	4.0E-02	3.8E-02	4.0E-02	3.8E-02
Pu-240/Pu-239	2.2E-01 to 2.6E-01	2.5E-01	2.3E-01	2.3E-01	2.3E-01	2.4E-01	2.4E-01	2.3E-01

These comparisons indicate that in the vast majority of cases, there is good agreement between the sample analysis results and the values based on receipt history. Specifically, in almost every case, the measured values were within the range specified by the receipt history and the magnitudes of the measured values were consistent with those of the averages indicated by receipt history. The only exceptions included a couple of the U mass and U-238 activity cases, where the measured concentrations were somewhat lower than the minimum concentrations indicated by the receipt history (lower by up to a factor of two). Given that U-238 is the dominant isotope driving Tank 18 uranium mass (the uranium in Tank 18 is depleted), it is expected that the trends of the U mass and the U-238 activity would track one another. Despite the uranium inconsistencies, it is clear that on the whole, the measured constituent concentrations in the floor residue samples were relatively consistent with the values indicated by the waste receipt history.

With respect to the Tank 18 wall samples, use of the waste receipt history provides the rationale for understanding why the Pu-238:Pu-239 ratio increases with wall elevation. Specifically, the receipt history identifies that the first 17,000 kg of solids received into Tank 18 contained ~600 Ci of Pu-239, with no significant Pu-238 (therefore, a Pu-238:Pu-239 ratio of zero). These receipts occurred between

August 1959 and March 1961. In contrast, over the later time period (from October 1966 to April 1997), an additional 470,000 kg of solids were received into the tank, with a Pu-239 content of ~2700 Ci and a Pu-238 content of ~18,000 Ci (corresponding to a Pu-238:Pu-239 activity ratio of about seven). Note that the plutonium activities in the receipt history have been adjusted for radiological decay). Given this history, it is expected that post-cleaning residue at the very bottom of the tank would contain minimal Pu-238, while higher elevation wall material that had been in contact with the later waste would contain significantly higher fractions of Pu-238, dependent on the relative contact times associated with the early and later receipts. Given the distribution of Pu-239 and Pu-238 reported in the waste receipt history, the average Pu-238:Pu-239 activity ratio for the later material should be about seven (taking decay into account).

A summary of the Pu-238:Pu-239 activity ratios based on the receipt records and the applicable sample analysis results is given in Table 4-4. As already discussed, the Tank 18 receipt records indicate that the Pu-238:Pu-239 activity ratios for the top layer of solids (late receipts) and the bottom layer of solids (early receipts) should be about 7 and 0, respectively. In contrast, wall samples collected at elevations of 17, 10-12, and 6-7 feet were found to have Pu-238:Pu-239 activity ratios of 19, 8.6, and 1.6, respectively; and floor samples from the tank bottom were found to have Pu-238:Pu-239 activity ratios averaging at 0.43.

Table 4-4. Pu-238:Pu-239 Activity Ratios Based on Tank 18 Receipt Records and Measured Values

Waste Description	Elevation, feet	Pu-238:Pu-239 Activity Ratio	
		Receipt Records	Measured Value
Top layer of solids		7	
Upper wall sample	17		19
Lower wall sample	10-12		8.6
Wall scale sample	6-7		1.6
Floor residue samples (average)	0		0.43
Bottom layer of solids		0	

Although two of the measured ratios (19 and 8.6) are higher than the maximum ratio identified by the receipt records (7), the trend of the data is still consistent with expectations – namely, that ratios at higher elevations are significantly larger than ratios at lower elevations, and ratios at lower elevations approach zero. Despite the absolute differences, all of the measured values are either within the range of the ratios indicated by the receipt records (0-7) or the same order of magnitude as the maximum ratio indicated by the receipt records (7). Clearly, use of the receipt records has limitations – however, it can certainly aid understanding of spatial variations within waste and when used wisely, can provide order or magnitude estimates of key waste constituent concentrations and/or constituent distributions.

4.4 Tank Waste Process Knowledge

Significant understanding of SRS tank waste compositions and variations has been developed through decades of waste handling, processing, characterization, and disposition activities. This includes understanding of the quantities and distributions of many key radionuclides, namely fission products, activation products, and actinides.[47,56-74] Such understanding comes from several sources, including data drawn from sample analyses, waste processing experiences, waste compositional studies, and theoretical relationships.

Coupling of data identifying the compositions of waste from the site separation facilities and tracking of the waste transfers occurring between tanks (or to applicable waste treatment facilities) provides a basis for quantifying the inventories of waste constituents stored in each tank, as well as quantifying the average constituent concentrations associated with each tank's waste. Such information has been consolidated in SRR's Waste Characterization System (WCS), an electronic information system targeting quantification of approximately ninety stable constituents and forty radionuclides.[75] Over time, WCS has been continuously updated to incorporate new data, as applicable, and to make modifications necessary to support new characterization needs.[76-79] This has resulted in the availability of waste characterization data for over fifty key radionuclides and over 100 elemental constituents, ions, and compounds. The current version of WCS is designated WCS 1.5, version 3.1.[80]

WCS is used for multiple site characterization purposes, such as: a) planning and scheduling of SRS tank waste system activities (which include receipt of waste from the canyons, transfer of waste within the Tank Farm, evaporation of supernatant phase waste, and processing of sludge and salt batches); b) calculating radiolytic heat loads and hydrogen generation rates; c) estimating source terms for accident analyses; d) estimating residual inventories for waste removal planning; and e) providing input for the annual DOE Integrated Database. Over its twenty year history, WCS has proven effective for these purposes – however, there is recognition that the system has limitations, due to the inherent variations and uncertainties associated with tank waste.

In 2002, an evaluation was performed to determine the consistency of constituent concentrations determined through sampling and analysis with those estimated by WCS.[81] Specifically, sampling and analysis data for two SRS sludge batches (SB1a and SB1b) were compared with WCS projections based on the expected distributions of constituents in the waste feeding the sludge batches (sludge waste from Tanks 8, 17, 19, 20, 40, and 42). A total of twenty-three radionuclides and twenty elemental constituents were addressed. This included: a) Co-60, Sr-90, Tc-99, Ru-106, Sb-125, Cs-134, Cs-137, Ce-144, Pm-147, Eu-154, U-233, U-234, U-235, U-236, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, and Cm-244; and b) Ag, Al, Ba, Ca, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Na, Ni, Pb, Si, Sr, Zn, and Zr.

On average, the WCS projections were relatively consistent with the sample analysis data. In the case of the radionuclides, the WCS values averaged 27% higher than the measured values. In contrast, in the case of the elemental constituents, the WCS values averaged 10% lower than the measured values. On a constituent by constituent basis, the range of deviations was higher, with some projections differing from measurements by two to three orders of magnitude. However, on the whole, the majority of deviations were one order of magnitude or less.

Specifically, the radionuclide deviations were within an order of magnitude 84% of the time, and the elemental deviations were within an order of magnitude 82% of the time. Clearly, these comparisons cannot be assumed representative of all sludge batches – and should not be considered definitive due to the inherent sampling and analysis uncertainties. However, they do provide an example of the extent that WCS projections will deviate from sample analysis data – but this example suggests that on the whole, the deviations were limited to one order of magnitude or less. Considering the inherent variability of the waste, the ranges of such deviations are not unexpected.

In 2005, after processing the first four sludge batches at DWPF (SB1a, SB1b, SB2, and SB3), a determination was made that the quantities of sludge solids projected by WCS were 56% lower than the quantities actually received at DWPF. Because of this difference, an investigation was performed to analyze the WCS sludge solids projections on a tank by tank basis, and on a waste type basis, focusing on the waste material comprising the first four sludge batches.[82] Results of the investigation led to the conclusion that underestimates of the quantities of $Fe(OH)_3$, $Al(OH)_3$, and MnO_2 (as indicated by WCS) were responsible for the lower mass of sludge sent to DWPF. Specifically, the investigation determined that the average quantities estimated by WCS were low by factors up to 2, 3, and 2, respectively [for $Fe(OH)_3$, $Al(OH)_3$, and MnO_2]. In the case of aluminum, WCS being low is consistent with the Tank 12H data comparison example presented in Table 4-1 and the Tank 18F data comparison example presented in Table 4-2. However, in the case of iron, the Tank 12H and Tank 18F data comparisons suggest otherwise – namely that the WCS values were high by a factor of 2-3 – and in the case of manganese, the Tank 18F WCS value was about right. Regardless of these differences, the sludge batch comparisons provide additional confidence that the WCS projections are usually the appropriate order of magnitude, although likely prone to greater uncertainties than typical sample analysis data.

Still, the use of such "process knowledge" data may be well suited for certain characterization applications, particularly those where high accuracy is not the primary concern.

4.5 Theoretical Scaling Factors

Use of scaling factors offers a potential means of estimating radionuclide concentrations based on process knowledge, radiological properties, solubility characteristics, and waste chemistry. Potential scaling factors are not available for all radionuclides, only those that meet criteria assuring consistency of radionuclide production, processing, and waste chemistry. Non-laboratory characterization methods such as scaling are important from the perspective of demonstrating analytical results are consistent with expectations, and for estimating radionuclide concentrations when a laboratory technique is unavailable, ineffective, or particularly cost prohibitive.

When assessing the possibility of utilizing scaling, it is typically assumed that processing has not changed the distribution of radionuclides from that in the original waste. In other words, no waste radionuclides have been selectively concentrated, removed, and/or redistributed through supplemental chemical recovery and/or operational processes. If such processes have been performed, the scaling assumptions may no longer be valid, and correspondingly, the application of scaling factors may no longer be appropriate. For this reason, scaling is typically less conducive to characterizing liquid-phase radionuclides than solid-phase radionuclides, due to the propensity for changing the liquid-phase elemental distributions through supernatant transfers, mixing, and/or evaporation processes. However, a couple of exceptions apply. One exception applies to isotopes of the same element, which will have identical chemical behavior and therefore can migrate in solution together, given that conditions support such behavior. Another exception applies to highly soluble radionuclides, which fully partition to the liquid phase (or near-fully partition to the liquid phase) and have the potential to migrate together in solution.

Three primary categories of radionuclide scaling factors are utilized at SRS: 1) those based on known radionuclide ratios; 2) those based on quantities of parent nuclides and age of the waste; and 3) those based on relative fission yields and atomic masses (as determined through application of fission yield scaling factors).

In the first category, radionuclide ratios are developed through available data and/or theoretical relationships, and then typically applied to estimate a concentration of one radionuclide based on a measured concentration of another radionuclide. Examples of usages at SRS include scaling of: a) Ni-59 based on Ni-63 (applicable to both solid-phase and liquid-phase wastes); [74,83,84] b) Tc-99 based on Cs-137 (applicable to liquid-phase waste); [72] c) Pd-107 based on Pd-105 (applicable to both solid-phase and liquid-phase waste); [8] d) I-129 based on Cs-137 (applicable to liquid-phase waste); [65] d) Sn-121m based on Sn-126 (applicable to solid-phase waste); [8] e) Cs-135 based on Cs-137 (applied to solid-phase and waste); [8,84] and f) Th-230 based on Th-232 (applicable to solid-phase and liquid-phase wastes).[56,74,84]

In the second category, concentrations of in-grown radionuclides are estimated based on known concentrations of the parent nuclides and the age of the waste. The concentrations of the parent nuclides are identified through sample analysis or an alternate technically-defendable basis. The age of the waste is typically estimated through process knowledge or through use of an applicable fission yield scaling factor.[8] Examples of usages at SRS include scaling of: a) Nb-93m based on Zr-93;[84] b) Ra-226 based on Th-230, U-234, and Pu-238;[84] c) Ra-228 based on Th-232;[84] d) Ac-227 and Pa-231 based on U-235;[68] and e) Th-229 based on U-233.[84] In all of these cases, the scaling was applied to solid-phase waste.

In the third category, concentrations of applicable fission products are estimated based on the concentrations of other applicable fission products, as typically measured by Inductively-Coupled Plasma Mass Spectrometry (ICP-MS)[8] or other mass spectrometry technology. Specifically, an average Fission Yield Scaling Factor (FYSF) is determined based upon a set of individual FYSFs applicable to isotopes meeting the following five criteria:

- The isotopes have low solubility in NaOH (and thus occur predominately in sludge) or will scaled against other fission product isotopes of the same element
- The isotopes have long half-lives and thus have not decayed significantly since the waste was generated
- The isotopes have low neutron cross sections and thus were not transmuted in the reactor during operations
- The isotopes could not be formed in the reactor by neutron absorption
- The isotopes have masses where interferences such as those from rare earth oxides formed in the argon plasma do not create a problem

Individual FYSFs for each applicable isotope are computed using the following relationship:

$FYSF_i = wt\%_i \div (FY_i * AM_i)$,

where $wt\%_i$ is the weight percent of isotope i (as measured by mass spectrometry), FY_i is the fission yield of isotope i, and AM_i is the atomic mass of isotope i. Since the primary sources of fission products in SRS waste are from fission of U-235, the bulk of the SRS cases utilize U-235 fission yield data. Alternately, Pu-239 or U-223 fission yield data would be utilized in the cases where fission products have been generated from Pu-239 or U-233, respectively.

The average FYSF is then used to estimate the concentrations of the unknown isotopes, based on the following relationship:

$Wt\%_i = FYSF_{avg} * FY_i * AM_i$

Examples of FYSF usages at SRS include scaling of: a) Se-79;[8] b) Zr-93;[84] c) Cd-113m;[84] d) In-115;[8] e) Sn-126;[8,84] and f) Sm-151.[84] In all of these cases, the scaling was applied to solid-phase waste (as inferred by the first "bulleted" criterion identified above).

In cases where the "scaled" radionuclide concentrations were compared to values determined through sampling and analysis, the scaled values were found to be reasonably consistent with the measured values.[84] For example, in comparisons of data applicable to characterization of Sludge Batch 7b, the scaled Zr-93 concentration was two to three times the analytical value, the scaled Cs-135 was about 10% lower than the analytical value, and the scaled Sm-151 was about 40% higher than the analytical value.

4.6 Historic Data Trends

Historic characterization data collected over an extended period of time provides a direct means of quantifying compositional variations and identifying potential constituent concentration trends. At SRS, a range of applicable data are currently available, through the extensive characterization work that has been performed in support of sludge processing, salt processing, and tank closure. As previously identified, available data include characterization results for several dozen radionuclides and stable constituents in the past nine sludge batches, the past six years of salt batch qualification and quarterly salt feed samples, and the post-cleaning residue associated with the five most recent waste tanks prepared for closure (four of these tanks are now closed and one is in the final stages of closure preparations). Given below is a summary of the radionuclide data associated with these campaigns, by waste type (sludge, salt feed, and post-cleaning residue). Note that the salt qualification characterization data have not been included in the summary, for the purposes of brevity.

Specifically, characterization results for twenty key radionuclides are captured in the tables, and characterization results for a smaller group of key radionuclides are captured in the plots. The twenty key radionuclides addressed in the tables include Ni-59, Ni-63, Sr-90, Zr-93, Tc-99, I-129, Cs-135, Cs-137, Sm-151, Th-232, U-233, U-234, U-235, U-238, Np-237, Pu-238, Pu-239, Am-241, Am-243, and Cm-244. These were selected because of their ubiquitousness in tank waste. In contrast, the key radionuclides addressed in the plots were specific to each waste type, based upon perceived significance of potential impacts (such as personnel dose, criticality concerns, and long-term environmental risks).

For the sludge batch cases, the radionuclide data that are presented are those identified in the WAPS reports as the present day input concentrations feeding the long-term decay calculations. For the salt feed cases, the data that are presented are the average radionuclide concentrations for each calendar year, based upon the quarterly results. In contrast, for the post-cleaning residue, the data that are presented are the average radionuclide concentrations for each tank, based on all the individual samples that were analyzed or the composited samples that were analyzed, as applicable. In cases where the original characterization data contained a combination of detectable concentrations and non-detectable concentrations, only the detectable concentrations were considered. In contrast, in cases where the original characterization data contained solely non-detectable concentrations, the data that are presented in the tables are the averages of the minimum detectable concentrations. Note that in the case of the Tank 16 residue, two sets of characterization data are reported – those for the interior of the tank (abbreviated INT) and those for the annulus (abbreviated ANN).

In addition to providing the radionuclide concentrations for each waste type and campaign, the tables also identify the span of concentrations associated with each radionuclide in each waste type. Specifically, the "span of concentrations" provides an indication of the radionuclide concentration variability. For a given radionuclide and waste type, the span of concentrations is defined as the ratio of the maximum radionuclide concentration divided by the minimum radionuclide concentration. In performing this calculation, the maximum concentration is the highest detectable concentration (as opposed to a concentration that is less than a minimum detection limit), and the minimum concentration is the lowest detectable concentration or minimum detection limit. In cases where the minimum concentration is designated by a minimum detection limit, the span of concentrations is preceded by an "approximately" sign, since the minimum concentration is actually "less than" the minimum detection limit identified.

4.6.1 SRS Sludge Batches

Measured concentrations of key radionuclides in SRS sludge batches 1b through 8 are given in Table 4-5. Note that these characterizations were performed between the years of 1999 and 2014. As shown in the table, the radionuclide concentrations vary over nine orders of magnitude, from a low concentration of ~8E-06 µCi/g I-129 in SB2 to a high concentration of ~2E+04 µCi/g Sr-90 in SB5. The variations were considerably smaller for each individual radionuclide, where the spans of concentrations ranged from 3x for U-235 to 340x for I-129. Of the twenty key radionuclides addressed, ten had concentration spans less than an order of magnitude (10x), including Sr-90, Tc-99, Cs-135, Cs-137, Sm-151, U-238, Np-237, Pu-239, and Am-241 (as well as U-235). Of these, the concentration spans for five radionuclides were limited to 3x-4x, indicative of general processing uniformity and consistency. The radionuclides in this group included Tc-99, Sm-151, U-235, U-238, and Pu-239. The radionuclides with the largest concentration variations included I-129 (340x), Th-232 (81x), and Am-243 (77x). The high I-129 variation is an impact of the unusually low concentration found in SB2, which is two orders of magnitude below the concentrations found in the other sludge batches. The reason for this low concentration is not known, but may be due to volatility impacts in the canyons during processing. In contrast, the high variability of Th-232 and Am-243 is attributed to their association with special processing campaigns. As such, their distribution in waste varies considerably from tank to tank.

Figure 4-1 provides a visual depiction of the consistency of concentrations for select radionuclides with limited concentration spans in the sludge batches – specifically Sr-90, Cs-137, U-235, Np-237, and Pu-239. Note that the slightly increased Sr-90, Cs-137, and Pu-238 concentrations occurring in the later sludge batches may correlate with the onset of salt processing, where the Sr/Cs/Pu waste streams were introduced into the sludge batches. Regardless, based on the plotted data, one can easily visualize an upper concentration bound, a lower concentration bound, and an average centerline concentration for each of these radionuclides. Typically, the upper bound in these cases is about twice that of the centerline concentration, and the lower bound is about half that of the centerline concentration. For future sludge batches comprised of similar source material, such characterization information provides a basis for estimating the corresponding ranges of concentrations that can be expected. Although the estimates will have clear uncertainties, it is likely that such an approach will provide some concentration estimates that are very good (within a factor of two or three), some that are moderately good (within an order of magnitude), and others that are too uncertain to be useful.

Table 4-5. Measured Concentrations of Key Radionuclides in Sludge Batches 1b through 8

Radionuclide	Radionuclide Concentration, µCi per gram of total solids									Span of Concentrations
	SB1b	SB2	SB3	SB4	SB5	SB6	SB7a	SB7b	SB8	
Ni-59	4.78E-02	3.13E-01	6.63E-01	5.67E-01	1.30E+00	1.05E+00	1.50E+00	2.57E+00	7.53E-01	54x
Ni-63	8.42E+00	2.05E+01	5.41E+01	6.58E+01	1.11E+02	1.08E+02	2.50E+01	1.60E+02	9.34E+01	19x
Sr-90	4.24E+03	4.52E+03	4.85E+03	7.23E+03	2.25E+04	1.87E+04	1.40E+04	1.27E+04	9.85E+03	5x
Zr-93	8.22E-02	1.36E-01	4.43E-02	2.64E-01	5.23E-01	4.59E-01	5.30E-01	4.63E-01	7.02E-01	16x
Tc-99	1.88E-01	1.26E-01	1.96E-01	1.41E-01	1.53E-01	<1.09E-01	1.00E-01	9.67E-02	3.43E-01	4x
I-129	1.09E-03	7.57E-06	<4.35E-04	5.20E-04	<3.77E-04	2.56E-03	1.70E-03	1.06E-03	2.04E-03	340x
Cs-135	7.54E-04	1.08E-03	1.34E-03	1.18E-03	1.61E-03	1.85E-03	2.80E-03	3.32E-03	4.55E-03	6x
Cs-137	1.59E+02	2.75E+02	3.15E+02	2.49E+02	3.23E+02	3.87E+02	5.90E+02	6.44E+02	8.52E+02	5x
Sm-151	6.32E+01	1.77E+02	1.96E+02	1.13E+02	1.82E+02	2.62E+02	2.30E+02	2.47E+02	1.63E+02	4x
Th-232	3.41E-04	3.76E-05	3.01E-05	5.07E-05	7.28E-05	2.44E-03	1.60E-03	1.19E-03	8.95E-04	81x
U-233	4.35E-02	1.07E-02	1.56E-02	<1.91E-02	2.22E-02	8.98E-02	1.30E-01	4.22E-02	6.00E-02	12x
U-234	2.89E-03	3.57E-02	3.03E-02	3.40E-02	6.15E-02	8.37E-02	4.80E-02	4.06E-02	4.39E-02	29x
U-235	2.81E-04	6.57E-04	7.29E-04	6.41E-04	7.00E-04	6.01E-04	6.40E-04	6.12E-04	5.41E-04	3x
U-238	6.46E-03	2.53E-02	2.26E-02	1.77E-02	1.67E-02	1.22E-02	1.50E-02	1.60E-02	1.25E-02	4x
Np-237	1.29E-02	1.33E-02	3.30E-02	2.60E-02	6.92E-02	3.61E-02	2.20E-02	2.46E-02	2.50E-02	5x
Pu-238	9.95E+01	3.85E+01	2.59E+01	1.22E+02	2.76E+02	3.83E+02	1.80E+02	1.42E+02	1.99E+02	15x
Pu-239	4.89E+00	7.68E+00	1.37E+01	1.03E+01	2.04E+01	1.82E+01	1.30E+01	1.08E+01	7.78E+00	4x
Am-241	6.65E+00	3.27E+01	1.85E+01	1.76E+01	3.25E+01	3.29E+01	3.70E+01	3.56E+01	2.74E+01	6x
Am-243	5.65E-02	5.34E-01	1.55E+00	1.64E+00	9.64E-01	4.33E+00	5.70E-01	4.99E-01	1.88E+00	77x
Cm-244	4.43E+00	4.42E+01	7.00E+01	9.07E+01	4.61E+01	1.49E+02	1.90E+01	1.81E+01	6.81E+01	34x

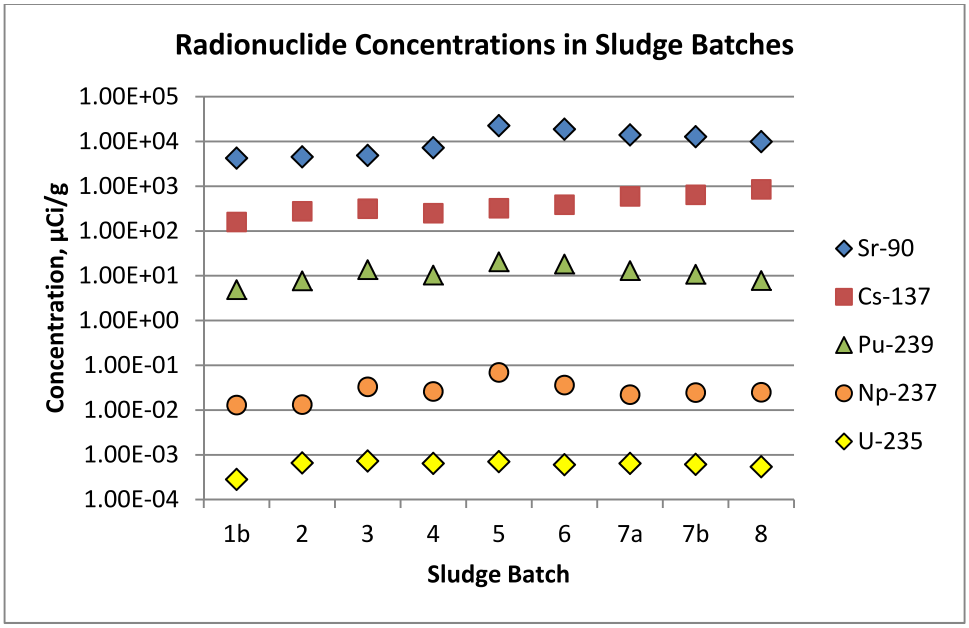


Figure 4-1. Concentrations of Select Radionuclides in SRS Sludge Batches 1B through 8

4.6.2 SRS Tank 50 Salt Feed

Average concentrations of key radionuclides in Tank 50 Salt Feed between the years of 2008 and 2014 are given in Table 4-6. As shown in the table, the radionuclide concentrations vary over almost ten orders of magnitude, from a low concentration of less than ~1E-03 pCi/mL Th-232 in CY 2014 to a high concentration of ~2E+07 pCi/mL Cs-137 in CY 2008. The variations were significantly smaller for the individual radionuclides, where the spans of concentrations ranged from ~1.3x for Np-237 to 170x for Pu-239. It may come as a surprise that the Pu-239 concentration in salt feed is so variable, when one considers the relative consistency of Pu-239 in the sludge batches (4x span). However, the significant drop in the Pu-239 concentration that has occurred since 2009 is an indicator of the onset of the actinide removal process (ARP) salt treatment, which removes soluble plutonium through monosodium titanate (MST) sorption and removes plutonium-containing entrained solid particles through cross-flow filtration.

Seven of the key radionuclides in salt feed had concentration spans less than an order of magnitude (10x), including Tc-99, I-129, Th-232, U-234, U-235, and U-238 (as well as Np-237). Of these, five radionuclides had concentration spans of 3x or less, demonstrating relatively high uniformity in waste. The relatively high variation of the Cs-137 concentration (18x) is attributed to the impacts of initiating salt processing (specifically cesium removal by caustic side solvent extraction), which have significantly reduced the cesium concentrations of the later batches. Regardless of the magnitudes of concentration spans for the 2008 to 2014 period, it is clear that in the vast majority of cases, the year to year concentration variation was an order of magnitude or less. This is apparent in Figure 4-2, where concentrations of select radionuclides are plotted as a function of the calendar year. In addition to the year to year variation, this figure also illustrates the trends of the concentrations. Such trends provide a basis for predicting whether future concentrations will be higher or lower, and for estimating by how much. For example, in the case of Cs-137, it can be seen that concentrations dropped by a factor of about twenty over the time period from 2008 to 2013. However, since 2013, the concentration has increased slightly, suggesting that the 2015 concentration could be slightly higher than that of 2014. Addition of a small volume of untreated salt waste into the 2014 feed is a likely source of the 2014 increase. Based on qualification testing, the expected cesium concentration of the next salt batch feed (salt batch 8) should increase by a factor of about four.

Table 4-6. Average Concentrations of Key Radionuclides in Tank 50 Salt Feed

Radionuclide	Mean Radionuclide Concentration, pCi/mL							Span of Concentrations
	2008	2009	2010	2011	2012	2013	2014	
Ni-59	<1.36E+02	<1.34E+01	<2.67E+00	<1.42E+01	<1.22E+01	<1.03E+01	<8.31E+00	N/A
Ni-63	<8.24E+01	7.11E+01	2.44E+02	5.29E+01	<7.41E+00	<1.01E+01	<5.49E+00	~44x
Sr-90	2.45E+05	1.06E+05	6.64E+04	3.57E+04	3.46E+03	3.55E+03	4.09E+03	71x
Zr-93	NM	NM	NM	NM	<7.40E+01	<5.47E+01	<1.50E+02	N/A
Tc-99	2.68E+04	2.69E+04	3.04E+04	3.29E+04	2.63E+04	1.95E+04	1.95E+04	2x
I-129	<4.17E+00	5.43E+00	5.92E+00	5.37E+00	1.04E+01	1.28E+01	1.37E+01	~3x
Cs-135	1.68E+02	9.05E+01	5.31E+01	4.51E+01	<4.06E+01	<2.50E+01	<1.73E+01	~10x
Cs-137	2.31E+07	1.33E+07	7.50E+06	3.00E+06	1.54E+06	1.26E+06	1.75E+06	18x
Sm-151	<2.33E+03	5.98E+02	6.38E+02	<5.62E+02	2.51E+01	<3.21E+01	<2.97E+01	25x
Th-232	1.13E-02	9.62E-03	9.47E-03	4.43E-03	<3.84E-03	<2.18E-03	<1.28E-03	~9x
U-233	<2.24E+02	<1.13E+02	<2.55E+02	<6.72E+02	<2.86E+02	<1.70E+02	<1.13E+02	N/A
U-234	1.46E+02	2.00E+02	1.45E+02	9.28E+01	<1.84E+02	<1.10E+02	<8.36E+01	~2x
U-235	1.70E-01	3.41E-01	3.74E-01	3.58E-01	2.55E-01	1.76E-01	1.76E-01	2x
U-238	7.79E-01	1.23E+00	1.60E+00	3.51E+00	5.00E+00	3.97E+00	3.90E+00	6x
Np-237	<1.63E+01	1.08E+01	9.48E+00	<3.16E+01	<2.07E+01	<2.56E+01	<8.22E+00	~1.3x
Pu-238	3.41E+04	9.59E+03	1.87E+04	8.49E+03	1.00E+03	1.22E+03	9.42E+02	36x
Pu-239	1.07E+03	1.06E+04	9.36E+02	5.32E+02	1.49E+02	8.68E+01	6.36E+01	170x
Am-241	7.75E+02	5.51E+02	1.09E+03	4.56E+02	1.39E+01	7.79E+00	1.80E+01	140x
Am-243	2.05E+01	1.49E+01	1.52E+01	5.70E+01	<8.35E-01	<6.62E-01	<4.76E-01	~120x
Cm-244	2.23E+03	1.29E+03	2.41E+03	9.19E+02	1.34E+02	1.94E+01	1.12E+02	120x

NM = not measured

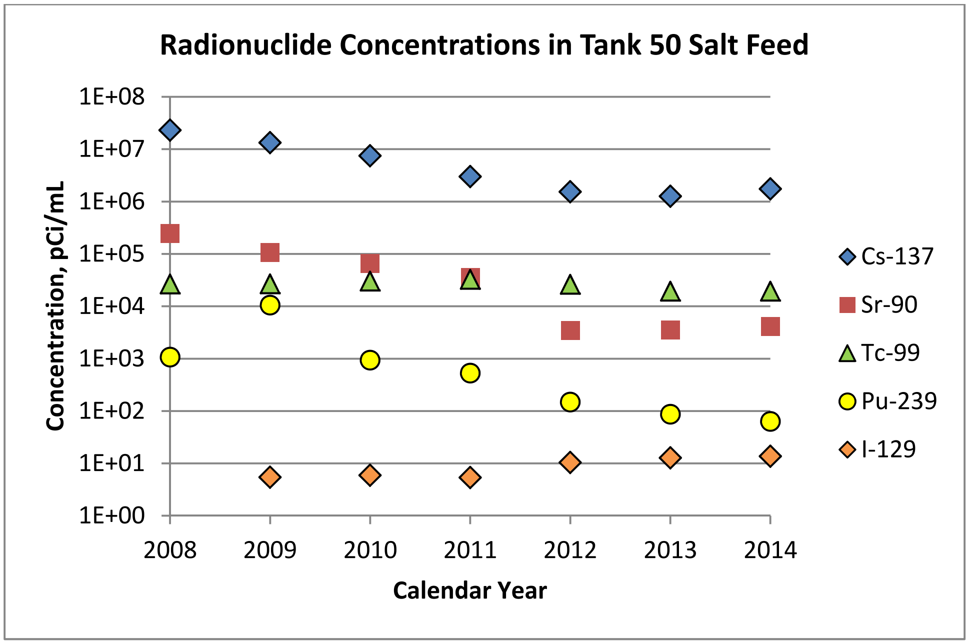


Figure 4-2. Concentrations of Select Radionuclides in SRS Tank 50 Salt Feed

Another example is the I-129 concentration, which was relatively constant between 2009 and 2011, increased by a factor of about two between 2011 and 2012, and then remained relatively constant between 2012 and 2014, with increases during the 2012-2014 period being limited to about 20-30%. Based on these data, one would expect that the 2015 concentration of I-129 would be close to that of 2014 or slightly higher. In contrast, the Tc-99 concentration has remained relatively stable from 2008 to 2014. As such, the 2015 Tc-99 concentration is expected to be similar to all those occurring before. It should be noted that prior to MCU treatment, the supernatant phase ratios of I-129 to Cs-137, and Tc-99 to Cs-137, have been reported to be relatively constant for a given point in time.[65,72] However, because of the relatively short half-life of Cs-137 (30 years) compared to the relatively long half-lives of I-129 and Tc-99 (~2E+07 and ~2E+05 years, respectively), the ratios will increase over the timeframe of the salt program.

Although there is inherent uncertainty in predicting future concentrations based on past history, the complete range of concentrations encountered in the past gives some indication of the magnitude of the uncertainty. Clearly, the anticipated uncertainty increases as the historic range of concentrations increases, and decreases as historic range of concentrations decreases. Still, there is utility in applying past history to gain an increased understanding of what may occur in the future.

4.6.3 SRS Post-Cleaning Tank Residue

Average concentrations of key radionuclides in SRS Post-Cleaning Tank Residue are given in Table 4-7. Note that these characterizations were performed between the years of 2010 and 2014. As shown in the table, the radionuclide concentrations vary over ten orders of magnitude, from a low concentration of ~2E-06 µCi/g U-238 in Tank 16 INT to a high concentration of ~2E+4 µCi/g Sr-90 in Tank 6. The variations were significantly smaller for the individual radionuclides, where the spans of concentrations ranged from 10x for Pu-238 to 46000x for Sm-151. Clearly, the concentration spans for the post-cleaning residue were significantly higher than those for the sludge batches and those for salt feed. This difference was attributed to two primary factors: 1) the waste at the bottom of a tank is often not representative of the bulk waste – because the earliest receipts are typically not representative of the later receipts – and because unusual waste solids, including particularly high density and/or large diameter solids, tend to migrate to the floor during bulk removal (this includes ion exchanger waste, for example); and 2) the distribution of constituents in the residue has been altered during chemical cleaning, and the method of cleaning and extent of cleaning has evolved from tank to tank. In addition, differences in the distribution of the dominant stable constituents (such as iron and aluminum) will affect the chemical cleaning efficacies.

Given the wide range of concentration variations, use of the historic waste residue data as a basis for predicting future waste residue compositions is limited. In almost every case, the uncertainties of the predictions would be multiple orders of magnitudes, which seem too high for most closure characterization applications. On the other hand, continued tracking of the characterization results is considered useful, to provide an up to date baseline understanding of the variations.

An illustration of the high relative concentration variations associated with post-cleaning residue is provided in Figure 4-3. As shown in the figure, the concentration variations for Sr-90, Cs-137, U-235, Np-237, and Pu-239 range from two to four orders of magnitude. Note that in the cases of Cs-137, U-235, and Pu-239, the very lowest concentrations were observed in the Tank 16H interior residue, and these concentrations were one to three orders of magnitude lower than the lowest concentrations observed in the other tank residues. As such, if the Tank 16H interior data were excluded, the concentration variations would be more manageable (particularly in the case of Cs-137) – however, on the whole, they would still be significantly greater than those of the sludge and salt cases of the past few years. Note that the low constituent concentrations observed for the Tank 16H interior residue are expected, given the extreme level of cleaning that was performed in this particular case.

Table 4-7. Average Concentrations of Key Radionuclides in Post-Cleaning Tank Residue

Radionuclide	Mean Radionuclide Concentration, µCi per gram of total solids						Span of Concentrations
	Tank 18	Tank 19	Tank 5	Tank 6	Tank 16 INT	Tank 16 ANN	
Ni-59	9.45E-03	<3.29E-02	5.52E+00	4.41E+00	<2.97E-01	<1.21E-02	580x
Ni-63	6.42E-01	1.14E-02	2.97E+02	3.18E+02	<2.27E+00	<2.80E-01	28000x
Sr-90	4.75E+01	7.01E-01	1.21E+04	1.60E+04	2.98E+03	1.57E+03	23000x
Zr-93	2.27E-03	1.35E-03	2.98E+00	9.90E-01	<1.89E-02	<1.52E-01	2200x
Tc-99	3.62E-02	3.67E-02	9.99E-03	1.07E-01	8.27E-02	2.50E-01	25x
I-129	1.18E-05	1.11E-05	3.48E-04	1.59E-04	1.76E-04	8.73E-04	79x
Cs-135	1.12E-03	2.64E-03	2.12E-03	2.75E-03	<1.20E-04	2.50E-03	~23x
Cs-137	2.99E+02	5.77E+02	4.15E+02	5.43E+02	7.13E-01	6.89E+02	970x
Sm-151	1.54E+00	1.70E-02	7.79E+02	2.05E+02	NM	NM	46000x
Th-232	NM	NM	NM	NM	NM	NM	N/A
U-233	8.22E-04	3.88E-04	<1.07E-03	3.28E-03	1.51E-04	<2.21E-03	22x
U-234	7.96E-03	4.82E-04	4.45E-03	9.26E-03	1.67E-04	1.57E-03	55x
U-235	3.10E-04	1.69E-05	1.97E-04	4.31E-04	6.67E-07	2.42E-05	650x
U-238	7.87E-03	5.52E-04	4.56E-03	1.40E-02	2.38E-06	9.95E-05	5900x
Np-237	4.79E-03	1.45E-04	2.31E-02	1.98E-02	<6.83E-04	1.97E-03	160x
Pu-238	3.75E+00	3.58E-01	2.59E+00	3.31E+00	1.17E+00	3.44E+00	10x
Pu-239	8.99E+00	3.81E-01	8.22E+00	2.91E+00	4.38E-02	4.46E-01	210x
Am-241	4.50E+00	2.33E-01	7.00E+01	6.85E+01	3.24E-01	7.72E-01	300x
Am-243	2.77E-02	5.90E-04	5.35E-01	1.47E+00	1.08E-02	1.19E-02	2500x
Cm-244	2.31E+00	3.61E-02	3.03E+00	5.55E+01	<5.80E-02	<3.90E-01	1500x

NM = not measured

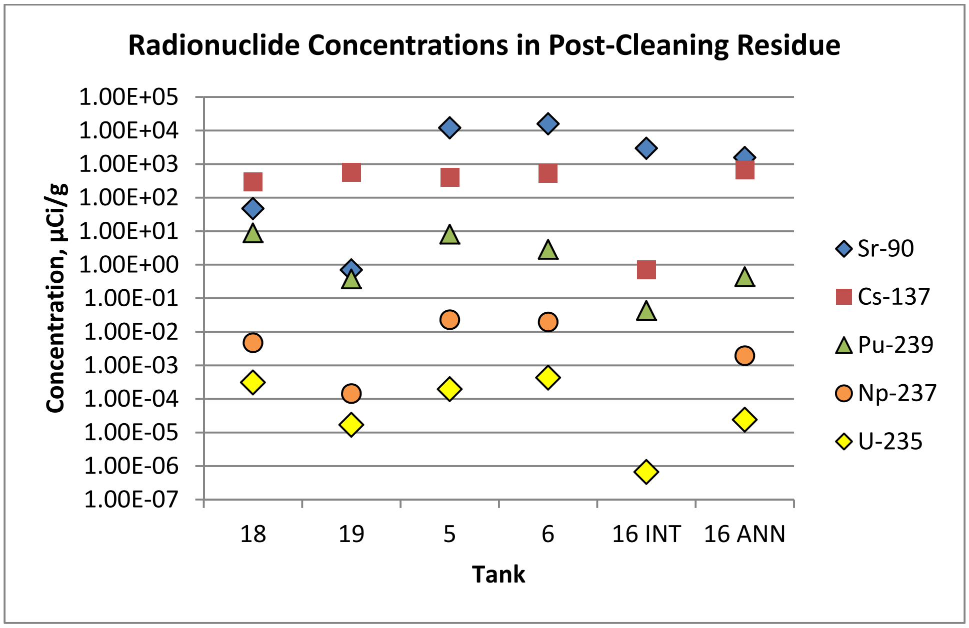


Figure 4-3. Concentrations of Select Radionuclides in SRS Post-Cleaning Tank Residue

4.7 Conclusions

In general, the uncertainties of sampling and analysis data are significantly lower than those associated with the alternative characterization approaches, due to the relatively high variability of waste compositions, mixing of multiple waste types, and the difficulties of tracking waste compositions as a function of location and time. However, judicious use of alternative characterization approaches may be adequate for many applications, particularly those where sufficient data consistency can be demonstrated and/or where somewhat higher characterization uncertainties are deemed acceptable. Because of the high costs of sampling and analysis, there is the clearly the potential to make characterization more cost-effective if some portion of the data is provided by an alternative means (by a non-sampling and analysis approach).

In many cases, use of the alternative characterization approaches are capable of providing constituent concentration estimates that are the appropriate order of magnitude, with deviations limited to the 2x-3x range. This includes estimates based on the waste receipt histories, process knowledge, use of scaling factors, and the historic data. Interestingly, the efficacies of the alternative characterization approaches appear to be functions of both the waste matrix (sludge, salt, or residue) and the particular constituent being addressed. On the whole, the alternative characterization approaches are more suited to sludge and salt, as opposed to post-cleaning residue, and to ubiquitous constituents that are present in every waste stream.

When high quality sampling and analysis data is available, the alternative sources of characterization data should still be considered, to assist in understanding the sampling and analysis data and to provide a level of confirmation that the sample analysis data is consistent with expectations.

Examples of cases where the alternative characterization approaches showed high potential for being effective included:

- Use of receipt records for understanding
  --the spatial distributions of plutonium isotopes in Tank 12 sludge and Tank 18 residue
  --the concentration ranges of key radioisotopes and metals in Tank 18 residue
- Use of process knowledge for estimating the concentrations of radioactive and stable constituents in sludge batches
- Use of scaling factors for estimating the concentrations of select radionuclides in sludge batches and salt solutions
- Use of historic data trends for
  --estimating the concentrations of key radionuclides in sludge batches and in salt feed
  --projecting the concentrations of key radionuclides in future salt feed solutions

Consideration of the importance of characterization accurateness should feed the potential for using alternate characterization approaches. In cases where constituents have little or no impacts on the disposition decisions and risks, use of alternative characterization approaches may be the best choice.

4.8 References

[1] Bannochie, C. J. and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 8 (Macrobatch 10)*," SRNL-STI-2014-00179, June 2014.

[2] Crawford, C. L. and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 7B (Macrobatch 9)*," SRNL-STI-2012-00294, Rev. 2, May 2014.

[3] Reboul, S. H, D. P. DiPrete, D. R. Click, and C. J. Bannochie, "*Reportable Radionuclides in DWPF Sludge Batch 7a (Macrobatch 8)*," SRNL-STI-2011, 00720, December 2011.

[4] Bannochie, C. J. and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 6 (Macrobatch 7)*," SRNL-STI-2011-00189, Revision 0, May 2011.

[5] Bannochie, C. J., N. E. Bibler, and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 5 (Macrobatch 6)*," SRNL-STI-2009-00821, Revision 0, February 2010.

[6] Bannochie, C. J., N. E. Bibler, and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 4 (Macrobatch 5)*," WSRC-STI-2008-00142, Revision 0, May 2008.

[7] Bannochie, C. J. and N. E. Bibler, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 3 (Macrobatch 4)*," WSRC-TR-2005-00157, Revision 0, May 2005.

[8] Bibler, N. E., D. P. DiPrete, and J. R. Harbour, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 2 (Macro Batch 3)*," WSRC-TR-2002-00255, September 2002.

[9] Fellinger, T. L., N. E. Bibler, and J. R. Harbour, "*Characterization and Waste Acceptance Radionuclides to be Reported for DWPF Macro Batch 2 (ESP 215 – ESP 221)*," WSRC-RP-99-00436, Revision 1, March 2004.

[10] Office of Environmental Management. *Waste Acceptance Product Specifications for Vitrified High-Level Waste Forms*, DOE/EM-0093, Revision 2, December 1996.

[11] Bannochie, C. J., "*Results for the Second Quarter 2014 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2014-00308, September 2014.

[12] Bannochie, C. J., "*Results for the First Quarter 2014 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2014-00203, May 2014.

[13] Bannochie, C. J., "*Results for the Fourth Quarter 2013 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2014-00074, April 2014.

[14] Bannochie, C. J., "*Results for the Third Quarter 2013 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2013-00651, November 2013.

[15] Bannochie, C. J., "*Results for the Second Quarter 2013 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2013-00390, July 2013.

[16] Bannochie, C. J., "*Results for the First Quarter 2013 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2013-00271, May 2013.

[17] Bannochie, C. J., "*Results for the Fourth Quarter 2012 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2012-00786, February 2013.

[18] Bannochie, C. J., "*Results for the Third Quarter 2012 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminants*," SRNL-STI-2012-00621, October 2012.

[19] Bannochie, C. J., "*Results for the First Quarter 2012 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2012-00286, Rev. 1, July 2012.

[20] Bannochie, C. J., "*Results for the Fourth Quarter 2011 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2012-00038, January 2012.

[21] Reigel, M. M., "*Results for the Third Quarter 2011 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2011-00574, October 2011.

[22] Eibling, R. E., "*Results for the Second Quarter 2011 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2011-00443, August 2011.

[23] Reigel, M. M., "*Results for the First Quarter 2011 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2011-00303, May 2011.

[24] Reigel, M. M., "*Results for the Fourth Quarter 2010 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2010-00713, February 2011.

[25] Reigel, M. M. and N. E. Bibler, "*Results for the Third Quarter 2010 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2010-00598, November 2010.

[26] Reigel, M. M. and N. E. Bibler, "*Results for the Second Quarter 2010 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2010-00437, July 2010.

[27] Reigel, M. M. and N. E. Bibler, "*Results for the First Quarter 2010 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2010-00210, April 2010.

[28] Reigel, M. M. and N. E. Bibler, "*Results for the Fourth Quarter 2009 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2009-00828, December 2009.

[29] Reigel, M. M., C. C. DiPrete, and N. E. Bibler, "*Results for the Third Quarter 2009 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2009-00585, November 2009.

[30] Reigel, M. M., C. C. DiPrete, and N. E. Bibler, "*Results for the First Quarter 2009 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNL-STI-2009-00452, October 2009.

[31] Bibler, N. E. and C. C. DiPrete, "*Results for the Fourth Quarter 2008 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNS-TR-2009-00144, April 2009.

[32] DiPrete, C. C. and N. E. Bibler, "*Results for the Third Quarter 2008 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," SRNS-TR-2008-00328, December 2008.

[33] Bibler, N. E. and C. C. DiPrete, "*Results for the First Quarter 2008 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," WSRC-TR-2008-00184, June 2008.

[34] Zeigler, K. E. and N. E. Bibler, "*Results for the Third Quarter 2007 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," WSRC-TR-2008-00080, Rev. 1, July 2008.

[35] Oji, L. N., D. P. DiPrete, C. J. Coleman, M. S. Hay, and E. P. Shine, "*Tank 16H Residual Sample Analysis Report*," SRNL-STI-2014-00321, Rev. 1, October 2014.

[36] Oji, L. N., D. P. DiPrete, C. J. Coleman, M. S. Hay, and E. P. Shine, "*Analysis of the Tank 6F Final Characterization Samples – 2012*," SRNL-STI-2012-00365, Rev. 2, January 2013.

[37] Oji, L. N., D. P. DiPrete, C. J. Coleman, and M. S. Hay, "*Analysis of the Tank 5F Final Characterization Samples – 2011*," SRNL-STI-2012-00034, Rev. 2, September 2012.

[38] Oji, L. N., D. P. DiPrete, and C. J. Coleman, "*Characterization of Additional Tank 19F Floor Samples*," SRNL-STI-2010-00439, August 2010.

[39] Oji, L. N., D. P. DiPrete, and C. J. Coleman, "*Characterization of Additional Tank 18F Samples*," SRNL-STI-2010-00386, September 2010.

[40] d'Entremont, P. D. and J. R. Hester, "*Characterization of Tank 20 Residual Waste*," WSRC-TR-96-0267, March 1997.

[41] Hay, M. S., "*Characterization of Tank 17F Samples in Support of Tank Closure*," WSRC-RP-97-066, January 1997.

[42] Reboul, S. H., L. N. Oji, D. P. DiPrete, C. J. Coleman, and E. P. Shine, "*Task Technical and Quality Assurance Plan for Tank 12H Residual Material Sample Analyses*," SRNL-RP-2014-00718, Rev. 1, October 2014.

[43] Cavin, W. S., "*Separations Waste Sludge Spreadsheet*," WSRC-TR-93-135, Rev. 1, July 1993.

[44] Cavin, W. S. and A. T. Crumm, "*Waste Transfers and Miscellaneous Additions*," WSRC-TR-93-425, Rev. 1, January 1994.

[45] Cavin, W. S. and A. T. Crumm, "*Fresh Waste Receipts*," WSRC-TR-93-426, Rev. 1, January 1994.

[46] Chandler, M. C., "*Estimated Sludge Inventory for Individual Tanks*," WSRC-TR-94-0191, Rev. 1, August 1994.

[47] Georgeton, G. K. and J. R. Hester, "*Characterization of Radionuclides in HLW Sludge Based on Isotopic Distribution in Irradiated Assemblies*," WSRC-TR-94-0562, Rev. 1, January 27, 1995.

[48] Reboul, S. H. and P. J. Hill, "*Characteristics and Understanding of SRS Tank Farm Waste*," CBU-PIT-2005-00090, presented to the National Academy of Science Tank Waste Committee, April 14, 2005.

[49] Reboul, S. H., "*Chemical Differences Between Sludge Solids at the F and H Area Tank Farms*," SRNL-STI-2012-00479, August 2012.

[50] Reboul, S. H., M. S. Hay, K. E. Zeigler, and M. E. Stone, "*Tank 12 Sludge Characterization and Aluminum Dissolution Demonstration*," SRNL-STI-2009-00180, March 2009.

[51] Martino, C. J. and W. D. King, "*Testing of Enhanced Chemical Cleaning of SRS Actual Waste Tank 5F and Tank 12H Sludges*," SRNL-STI-2011-00360, August 2011.

[52] Martino, C. J., S. H. Reboul, B. J. Wiersma, and C. J. Coleman, "*Tank 12H Acidic Cleaning Sample Analysis and Material Balance*," SRNL-STI-2013-00459, November 2013.

[53] Sludge 1.5 Database, "Tank 12 Spreadsheet, Version 3.1" Savannah River Remediation, Aiken, SC, January 2015.

[54] Hay, M. S., D. R. Click, D. P. DiPrete, and C. C. DiPrete, "*Characterization of Tank 18F Wall and Scale Samples*," SRNL-STI-2009-00802, March 2010.

[55] Dean, W. B., "*Tanks 18 and 19 Final Characterization Data Quality Assessment*," SRR-CWDA-2011-00149, Rev. 1, February 2012.

[56] Monier, J. A. and T. C. Evans, "*Thoria Powder Specifications – EM Specification 162*," E. I. du Pont de Nemours & Company, Savannah River Site, Aiken, SC, May 10, 1968.

[57] Fowler, J. R. and C. J. Coleman, "*Carbon-14 in Sludge*," DPST-83-2001, December 28, 1983.

[58] Fowler, J. R., B. A. Hamm, and J. R. Cook, "*Technetium-99 in SRP High-Level Waste and Saltstone*," DPST-84-334, February 24, 1984.

[59] Fowler, J. R. and J. R. Cook, "*I-129 in SRP High-Level Waste and Saltstone*," DPST-84-343, February 29, 1984.

[60] Bess, C. E., "*Waste Characterization of Activation Product Radionuclides in High Level Waste Supernate*," WSRC-TR-94-0291, June 1994.

[61] Reboul, S. H., "*Conclusions and Path Forward for Sn-126 in Low Curie Salt*," HLW-PRE-2002-0030, August 19, 2002.

[62] Reboul, S. H. and E. T. Ketusky, "*Compilation and Comparison of Radionuclide Concentrations Reported for SRS HLW Supernates*," CBU-SPT-2003-00020, February 18, 2003.

[63] Ledbetter, L. A., "*Waste Characterization of Ni-63 in High Level Waste Sludge*," CBU-PIT-2005-00017, January 2005.

[64] Tran, H. Q., "*Waste Characterization of Nb-94 in High Level Waste Sludge*," CBU-PIT-2005-00044, February 2005.

[65] Tran, H. Q., "*Supernatant Phase Iodine-129 Inventory*," CBU-PIT-2005-00050, February 2005.

[66] Tran, H. Q., "*Total Iodine-129 Curie Inventory Report*," CBU-PIT-2005-00033, February 2005.

[67] Hutchens, G. J., "*Estimate of Pu-244 Abundance in SRS High Level Waste Sludge*," CBU-PIT-2005-00039, March 2005.

[68] Hutchens, G. J., "*Estimate of Actinide Concentration by Radioactive Decay*," CBU-PIT-2005-00040, March 2005.

[69] Hutchens, G. J., "*Estimate of Al-26 Abundance in SRS High Level Waste*," CBU-PIT-2005-00041, March 2005.

[70] Reboul, S. H. and P. D. d'Entremont, "*Improving WCS Supernatant Actinide Projections*," CBU-PIT-2005-00076, March 2005.

[71] Hutchens, G. J. and H. Q. Tran, "*Estimate of Na-22 Abundance in SRS High Level Waste*," CBU-PIT-2005-00097, April 2005.

[72] Hill, P. J., "*Soluble Phase Selenium-79, Technetium-99, and Tin-126 Inventories*," CBU-PIT-2005-00127, May 2005.

[73] Tran, H. Q., "*Tank Radionuclide Inventories*," CBU-PIT-2005-00138, Rev. 1, October 2005.

[74] Reboul, S. H., "Nickel-59, Cerium-144/Praseodymium-144, and Radium-226 in Salt Feed," SRNL-L3100-2009-00189, September 8, 2009.

[75] Hester, J. R., "*High Level Waste Characterization System (WCS)*," WSRC-TR-96-0264, December 1996.

[76] Hester, J. R., "*WCS Supernate Radionuclide Algorithms*," X-ESR-G-00004, September 16, 2004.

[77] Tran, H. Q., "*Compilation of Additional Radionuclide Data for SRS HLW Sludge to be Included in Waste Characterization System (WCS II)*," CBU-PIT-2005-00034, March 2005.

[78] Tran, H. Q., "*Compilation of Additional Chemical Data for SRS HLW to be Included in Waste Characterization System (WCS II)*," CBU-PIT-2005-00182, July 2005.

[79] Tran, H. Q., "*HLW Inventories of Additional Radionuclides Identified in the 2005 Saltstone Special Analysis*," CBU-PIT-2005-00178, August 2005.

[80] WCS 1.5 Database, Version 3.1, Savannah River Remediation, Aiken, SC, January 2015.

[81] Hill, P. J., "*Comparison of Waste Characterization System (WCS) Estimations to Waste Tank Sample Results for Tank Farm Sludge*," WSRC-TR-2002-00312, July 2002.

[82] Hamm, B. A. and H. H. Elder, "*Savannah River Site Sludge Characterization Model Using Dial-Up Factors*," CBU-PIT-2006-00058, March 2006.

[83] Chandler, J. R., "*Ni-59, Ni-63, and Nb-94 in DWPF Saltcrete*," DPST-82-236, E. I. du Pont de Nemours & Company, January 1982.

[84] Reboul, S. H., "*Potential Application of Radionuclide Scaling Factors to High Level Waste Characterization*," SRNL-STI-2013-00492, September 2013.

5.0 Honing Characterization Needs as a Function of Waste Type

5.1 Approach

This section addresses differences between sludge, salt, and post-cleaning residue with the goal of honing characterization needs as a function of waste type. To accomplish this, documentation of sludge, salt, and post tank cleaning residue characterization was reviewed. For sludge, documentation included the Defense Waste Processing Facility (DWPF) Waste Acceptance Product Specification (WAPS) documents.[1-9] At the start of each DWPF sludge batch, extensive radionuclide characterization of the DWPF feed (Tank 40H) is completed. For salt waste, a spreadsheet showing all radionuclide characterization results for Tank 50, the Savannah River Site (SRS) Saltstone Disposal Facility (SDF) feed tank, was reviewed. For post tank cleaning residue, reports documenting analysis of material remaining in a tank after cleaning were reviewed,[10-14] Finally, several performance assessments (PAs) were reviewed to evaluate radionuclides that contributed to short and long term environmental dose.[15-17]

5.2 Sludge Characterization

SRS Tank Farm sludge is prepared for vitrification at the DWPF facility in a batch process. Sludge from a tank or various tanks is combined in Tank 51H where it is washed (a large portion of sodium is removed and the concentrations of major anions may be adjusted). The washed Tank 51H material is then transferred to the DWPF feed tank – Tank 40H. At this point a new sludge batch (SB) is “declared” and a sample from Tank 40H is extensively characterized. The radionuclide characterization is governed by the WAPS.[18] In summary, the WAPS requires that the inventory of radionuclides (in curies) that have half-lives longer than 10 years and that are, or will be, present in concentrations greater than 0.05 percent of the total inventory between the years 2015 and 3115 be reported. The WAPS also requires quantification of U and Pu isotopes. As part of the strategy to comply with WAPS requirements, the DWPF reports all radionuclides (with half-lives greater than 10 years) that have concentrations greater than 0.01 percent of the total curie inventory from time of production through the 1100 year period from 2015 through 3115.[19] Note that the criterion of using greater than 0.01 curie percent to identify “reportable” radionuclides was an SRS decision made for the purpose of conservatism, as the actual “reportability” criterion stated in the WAPS is greater than 0.05 curie percent.

To determine curie content over the required time period, an extensive list of radionuclides in a Tank 40H sample is quantified. The radionuclide content is then input into a radioactive decay calculator to calculate activity over the prescribed time period in 100 year increments. Each WAPS report[1-9] describes the radionuclides quantified, reasons for not quantifying some radionuclides, and details on quantification methodology (measurement or calculation). The quantified radionuclides from Sludge Batch 1B to Sludge Batch 8 are given in Table 5-1. As can be seen, the number of quantified radionuclides has varied over time. Some of this change is due to improvements in analysis techniques, a better knowledge of the radionuclides in the SRS sludge, and better understanding of the DWPF process. For example, C-14 is no longer quantified since carbon is not present in the final DWPF vitrified waste form.

As stated above, Table 5-1 shows the radionuclide concentrations in the past nine sludge batches (SB1b to SB8). Blank cells indicate that the radionuclide was not quantified in the Tank 40H sample. Gray shading indicates that the radionuclide was reportable – which means that the radionuclide contributed greater than 0.01% of the total curie content at some point in the 1100 year evaluation time period.

An additional table (Table 5-2) shows the minimum and maximum concentrations of radionuclides over the past 9 sludge batches. Only radionuclides quantified/detected in five or more sludge batches are included in the table. Also included is the ratio of the maximum and minimum concentrations. This ratio gives a crude indication of variability over the sludge batches.

In a review of the tables and the general WAPS reportable radionuclide process, several observations can be made:

- Although the number of quantified radionuclides in each batch has varied significantly (from 32 to 66), the number of reportable radionuclides has been relatively consistent (from 26 to 31). Some of these reportable radionuclides are based on inadequate minimum detection limits, particularly the high mass number radionuclides. A more detailed evaluation of the consistently reportable radionuclides could reduce the number of radionuclides requiring quantification.

- With only a few exceptions, the variation in individual radionuclide concentrations in Tank 40H is less than 100X (see Table 5-2). Using historical data or calculations could decrease the need for extensive characterization.

- Only considering radionuclides that do indeed meet the WAPS requirement (0.05% instead of the DWPF practice of 0.01%) may reduce the need for quantification of some radionuclides.

In evaluating the WAPS requirements/process alongside the Tank Farm and Saltstone Disposal Facility Performance Assessments (PAs)[15-17], the following observations are made.

- The practice of calculating curie concentrations for reportability is questionable. The WAPS decay timeframe of 1100 years is short compared to the PAs, the long-lived nature of the sludge (and ultimately vitrified) radionuclides, and the geologic storage timeframe of the vitrified sludge. Decay calculations could be completed at any time; calculations at the time of characterization are not necessary.

- Reportability based on curie content does not directly account for risk or dose. That is, not all curies are equal with respect to potential environmental and biological effects.

- The requirement to report radionuclides based on a fraction of total curies may prevent "high risk" radionuclides from being reported. Lower risk radionuclides in essence "dilute" higher risk radionuclides. Ra-226 is an example. This isotope is a major contributor to dose (based on review of the PAs[15-17]), yet it is not DWPF reportable in any sludge batches.

Table 5-1. Tank 40H Radionuclide Concentrations With WAPS-Reportable Radionuclides Highlighted in Gray From Sludge Batch 1B to Sludge Batch 8 (blanks indicate radionuclide concentration was not quantified)

	SB1b	SB2	SB3	SB4	SB5	SB6	SB7a	SB7b	SB8
Radionuclide	µCi/g of total solids								
C-14	4.41E-03	8.13E-03							
Cl-36					<9.75E-01	<2.32E-03	<2.5E-04	<4.07E-03	<1.32E-04
Co-60	7.27E-01	5.47E+00	1.62E+00	8.56E-01	2.40E+00	2.15E+00	2.70E+00	2.76E+00	1.25E+00
Ni-59	4.78E-02	3.13E-01	6.63E-01	5.67E-01	1.30E+00	1.05E+00	1.50E+00	2.57E+00	7.53E-01
Ni-63	8.42E+00	2.05E+01	5.41E+01	6.58E+01	1.11E+02	1.08E+02	2.50E+01	1.60E+02	9.34E+01
Se-79	4.16E-02	6.23E-02	<1.97E-02	1.61E-02	<2.78E-03	9.10E-03	7.60E-03	1.54E-02	1.04E-02
Sr-90	4.24E+03	4.52E+03	4.85E+03	7.23E+03	2.25E+04	1.87E+04	1.40E+04	1.27E+04	9.85E+03
Y-90			4.85E+03	7.23E+03	2.25E+04	1.87E+04	1.40E+04	1.27E+04	9.85E+03
Zr-93	8.22E-02	1.36E-01	4.43E-02	2.64E-01	5.23E-01	4.59E-01	5.30E-01	4.63E-01	7.02E-01
Nb-93m				2.09E-01	3.93E-01	3.79E-01	4.60E-01	4.21E-01	6.15E-01
Tc-99	1.88E-01	1.26E-01	1.96E-01	1.41E-01	1.53E-01	<1.09E-01	1.00E-01	9.67E-02	3.43E-01
Ru-106							<6.4E-01		
Rh-106							<6.4E-01		
Pd-107	5.30E-04	4.16E-04	6.89E-04	5.98E-04	1.29E-03	1.38E-03	1.20E-03	1.22E-03	1.52E-03
Cd-113m				1.29E+00	2.91E+00	3.35E+00	3.10E+00	3.33E+00	2.35E+00
Sn-121m		3.64E+00	<2.34E+00	<3.18E-02	<3.05E-01	<4.73E+00	<5.7E-02	4.95E-01	3.70E-01
Sn-126	1.42E-02	4.49E-02	<1.42E-02	<3.24E-01	<3.60E-01	<1.48E-01	<6.4E-01	<7.03E-01	<1.28E-01
Sb-125			<8.29E-02	<1.86E-01	<9.90E-02	1.20E-01	<2.4E-01	<3.64E-01	<2.82E-01
Te-125m			<8.29E-02	<1.86E-01	<9.90E-02	1.20E-01		<3.64E-01	<1.28E-01
I-129	1.09E-03	7.57E-06	<4.35E-04	5.20E-04	<3.77E-04	2.56E-03	1.70E-03	1.06E-03	2.04E-03
Cs-134							<7.9E-01		
Cs-135	7.54E-04	1.08E-03	1.34E-03	1.18E-03	1.61E-03	1.85E-03	2.80E-03	3.32E-03	4.55E-03
Cs-137	1.59E+02	2.75E+02	3.15E+02	2.49E+02	3.23E+02	3.87E+02	5.90E+02	6.44E+02	8.52E+02
Ba-133				<9.14E-02	<8.42E-02	<2.64E-02	<2.0E-01	<1.87E-01	<6.67E-02
Ba-137m			3.03E+02	2.39E+02	3.10E+02	3.71E+02	5.60E+02	6.18E+02	8.06E+02
Ce-144							<1.3E+00		
Pr-144							<1.3E+00		
Pm-147		1.30E+02	1.20E+03	<1.44E+01	<1.60E+02	<1.48E+02	<1.6E+02	<1.73E+02	<1.25E+02
Sm-151	6.32E+01	1.77E+02	1.96E+02	1.13E+02	1.82E+02	2.62E+02	2.30E+02	2.47E+02	1.63E+02
Eu-152									2.87E-01
Eu-154	1.05E+01	8.01E+00	7.73E+00	1.15E+01	2.33E+01	2.23E+01	1.70E+01	1.39E+01	1.11E+01
Eu-155			2.12E+00	1.22E+00	2.45E+00	2.80E+00	2.60E+00	3.34E+00	1.59E+00
Pb-210							3.80E-06	2.69E-06	1.56E-06

Table 5-1. Tank 40H Radionuclide Concentrations With WAPS-Reportable Radionuclides Highlighted in Gray From Sludge Batch 1B to Sludge Batch 8 (blanks indicate radionuclide concentration was not quantified)

	SB1b	SB2	SB3	SB4	SB5	SB6	SB7a	SB7b	SB8
Radionuclide	µCi/g of total solids								
Ra-226							7.70E-06	5.30E-06	3.42E-06
Ra-228							1.60E-03		
Ac-227							3.40E-07	3.55E-07	9.02E-04
Th-228							1.60E-03		
Th-229							6.30E-04	2.11E-04	2.35E-04
Th-230							3.80E-04	2.41E-04	7.30E-05
Th-232	3.41E-04	3.76E-05	3.01E-05	5.07E-05	7.28E-05	2.44E-03	1.60E-03	1.19E-03	8.95E-04
Pa-231							6.80E-07	6.86E-07	<4.34E-04
U-233	4.35E-02	1.07E-02	1.56E-02	<1.91E-02	2.22E-02	8.98E-02	1.30E-01	4.22E-02	6.00E-02
U-234	2.89E-03	3.57E-02	3.03E-02	3.40E-02	6.15E-02	8.37E-02	4.80E-02	4.06E-02	4.39E-02
U-235	2.81E-04	6.57E-04	7.29E-04	6.41E-04	7.00E-04	6.01E-04	6.40E-04	6.12E-04	5.41E-04
U-235m							1.30E+01		
U-236	7.38E-04	9.44E-04	8.46E-04	8.12E-04	1.34E-03	1.33E-03	1.10E-03	9.03E-04	1.01E-03
U-238	6.46E-03	2.53E-02	2.26E-02	1.77E-02	1.67E-02	1.22E-02	1.50E-02	1.60E-02	1.25E-02
Np-237	1.29E-02	1.33E-02	3.30E-02	2.60E-02	6.92E-02	3.61E-02	2.20E-02	2.46E-02	2.50E-02
Pu-238	9.95E+01	3.85E+01	2.59E+01	1.22E+02	2.76E+02	3.83E+02	1.80E+02	1.42E+02	1.99E+02
Pu-239	4.89E+00	7.68E+00	1.37E+01	1.03E+01	2.04E+01	1.82E+01	1.30E+01	1.08E+01	7.78E+00
Pu-240	1.78E+00	2.38E+00	4.80E+00	4.51E+00	6.59E+00	6.51E+00	4.10E+00	3.59E+00	2.82E+00
Pu-241	2.84E+01	2.80E+01	5.42E+01	1.08E+02	9.74E+01	<8.33E+01	5.60E+01	5.09E+01	4.32E+01
Pu-242	3.73E-03	4.59E-03	5.46E-03	5.83E-03	6.82E-03	<1.53E-02	<2.3E-02	<8.98E-03	<7.31E-03
Pu-244							<1.2E-10		
Am-241	6.65E+00	3.27E+01	1.85E+01	1.76E+01	3.25E+01	3.29E+01	3.70E+01	3.56E+01	2.74E+01
Am-242m		1.10E-01	<8.93E-02	7.00E-02	<4.54E-01	2.26E-01	3.40E-02	3.25E-02	1.42E-01
Am-243	5.65E-02	5.34E-01	1.55E+00	1.64E+00	9.64E-01	4.33E+00	5.70E-01	4.99E-01	1.88E+00
Cm-242			<7.46E-03	5.78E-02	<3.75E-01	1.87E-01	2.80E-02	2.68E-02	1.18E-01
Cm-243	1.41E-01	6.01E-01	<2.55E-01	<5.96E-01	<2.42E-01	<1.24E+00	<3.9E-01	<2.89E-01	<7.33E-02
Cm-244	4.43E+00	4.42E+01	7.00E+01	9.07E+01	4.61E+01	1.49E+02	1.90E+01	1.81E+01	6.81E+01
Cm-245		3.86E-03	1.09E-02	<3.49E-02	<4.29E-02	2.04E-02	2.80E-03	2.63E-03	1.08E-02
Cm-246	4.05E-02	2.48E-02	1.62E-02	1.90E-02	1.29E-02	6.46E-02	7.30E-03	6.60E-03	3.10E-02
Cm-247	2.30E-06	9.75E-07	<2.65E-02	<5.65E-03	<2.43E-07	<4.52E-07	<9.2E-08	<3.25E-08	<1.40E-07
Cm-248			<2.77E-02	<5.91E-03	3.77E-05	<7.07E-03	<3.1E-04	<1.23E-06	<8.43E-06
Bk-247				<1.36E-02	<2.71E-03	<5.05E-03	<1.0E-03	<3.62E-04	<1.55E-03
Cf-249		5.20E-03	<2.83E-02	<7.39E-03	<8.96E-03	<2.30E-02	<4.0E-03	<2.35E-03	<1.69E-02
Cf-250			<7.46E-03	5.78E-02	<1.36E-04	<8.40E-04	<9.6E-05	<3.59E-05	<6.52E-05
Cf-251		1.19E-02	<2.24E-02	<1.88E-02	<2.31E-02	<5.49E-02	<9.2E-03	<5.10E-03	<4.79E-02

Table 5-1. Tank 40H Radionuclide Concentrations With WAPS-Reportable Radionuclides Highlighted in Gray From Sludge Batch 1B to Sludge Batch 8 (blanks indicate radionuclide concentration was not quantified)

	SB1b	SB2	SB3	SB4	SB5	SB6	SB7a	SB7b	SB8
Radionuclide	µCi/g of total solids								
Cf-252							<2.8E-02	<2.09E-02	<8.30E-02
Number of Radionuclides Quantified	32	39	46	50	51	51	66	58	59
Number of Reportable Radionuclides	27	27	31	30	27	31	26	27	30

Table 5-2. Comparison of Minimum and Maximum Concentrations of Selected Radionuclides in DWPF Sludge Batches

Radionuclide	Minimum µCi/g	Maximum µCi/g	Maximum:Minimum Ratio
Co-60	7.27E-01	5.47E+00	8
Ni-59	4.78E-02	2.57E+00	54
Ni-63	8.42E+00	1.60E+02	19
Se-79	7.60E-03	6.23E-02	8
Sr-90	4.24E+03	2.25E+04	5
Zr-93	4.43E-02	7.02E-01	16
Nb-93m	2.09E-01	6.15E-01	3
Tc-99	9.67E-02	3.43E-01	4
Pd-107	4.16E-04	1.52E-03	4
Cd-113m	1.29E+00	3.35E+00	3
I-129	7.57E-06	2.56E-03	338
Cs-135	7.54E-04	4.55E-03	6
Cs-137	1.59E+02	8.52E+02	5
Sm-151	6.32E+01	2.62E+02	4
Eu-154	7.73E+00	2.33E+01	3
Eu-155	1.22E+00	3.34E+00	3
Th-232	3.01E-05	2.44E-03	81
U-233	1.07E-02	1.30E-01	12
U-234	2.89E-03	8.37E-02	29
U-235	2.81E-04	7.29E-04	3
U-236	7.38E-04	1.34E-03	2
U-238	6.46E-03	2.53E-02	4
Np-237	1.29E-02	6.92E-02	5
Pu-238	2.59E+01	3.83E+02	15
Pu-239	4.89E+00	2.04E+01	4
Pu-240	1.78E+00	6.59E+00	4
Pu-241	2.80E+01	1.08E+02	4
Pu-242	3.73E-03	6.82E-03	2
Am-241	6.65E+00	3.70E+01	6
Am-242m	3.25E-02	2.26E-01	7
Am-243	5.65E-02	4.33E+00	77
Cm-242	2.68E-02	1.87E-01	7
Cm-244	4.43E+00	1.49E+02	34
Cm-245	2.63E-03	2.04E-02	8
Cm-246	6.60E-03	6.46E-02	10

5.3 Tank Closure Samples

Following tank cleaning, residual material from waste tanks is sampled and extensively characterized prior to closure; over 60 radionuclides are quantified for several samples per tank.[10-14] To date, six tanks have been closed (Tanks 5F, 6F, 17F, 18F, 19F, and 20F) and an additional tank is in the process of being closed (Tank 16H). Five of these tanks have gone through the extensive radionuclide characterization needed to demonstrate removal of highly radioactive radionuclides (HRRs) to the maximum extent practical, as required by Section 3116 of the Ronald W. Reagan National Defense Authorization Act for Fiscal Year 2005. A cursory review of a select set of radionuclide characterization data from these samples is presented in Table 5-3 below. Sr-90 and Pu-239 were selected for comparison because they are dominant beta and alpha emitters, respectively, in the residual solid-phase waste. Tc-99 and I-129 were selected because they are long-lived mobile radionuclides that typically have potential impact on long term environmental risk.

As can be seen, there is significant variability between the radionuclide concentrations in these samples. For Sr-90, the concentrations ranged over four orders of magnitudes; and for Tc-99, I-129, and Pu-239, the concentrations ranged over two orders of magnitude. Thus, utilizing historical data may not be the best approach for reducing characterization requirements of closure radionuclides.

However, a focus on long term risk (see PAs[15,16]) could be used to guide the selection of radionuclides for quantification. For example, from the tank closure PAs, only 16 radionuclides were identified as being significant contributors to dose to the public and to an inadvertent intruder: C-14, Sr-90/Y-90, Nb-94, Tc-99, I-129, Sn-126, Cs-135, Cs-137/Ba-137m, Ra-226, Th-229, Pa-231, U-233, U-234, Np-237, Pu-238, and Am-241. Of these nuclides, Tc-99, Ra-226, Pa-231, and Np-237 were the primary risk drivers associated with the closed tanks. Thus, focusing on those nuclides driving the risk (and the applicable parent nuclides) could reduce the characterization needs.

Table 5-3. Tank Closure Sample Results

Tank		Sr-90	Tc-99	I-129	Pu-239
		µCi/g dried solids			
5F	Composite 1	1.21E+04	8.78E-03	1.57E-04	8.17E+00
	Composite 2	1.26E+04	1.09E-02	<1.60E-03	7.72E+00
	Composite 3	1.15E+04	1.03E-02	2.28E-04	8.78E+00
6F	Composite 1	1.57E+04	1.17E-01	1.33E-04	2.94E+00
	Composite 2	1.58E+04	1.02E-01	1.57E-04	2.75E+00
	Composite 3	1.65E+04	1.03E-01	1.87E-04	3.05E+00
16H (interior)	1-P	2.80E+03	1.07E-01	2.26E-04	4.14E-02
	2-P	<5.06E+00	9.88E-04	≤7.07E-06	<6.03E-02
	3-P	3.15E+03	1.40E-01	2.95E-04	4.61E-02
16H (annulus)	Composite 1	1.36E+03	2.23E-01	7.76E-04	4.39E-01
	Composite 2	2.04E+03	2.53E-01	7.64E-04	5.59E-01
	Composite 3	1.31E+03	2.73E-01	1.08E-03	3.39E-01
18F	North	5.01E+01	3.77E-02	1.22E-05	7.08E+00
	South	4.49E+01	3.47E-02	1.14E-05	1.09E+01
19F	North	6.32E-01	3.71E-02	1.11E-05	3.92E-01
	South	7.69E-01	3.63E-02	<1.04E-05	3.70E-01

5.4 Salt Waste Samples

Salt waste at SRS is dispositioned to the SDF. The SRS feed tank for the SDF is Tank 50H. This tank receives material from several places: ARP/MCU (where actinide, strontium, and cesium content of the salt is reduced), low level waste from H Canyon, and material from the SRS Effluent Treatment Facility. Tank 50H is sampled and characterized quarterly. Approximately fifty radionuclides are included as part of the characterization. Characterization results are compared to Waste Acceptance Criteria (WAC) limits and targets.[20] See Reference 21 for an example report showing analytical results and targets. Long-term environmental risk is projected in the Saltstone PA,[17] which identifies Tc-99, I-129, and Ra-226 as the primary dose drivers.

Tank 50H WAC analysis results from the third quarter of 2008 to the second quarter of 2014 have been compiled in a spreadsheet. These results were compared to the current (January 2014) WAC limits/targets. Results of this comparison are presented in Table 5-4 and Figure 5-1.

Table 5-4. Saltstone WAC Radionuclides Grouped by Percent of 2014 WAC Limits/Targets (Peak Concentrations from 3Q2008 to 2Q2014 Used for Grouping)

Not Detected	<0.01% of WAC	0.01%-0.1% of WAC	0.1%-1% of WAC	1%-10% of WAC	>10% of WAC
U-233 Ni-59 Zr-93 Nb-94 Ru-106/Rh-106 Cs-134 Ce-144 Ra-226 Ra-228 Ac-227 Th-230 Pa-231 Pu-242 Pu-244 Cm-245	Al-26 Th-232 Am-242m Cm-242	Pm-147 Eu-154 U-236 U-238 Am-243	H-3 C-14 Ni-63 U-235 Co-60 Te-125m Eu-155 Th-229 U-232 Np-237 Am-241	Sr-90/Y-90 I-129 Pu-241 K-40 Se-79 Sn-126 Sm-151 U-234 Pu-239 Pu-240 Cm-244	Tc-99 Cs-137/Ba-137m Sb-125 Cs-135 Pu-238

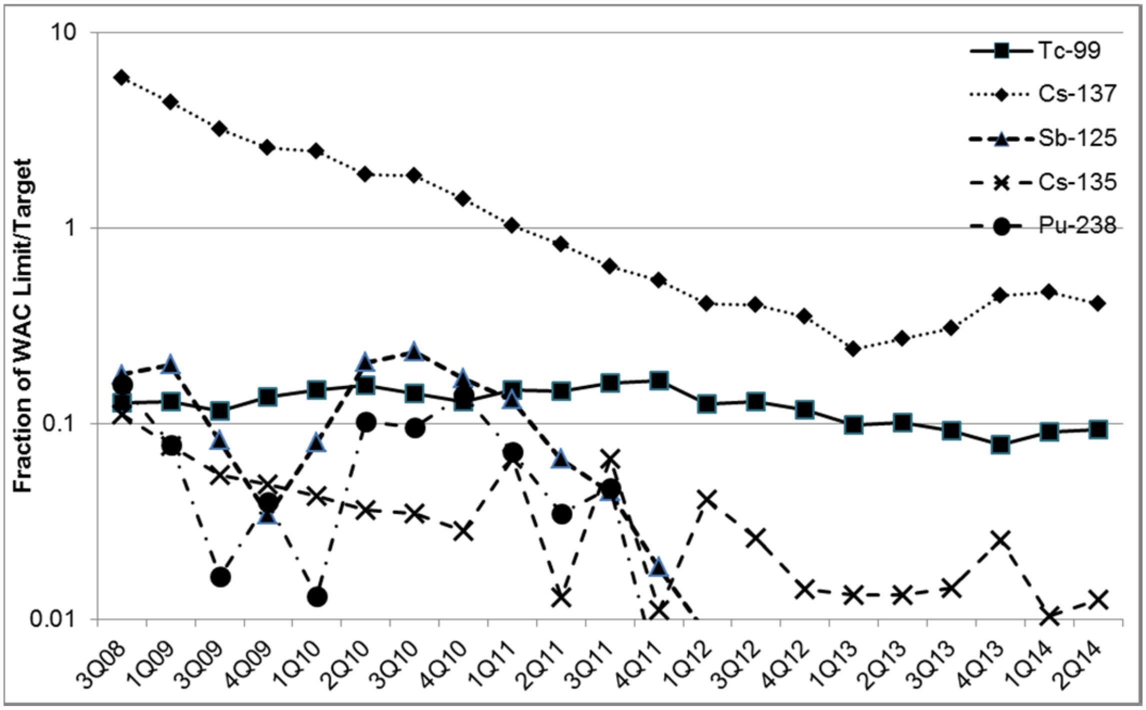


Figure 5-1. Radionuclides Exceeding 10% of the 2014 WAC Limits/Targets

The first column of Table 5-4 lists all the radionuclides that have not been detected in the quarterly samples. The remaining columns show radionuclides that have been detected at least once. The peak detected concentration was then used to group the radionuclides relative to the 2014 WAC targets/limits. For example, Eu-154 concentrations ranged from <0.01% to 0.02% of WAC limits/targets. Because the maximum was 0.02%, Eu-154 was placed in the third column. The radionuclides that approached limits/targets (i.e., >10%of WAC) are shown in Figure 5-1.

In reviewing the table and figure:

- The majority of the radionuclides are well below the WAC limits and targets (<1% of the limits and targets) and have been well below the limits/targets since the start of disposition. Fifteen radionuclides have not been detected, and four have peak concentrations less than 0.01% of limits. Consideration to not analyze some or all of these radionuclides and/or to reduce the frequency of these analyses should be pursued.

- To date, the most significant radionuclide in salt feed has been Cs-137/Ba-137m, with concentrations initially exceeding the WAC limit/target, but declining since the start of MCU processing, and maintaining a concentration less than the limit since the middle of 2011. Cs-137/Ba-137m concentrations over the past year have risen slightly, but are still significantly less than the limit (by a factor of about four). Because of the significance of Cs-137/Ba-137m and relative ease of measurement (by gamma spectroscopy), it is one of the few radionuclides that still warrants continuous frequent (quarterly) analysis.

- The other “most significant” radionuclides include Tc-99, Sb-125, Cs-135, and Pu-238, with peak concentrations exceeding 10% of the limits/targets at least once since the start of the monitoring program. Of these nuclides, Tc-99 is the one with the most dominant and consistent concentration over time, teetering at around 10% of the limit since the start of the program. In contrast, the concentrations of Sb-125, Cs-135, and Pu-238 have been less than 10% of the limits for the majority of the monitoring events. In fact, since 2012, most of their concentrations have been significantly lower than 10% of the limits (the majority less than 1% of the limits). As such, there seems to be a basis for reducing the frequency of characterizing these three nuclides, except in cases where process knowledge and/or salt qualification results indicate otherwise. Certainly, if MCU and ARP processing were stopped, the concentrations of Cs-135 and plutonium isotopes in the salt feed would be higher. If this occurred and the cesium and/or plutonium isotope concentrations were expected to challenge the limits, the move to lower the frequency of monitoring these isotopes could be lifted.

5.5 Conclusions

Possibilities for reducing the various characterization campaigns are a function of waste type, due to differences in the characterization objectives. Decisions on how to reduce characterization scope should be based on the following considerations:

- For sludge – historic concentration trends, MDL impacts, conservatism of curie fraction (0.01% vs 0.05%), and consistency of “reportable list” (consider defining a standard list)
- For tank closure – high potential concentration variability, but relatively low number of risk drivers
- For salt feed – most constituents are well below the WAC limits and are predictable based on historic trends, process knowledge, and qualification testing

Based on the PAs, the primary risk-driving constituents associated with closed tanks are Tc-99, Ra-226, Pa-231, and Np-237, while the primary risk-driving constituents associated with Saltstone are Tc-99, I-129, and Ra-226.

5.6 References

[1] C. J. Bannochie and N. E. Bibler, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 3 (Macrobatch 4)*," WSRC-TR-2005-00157, May 2005.

[2] C. J. Bannochie, N. E. Bibler, and D .P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 4 (Macrobatch 5)*," Savannah River National Laboratory, Aiken, SC, WSRC-STI-2008-00142, May 2008.

[3] C. J. Bannochie, N. E. Bibler, and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 5 (Macrobatch 6)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2009-00821, February 2010.

[4] C. J. Bannochie and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 6 (Macrobatch 7)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2011-00189, May 2011.

[5] C. J. Bannochie and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 8 (Macrobatch 10)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2014-00179, June 2014.

[6] N. E. Bibler, D. P. DiPrete, and J. R. Harbour, "*Determination of Reportable Batch 2 (Macro Batch 3)*," Savannah River Site, Aiken, SC, WSRC-TR-2002-00255, September 2002.

[7] C. L. Crawford and D. P. DiPrete, "*Determination of Reportable Radionuclides for DWPF Sludge Batch 7b (Macrobatch 9)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2012-00294, Rev. 2, May 2014.

[8] T. L. Fellinger, J. R. Harbour, and N. E. Bibler, "*Characterization of and Waste Acceptance Radionuclides to be Reported for Macro Batch 2 (ESP 215-ESP 221)*," Westinghouse Savannah River Company, Aiken, SC, WSRC-RP-99-00436, Rev. 1, March 2004.

[9] S. H. Reboul, D. P. DiPrete, D. R. Click, and C. J. Bannochie, "*Reportable Radionuclides in DWPF Sludge Batch 7a (Macrobatch 8)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2011-00720, December 2011.

[10] L. N. Oji, D. P. DiPrete, and C. J. Coleman, "*Characterization of Additional Tank 18F Samples*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2010-00386, September 2010.

[11] L. N. Oji, D. P. DiPrete, and C. J. Coleman, "*Characterization of Additional Tank 19F Floor Samples*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2010-00439, August 2010.

[12] L. N. Oji, D. P. DiPrete, C. J. Coleman, and M. S. Hay, "*Analysis of the Tank 5F Final Characterization Samples-2011 (U)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2012-00034, Rev. 2, September 2012.

[13] L. N. Oji, D. P. DiPrete, C. J. Coleman, M. S. Hay, and E. P. Shine, "*Tank 16H Residual Sample Analysis Report*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2014-00321, Rev. 1, October 2014.

[14] L. N. Oji, D. P. Diprete, M. S. Hay, and E. P. Shine, "*Analysis of the Tank 6F Final Characterization Samples-2012 (U)*," Savannah River National Laboratory, Aiken, SC, SRNL-STI-2012-00365, Rev. 2, January 2013.

[15] SRR, "*Performance Assessment for F-Tank Farm at the Savannah River Site*," Savannah River Remediation, Aiken, SC, SRS-REG-2007-00002, Rev. 1, 2010.

[16] SRR, "*Performance Assessment for the H-Area Tank Farm at the Savannah River Site*," Savannah River Remediation, Aiken, SC, SRR-CWDA-2010-00128, Rev. 1, 2012.

[17] SRR, "*Performance Assessment for the Saltstone Disposal Facility at the Savannah River Site*," Savannah River Remediation, Aiken, SC, SRR-CWDA-2009-00017, 2009.

[18] Office of Environmental Management, "*Waste Acceptance Product Specifications for Vitrified High-Level Waste Forms*," USDOE Document DOE/EM-0093, Rev. 2, 1996.

[19] SRR, "*DWPF Waste Form Compliance Plan*," Savannah River Site, Aiken, SC, WSRC-IM-91-116-0, Rev. 9, 2012.

[20] K. D. Dixon, "*Waste Acceptance Criteria for Aqueous Waste Sent to the Z-Area Saltstone Production Facility (U)*," Savannah River Remediation, Aiken, SC, X-SD-Z-00001, Rev. 13, January 2014.

[21] C. L. Crawford, "*Tables Containing Results for the Third Quarter 2014 Tank 50 WAC Slurry Sample: Chemical and Radionuclide Contaminant Results*," Savannah River National Laboratory, Aiken, SC, SRNL-L3100-2014-00221, September 2014.

6.0 Alternative Laboratory Methods

6.1 Approach

This section addresses potential alternative laboratory characterization methods holding promise for being less resource intensive, less time consuming, capable of improving data quality, and/or reducing personnel dose. Three primary categories of potential alternative characterization methods are addressed, those involving: 1) automation or semi-automation of radiochemical separation and waste removal processes; 2) development of improved radiochemical separation methods; and 3) improved measurement instrumentation and approaches. These three categories of alternatives were identified based upon known program needs/limitations as determined through decades of laboratory experience characterizing tank waste including sludge batches dispositioned at the Defense Waste Processing Facility, salt batches dispositioned at the Saltstone Disposition Facility, and post-cleaning waste residue quantified in support of tank closure.

Discussions of the most promising characterization alternatives and the rationales for their potential application are provided in this section, along with a summary of the estimated costs and time requirements associated with fabrication, testing, and implementation. This information serves as a basis for developing a path forward for integrating the improved laboratory methods into SRNL's existing tank waste characterization program.

6.2 Automation/Semi-Automation of Radiochemical Separation and Waste Removal Processes

Laboratory characterizations on highly radioactive tank waste sample matrices require numerous resource intensive radiochemical separations in both manpower and laboratory space. Due to the high resource loading, these separations are highly serial in nature, requiring one to be completed before the second one can start. In addition, tank closure samples are highly radioactive. As these separations are very "hands on" in nature, their execution on sample matrices of this type carries significant risk of personnel exposure and personnel contamination. Personnel exposure risks often result in radiochemical separation protocols being conducted in two stages. The first stage is conducted in remote handled analytical cells to reduce sample dose to a degree, and then a second separation is performed to generate an analytical quality purified fraction of the radioisotope in question. The majority of these analyses require radiochemical separations based on highly specific solid phase extractants. These separations pose risks to hands-on personnel due to associated dose and risk of contamination. They also present risks to schedules since they are extremely time and labor intensive. The development of a flexible system to automate the initial steps of the radiochemical separations is proposed and is expected to maximize productivity, optimize the use of strained labor resources, minimize personnel risks from dose and contamination, and facilitate accelerated schedules.

6.2.1 Current Limitations:

The majority of the radiochemical analyses performed on highly radioactive samples at SRS are completed using a vacuum box and series of columns containing solid phase extractants. The waste material that passes through the columns during the initial steps contains much of the dose associated with the sample. The columns are attached to various external ports on the vacuum box, as shown in Figure 6-1.

Figure 6-1. Vacuum Box Arrangement Typically Employed for Solid Phase Extractions

The solid phase extractants are conditioned with a particular chemical agent. Treated radioactive sample aliquots are added to the solid phase extractant columns. Numerous hands on operations are then conducted involving various reagent additions, waste removal, and finally purified product removal. For highly radioactive samples, often the analyte of interest is quite low in concentration compared to the interfering analytes. As a result, the separations often require significant rinse volumes. Quite often the separations require significant volumes of different chemical rinses to remove various elements which have a radioactive isotope that interferes with the analyte of interest. These large volumes of rinse solutions require multiple entries into the vacuum boxes for waste removal. Each entry is time consuming, increases personnel exposure, and increases risk of personal contamination.

An automated system would help increase efficiency and reduce risk. Automated systems for radiochemical sample preparation in radioanalytical laboratories have not enjoyed the same level of investment as radiochemical sample preparation methods conducted by the biomedical industry. Automatic radioisotopic separators, such as the one shown in Figure 6-2, are used in the biomedical industry for the repeated production of a specific radioisotope from a specific parent sample matrix. In such cases, the protocol does not change. The radiochemical separations required for samples of radiological tank waste do not fit this model since the chemical form and isotopic distribution will vary. This variation requires customization of the radiochemical analyses.

Figure 6-2. An Automated Radionuclide Separator Developed by Northstar Industries for ^{99m}Tc

Cross contamination is of particular concern when conducting radiochemical extractions for trace analytes in highly radioactive sample matrices, such as those associated with tank waste. As the automated radionuclide separators commercially available are designed for a specific separation for a specific analyte, the plumbing of the device becomes highly contaminated after a separation. This contamination is not a concern in typical biomedical applications since the input sample matrix and output radioisotopic product remain constant. However, the presence of internal contamination would lead to cross contamination if sample matrices and isotopes of interest vary between runs. The changing nature of the radiochemical analyses often required for tank waste samples makes the use of a commercially available isotope separator like those found in the biomedical community impractical. The reuse of the device by customizing analysis protocols and running a separation for one radiochemical species, and then using the same device to run a separation for a different species is not feasible. As some isotopes of interest are low in activity relative to interfering isotopes, robust separation protocols are required to remove interferences. The materials used for the separations become highly contaminated with these interfering isotopes and must be disposed of between sample extraction runs. The interfering isotopes can change from sample to sample, compromising a separation that was robust for one sample matrix but not for the next.

6.2.2 Proposed Research Approach:

The successful deployment of automated hands-off radiochemical separation systems would result in multiple benefits. Laboratory efficiency would be maximized because separations could be carried out during off-shift hours since much of the work could be automated and unattended. For example, round-the-clock separations could be conducted in laboratories staffed only with day personnel. Personnel exposure and dose would be reduced since many of the initial steps in the separation process would be automated, effectively removing personnel from the high-dose areas. The risk of personal contamination from sample manipulation within a radiological containment unit would also be greatly reduced.

In addition to reducing dose and contamination risks, the automated process would improve measurement sensitivity because larger aliquots of high-dose samples could be utilized. Currently, many of the preliminary separation steps must be carried out in the shielded cells facility. Only after these steps have successfully removed much of the dose from isotopes not being quantified can the remainder of the material be brought to the laboratories for completion of the analysis. De-coupling analyses from the Shielded Cells Facility will also accelerate schedule.

In order to develop this system, an automated system will be designed to perform automated reagent preparations, reagent additions and waste removal steps. The creation of this system will require the development of computer interfaced mechanisms for reagent mixing and delivery as well as waste neutralization and disposal. The environment will be highly acidic due to the reagents, which would reduce the lifetime of moving parts. Therefore, a peristaltic reagent pumping mechanism will be used to introduce reagents to the resin columns mounted on simple vacuum manifolds containing sample aliquots. An illustration of simple polyvinylchloride (PVC) vacuum manifolds is given in Figure 6-3.

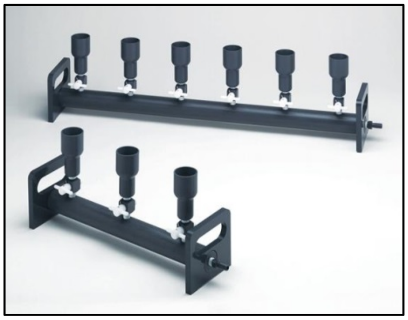


Figure 6-3. Simple PVC Vacuum Manifolds

This automated system will introduce reagents to the resin columns as programmed, will transfer the waste to a holding vessel, will adjust the pH of the liquid waste, and will finally discharge the waste into the radioactive drain system. The analyte of interest would remain on the resin columns, which would then be manually transferred to a vacuum box for the final step of the separation. The vacuum box which will be utilized for the final elution of the analyte of interest is shown in Figure 6-1.

The proposed system will consist of four computer-controlled pump systems: one to prepare and mix reagents, one to transfer prepared reagents to resin columns containing sample aliquots, one to transfer liquid waste to and from a mixing vessel, and one to add reagents to the liquid waste for pH adjustment. The first pump system will follow a programmed protocol to create the appropriate solutions used for the sample preparation and elution. The method will be programmed into the computer, which will control how the pump dispenses the solutions into the mixing vessel, allowing the in-situ dilution and mixing of solutions. The second pump, which transfers the prepared reagents to the resin columns containing sample aliquots, will have numerous disposable outlet lines, each attached to a disposable cap which will fit over the resin columns. Each cap will have the inlet tube, a vent port, and an attachable light sensor. The light sensor will act as a safety mechanism to detect clogs which could potentially lead to spills; if the solid phase extractant resin system becomes clogged, the pump will be switched off to prevent a spill. A third pump will provide reagent addition to the waste receptacle for pH adjustment, and the forth pump will facilitate waste transfer to the radioactive drain system.

The system is designed to provide hands-free analyte addition to resin columns and hands-free waste disposal. Following the addition of all of the reagents to the resin columns, the columns will contain only the analyte of interest since the interfering analytes (and their associated dose) will have been eluted from the columns and discarded. At this point in the analysis, the analyst would remove each resin column, transfer it into a vacuum box, and elute the analyte of interest.

The automated system would have a user friendly graphical user interface, programmed in LabVIEW or an equivalent. Various protocols will be defined to enable use with a wide range of radiochemical separation methods. The interface will be flexible enough to allow simple editing of protocols when adjustments are needed due to a specific sample matrix, or unique customer requirements.

6.2.3 Estimated Cost

The total R&D cost necessary to develop and test the automated system is estimated to be $250K.

6.3 <u>Improvements in Separation Protocols</u>

The recent Tank 12 Closure radiological characterization campaign called for the characterization of thirty-five radioisotopes. No substantial R&D effort was ever put into development of the radiochemistry protocols for SRS Closure Waste Tank Campaigns. Protocols were assembled quickly at the onset of the Tank 19 campaign, to meet aggressive characterization schedules. Incremental improvements to methodology have been carried out from Tank Campaign to Tank Campaign. These improvements were typically required as tank matrices and interfering radioisotopes changed from tank to tank. However, more deliberate improvements in some of the separation protocols could result in time and cost savings in a number of areas.

6.3.1 Lowering of Detection Limits for Targeted Radionuclides

One area would be to improve the chemical yields of the various radiochemical separations while maintaining the high levels of decontamination of the interfering radioisotopes. Improved yields can improve detection limits, and improved detection limits can result in less sample required for the analyses. Lower sample requirements can reduce the number of operations depending on both shielded cells and radiohood operations, which would accelerate schedule and reduce cost. In addition, lower sample requirements could significantly reduce the scope of the tank waste sampling campaigns and would result in lower dose to personnel.

Ra-226 is currently a candidate for which improving detection limits of the analysis could remove this isotope from the suite of isotopes being analyzed for in the Shielded Cells. Currently, aliquots of samples are digested by peroxide fusion in the Shielded Cells. The dissolutions are subjected to decontamination steps using chromatographic resins to remove Sr-90 and Y-90 as well as ammonium phosphomolybdate to remove Cs-137. Samples are removed from the Cells and the radium is extracted with cation exchange resin in the radiohoods. The radium extract is then analyzed using a high purity germanium well detector.

There are two potential areas of improvement in the radium analysis methodology. Optimization of the ion exchange extraction process, as well as exploration into the capabilities of some different extractions could lead to improved chemical yields. Optimization of the analysis equipment could also lead to other benefits. As the emissions measured for the radium measurements are relatively low in energy, a smaller well detector than the one currently used could be potentially be used, with the benefit of greater potential Compton suppression.

Efforts to reduce ambient radon, such as with a liquid nitrogen purge, or filling up the detector cavity with radon free material, could be investigated to further reduce the background and thus the minimum detection limits for the radium measurements. Pa-231 is another isotope that would likely benefit from optimization of radiochemical separation protocol, with the end goal of improving detection limits such that the initial Shielded Cells separations processing would no longer be necessary. In addition to addressing the Rad-226 and Pa-231 analyses, an in-depth review would be performed to identify other radiochemical methodologies that would benefit from improvement.

6.3.2 Consolidation of Shielded Cells Operations

An additional area of focus could be in combining the Shielded Cells decontamination steps for several protocols into a single protocol. This could reduce the number of Shielded Cells operations, both accelerating schedule and reducing cost. Additionally, it could reduce the amount of sample mass required, which could reduce the amount of supplies needed for these characterization campaigns – note that some of the chromatographic resins needed are also quite expensive.

There are two highly radioactive radionuclides (HRRs), Technetium-99 and Iodine-129, that undergo decontamination steps in the Shielded Cells prior to the hands on radiochemistry in the lab modules. The decontamination steps target removing the primary contributor to the whole body dose (Cs-137) and the primary contributors to the extremity dose (Sr-90 & Y-90). Currently, the decontamination steps for the I-

129 method are based primarily on the fact that strontium and yttrium are relatively insoluble in caustic solutions. The tank material is digested in nitric acid, the solution is rendered caustic, and Sr-90 and Y-90 precipitate out to a degree along with actinides and lanthanides. Additional decontamination is achieved with strikes of monosodium titanate (MST). Cs-137 levels are reduced with crystalline silicotitanate (CST). All of this decontamination is carried out in the presence of a potassium iodide tracer, to track recoveries of chemical iodine throughout the I-129 separation. As the oxidized state of technetium is also soluble in caustic, it is possible a similar decontamination would also be effective with Tc-99, and if the I-129 and Tc-99 decontamination steps could be combined into one Shielded Cells operation instead of two, there would be considerable time savings. Eliminating the use of chromatographic extraction resins for the decontamination of Tc-99 in the Shielded Cells, and instead utilizing solubility principles and relatively inexpensive titanates, would also offer a cost savings.

As mentioned previously, some of the radiochemical methods require extraction chromatography steps to be conducted in the Shielded Cells, prior to the further work-up in the lab modules. Some isotope protocols could be initially combined so that various elements could be extracted from one peroxide fusion digestion instead of multiple ones. Currently fusions are conducted for several different groups of actinides, one for thorium, one for protactinium, and one for americium, curium and californium isotopes. Experiments will be conducted to assess if it is practical to combine the initial steps in the Shielded Cells and then subsequently run several of these methods off the single peroxide fusion prep. This would result in savings, both from the aspect of time and in the volume of sample needed to complete a tank waste sample characterization campaign.

6.3.3 Separation of Yttrium from Trivalent Actinides

A third area that could be investigated would that of adjusting the protocols to separate Y-90 from the trivalent actinides. Y-90 is the short-lived daughter of Sr-90 and is the dominant beta emitter in tank waste solid samples. As such, Y-90 contributes the lions share to the extremity dose from tank closure samples. Yttrium is a trivalent element, like many of the actinides and lanthanides. It is often separated and trails along with radiochemical separations for these elements. Due to its high dose, following strontium separations in which it tags along, samples have to sit in the Shielded Cells for several weeks until the Y-90 decays away. Removing Y-90 from other analytes would eliminate the delay required for the Y-90 to decay, which would allow samples to exit the Shielded Cells quickly, accelerating the schedule for the tank waste radiological characterization campaigns. Aliquat 336 is an ammonium thiocyanate matrix that has been successfully used in the past to separate americium from the rare earth elements. Experiments will be conducted to determine if yttrium can also be removed. Experiments will also be conducted to establish that curium and californium will also co-extract with americium under these conditions. Additional extractants other than Aliquat 336 could also be explored as well.

6.3.4 Estimated Cost

The total R&D necessary to develop and test the proposed alternative separation protocols is estimated to be $250K.

6.4 Benefits from Investments into More Sensitive Instrumentation

The current Tank 12 Closure radiological characterization campaign calls for the characterization of 35 radioisotopes. Eight of these radioisotopes have half-lives long enough that mass spectrometry becomes a more sensitive analysis compared to a radioactivity analysis. Currently, SRNL’s mass spectrometry analyses are conducted using an Agilent quadrupole inductively coupled plasma mass spectrometer (ICP-MS) (see Figure 6-4).

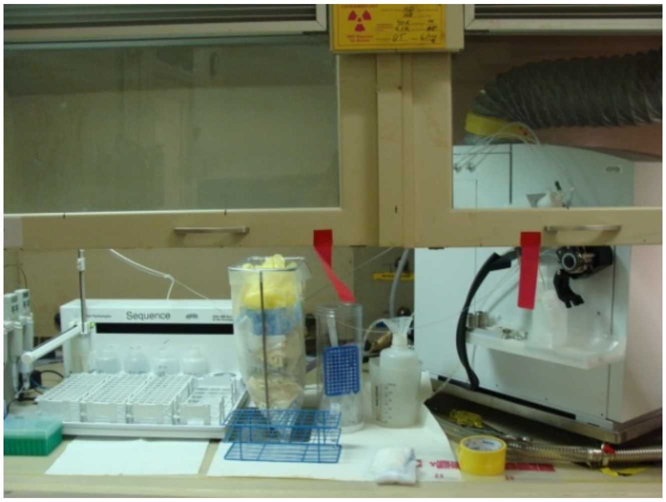


Figure 6-4. SRNL's Agilent Quadrupole ICP-MS

An ICP-MS combines a high-temperature ICP (Inductively Coupled Plasma) source with a mass spectrometer. The ICP source converts the atoms of the elements in the sample to ions. These ions are then separated and detected by the mass spectrometer. The mass spectrometer in the SRNL unit is based on a quadrupole mass filter. In a quadrupole mass filter, alternating AC and DC voltages are applied to opposite pairs of the rods. These voltages are then rapidly switched along with a radiofrequency (RF) field. The result is that an electrostatic filter is established that only allows ions of a single mass-to-charge ratio (m/e) pass through the rods to the faraday cup detector at a given instant in time. The voltages on the rods can be switched at a very rapid rate, so that the quadrupole mass filter can separate up to 2400 amu (atomic mass units) per second. The typical quadrupole mass spectrometers used in ICP-MS have resolutions between 0.7 - 1.0 amu.

In recent years, the state of the art in ICP-MS has transitioned from quadrupole mass spectrometers to high resolution magnetic sector mass spectrometers coupled to the ICP (see Figure 6-5). The resolutions of these systems are sufficient to separate overlapping molecular or isobaric interferences from the elemental isotope of interest. Besides high resolving power, another attractive feature of magnetic-sector instruments is their very high sensitivity combined with extremely low background levels. For this reason, minimum detection limits, especially for high-mass elements like uranium, are typically an order of magnitude lower than those provided by a quadrupole-based instrument. High resolution ICP-MS instruments are typically ordered with multiple magnetic sector detectors (multicollector high resolution ICP-MS or MC-ICP-MS), which both speeds up the time for scans and increases the precision of the measurements of isotopics in the scan.

Improvements in the detection limit capabilities of the SRNL mass spectrometer would benefit the tank closure radiological characterization campaign in several ways. Current protocols require initial treatment of sample aliquots in the Shielded Cells, followed by work up in the radiological hoods. Shielded Cells work puts considerable pressure on meeting schedules and budgets. To meet the currently required detection limits, considerable sample is used in the analysis. By using a more sensitive instrument, smaller aliquots could be used. Smaller aliquots translate into lower radiological doses, which would allow work to be conducted exclusively in the radiological hoods. Smaller sample aliquot needs would also significantly reduce waste tank sampling requirements for the tank closure operations. This would reduce both cost and risk to the schedule that arises from repeated entries into the tanks to generate sufficient sample material for the characterization campaign. In summary, a more sensitive ICP-MS has the potential to shave both significant time and costs from the tank closure characterization campaigns.

Figure 6-5. Thermo ELEMENT XR™ High Resolution (magnetic sector) ICP-MS

6.4.1 Estimated Cost

The total procurement and installation cost of the high resolution ICP-MS unit is estimated to be $1M. Use of this unit offers the potential for significant cost savings, through use of smaller sample aliquots (due to the higher measurement sensitivity), which offer the potential for eliminating a number of the highly resource-intensive Shielded Cells preparation activities. On average, each Shielded Cell prep increases the analysis cost on the order of $20K to $30K. This is particularly important in the Tank Closure characterization campaigns, where several Shielded Cells preps are typically performed, due to the low targeted minimum detection limits, the high relative radioactivities, and the complex sample matrices. The anticipated payback time for the high sensitivity ICP-MS unit is approximately five years, assuming two tank closure characterization campaigns are performed each year.

6.5 Conclusion

The potential exists for making current radiochemical laboratory methods more cost effective and rapid through various approaches, including: a) automation of radiochemical separation and waste removal processes; b) optimization of radiochemical separation protocols; and c) utilization of state-of-the- art mass spectrometric measurement technologies. Although such improvements will require R&D to be brought to fruition, the advantages of the new methods will ultimately benefit the full range of site tank waste characterization programs.

7.0 Overall Conclusions

1) At present, the greatest potential for reducing costs and schedule is in the tank closure characterization program. The second greatest potential is in the salt waste characterization program. The current focus should not be on sludge characterization, since it is a relatively small portion of the current characterization scope.

2) Potential approaches for increasing cost-effectiveness include:
- Elimination of characterization requirements for "negligible risk" constituents
- Improved lab methods that reduce Shielded Cells processing requirements and/or standard laboratory "hands on" processing times
- Replacement of labor-intensive methods with simpler methods, as appropriate
- Utilization of non-lab methods for characterizing "low risk" constituents
    - Waste receipt history, process knowledge, scaling factors, historic trends
- Reduce characterization frequency for constituents with "low risk" or stable history
- Raise targeted Minimum Detection Limits, as appropriate

3) Consideration of accuracy needs should feed characterization requirements
- "High risk" constituents are candidates for high accuracy quantification (± 20%)
- "Low risk" constituents may be candidates for order of magnitude estimates
- "Negligible risk" constituents may be candidates for elimination

4) Potential approaches for streamlining of characterization are a function of waste type, due to differences in program objectives and principal radionuclide impact measures
- Environmental risk (post-cleaning tank residue)
- WAC compliance (salt feed)
- Fraction of radioactivity (sludge batches)

5) Most promising options for streamlining tank closure characterization include:
- Utilization of methods that minimize need for Shielded Cells processing
- Development of alternative laboratory methods that increase productivity & reduce TATs
- Utilization of theoretical relationships to estimate long-term quantities of decay products
- Elimination or reduction of characterization of "negligible risk" constituents

6) Most promising options for streamlining salt characterization include:
- Reduction of frequency for characterizing "low risk" constituents
- Working with regulators to move from quarterly feed samples to bi-annually or annually, particularly for "low risk" constituents
    - Given current level of understanding, existing program seems excessive

7) Applicability to other DOE sites
- Provides a baseline for PUREX and HM tank waste characterization

8.0 Path Forward

1) Develop technical bases and strategies for improving cost effectiveness of:
   - Tank closure characterization program
   - Salt characterization program

2) Pursue development and testing of improved laboratory characterization methods
   - Automation of radiochemical separation and waste removal processes
   - Optimization of radiochemical separation protocols
   - High sensitivity instrumentation

3) Continue pursuing opportunities for making waste characterization more cost effective

4) Proposed scope over the next year:
   - Develop technical basis and strategy for improving cost effectiveness and schedule of SRNL's tank closure characterization program
   - Initiate design of hardware, plumbing, and software for automation of radiochemical separation and waste removal processes
   - Complete feasibility testing for at least two alternative radiochemical separation protocols (Ra-226, Pa-231, Tc-99/I-129, and/or Y/trivalent actinide separations)

Appendix A: Tank 50 Campaigns

METHOD	Radiochemical Method	DURATION -3Q13	DURATION - 4Q13	DURATION - 1Q14	DURATION - 2Q14	DURATION - 3Q14	Average of Five Batches
AAAS	N	25	25	19	56	18	29
AAK	N	26	26	33	34	18	27
AANA	N	26	26	32	34	18	27
AASE	N	25	20	19	34	18	23
CV HG	N	22	14	13	20	18	17
DBP BY IC ANIONS	N	18	16	63	55	56	42
HPLC	N	33	8	42	63	42	38
IC ANIONS	N	7	3	33	59	21	25
IC CATIONS	N	12	8	33	32	2	17
RAD ICPES LEEMAN	N	14	13	13	26	28	19
RAD ICPMS	N	22	32	32	37	36	32
RAD SPECIAL PREP	N	19	12	14	15	3	13
SVOA	N	22	40	55	43	32	38
T BASE/OH/OTHER BASE EXC CO3	N	18	29	29	7	15	20
TIC/TOC	N	11	15	19	8	15	14
VOA	N	42	42	59	55	32	46
AM/CM	Y	66	76	67	51	52	62
C-14	Y	14	32	33	15	46	28
GAMMA SCAN	Y	11	2	19	9	7	10
GAMMA SCAN CS REMOVED	Y	28	34	28	28	25	29
I-129 WITH SEPARATION	Y	11	22	19	27	66	29
LIQUID SCINT COUNTING	Y	11	20	19	23	21	19
LIQUID SCINT COUNTING (Cs-remove	Y	19	33	NA	20	22	24
NB-94	Y	NA	76	42	51	63	58
NI59/63	Y	25	76	34	30	35	40
PA-231	Y	68	NA	67	52	63	63
PM-147/SM-151	Y	27	32	34	30	43	33
PU238/PU241	Y	35	47	27	30	23	32
RA-226	Y	68	76	69	62	63	68
SE-79	Y	25	18	60	15	43	32
SR-90	Y	32	21	28	28	23	26
TC-99	Y	33	26	32	15	27	27
TH-229/230	Y	69	49	43	33	44	48
TRITIUM	Y	14	14	19	20	8	15
U-232	Y	25	22	40	58	52	39
	For Non-radiochemical Methods:						**Grand Metrics for Average (below):**
	Minimum =	**7**	**3**	**13**	**7**	**2**	**13**
	Maximum =	**42**	**42**	**63**	**63**	**56**	**46**
	Average for all methods =	**21**	**21**	**32**	**36**	**23**	**27**
	For Radiochemical Methods:						
	Minimum =	**11**	**2**	**19**	**9**	**7**	**10**
	Maximum =	**69**	**76**	**69**	**62**	**66**	**68**
	Average for all methods =	**32**	**38**	**38**	**31**	**38**	**36**
			🟩 Best-case				
			🟥 Worst-case				

Appendix B: Salt Batch Campaigns

METHOD	**Radiochemical Method**	**DURATION - Salt Batch 5**	**DURATION - Salt Batch 6**	**DURATION - Salt Batch 7**	**Average for 3 Batches**
AAAS	N	7	19	19	15
AASE	N	7	13	19	13
CV HG	N	8	13	19	13
HPLC	N	35	20	45	33
IC ANIONS	N	NA	6	8	7
IC CATIONS	N	34	6	26	22
RAD ICPES LEEMAN	N	NA	7	9	8
RAD ICPMS	N	7	21	30	19
SVOA	N	2	NA	45	24
T BASE/OH/OTHER BASE EXC CO3	N	NA	8	14	11
TIC/TOC	N	NA	8	11	10
VOA	N	24	NA	NA	24
AM/CM	Y	55	46	45	49
C-14	Y	41	20	31	31
GAMMA SCAN	Y	NA	19	7	13
GAMMA SCAN CS REMOVED	Y	49	NA	27	38
I-129 WITH SEPARATION	Y	29	26	21	25
LIQUID SCINT COUNTING	Y	37	22	21	27
NB-94	Y	NA	46	69	58
NI59/63	Y	41	21	35	32
NP-237 W MS	Y	52	NA	NA	52
PM-147/SM-151	Y	41	42	21	35
PU238/PU241	Y	30	22	27	26
SR-90	Y	50	48	31	43
TC-99	Y	29	42	15	29
TRITIUM	Y	27	14	27	23
U-232	Y	36	43	20	33
	For Non-radiochemical Methods:				**Grand Metrics for Average (below):**
	Minimum =	**2**	**6**	**8**	**7**
	Maximum =	**35**	**21**	**45**	**33**
	Average for all methods =	**16**	**12**	**22**	**17**
	For Radiochemical Methods:				
	Minimum =	**27**	**14**	**7**	**13**
	Maximum =	**55**	**48**	**69**	**58**
	Average for all methods =	**40**	**32**	**28**	**34**
			🟩 Best-case		
			🟥 Worst-case		

Appendix C: Sludge Batch Campaigns

METHOD	Radiochemical Method	DURATION - Sludge Batch 7A	DURATION - Sludge Batch 7B	DURATION - Sludge Batch 8	Average of Three Batches
AAAS	N	NA	36	28	32
AASE	N	NA	36	28	32
CV HG	N	17	23	2	14
HLC-ALKALI FUSION-PREP CHARGE	N	2	9	16	9
HLC-AQUAREGIA-PREP CHARGE	N	2	15	5	7
IC ANIONS	N	NA	NA	5	5
RAD ICPES LEEMAN	N	14	22	10	15
RAD ICPES LOW S LEEMAN	N	14	16	8	13
RAD ICPMS	N	1	21	7	10
RAD ICPMS FOR IODIDE	N	NA	27	NA	27
AM/CM	Y	58	78	147	94
CL-36	Y	114	91	168	124
CS-135	Y	70	92	NA	81
GAMMA SCAN	Y	7	3	28	13
GAMMA SCAN CS REMOVED*	Y	14	7	28	16
I-129 WITH SEPARATION	Y	37	36	38	37
LIQUID SCINT COUNTING	Y	8	9	28	15
NI59/63	Y	32	10	28	23
PA-231	Y	NA	NA	192	192
PM-147/SM-151	Y	37	23	28	29
PU238/PU241	Y	14	15	28	19
RA-226	Y	NA	NA	170	170
SE-79	Y	38	76	128	81
SR-90	Y	37	29	29	32
TH-229/230	Y	NA	NA	189	189
TRITIUM	Y	3	8	26	12
U-233, 234, 235, 236	Y	NA	NA	189	189
	For Non-radiochemical Methods:				**Grand Metrics for Average (below):**
	Minimum =	**1**	**9**	**2**	**5**
	Maximum =	**17**	**36**	**28**	**32**
	Average for all methods =	**8**	**23**	**12**	**16**
				14	
	For Radiochemical Methods:				
	Minimum =	**3**	**3**	**26**	**12**
	Maximum =	**114**	**92**	**192**	**192**
	Average for all methods =	**36**	**37**	**90**	**77**
				54	
			🟩 Best-case		
			🟥 Worst-case		

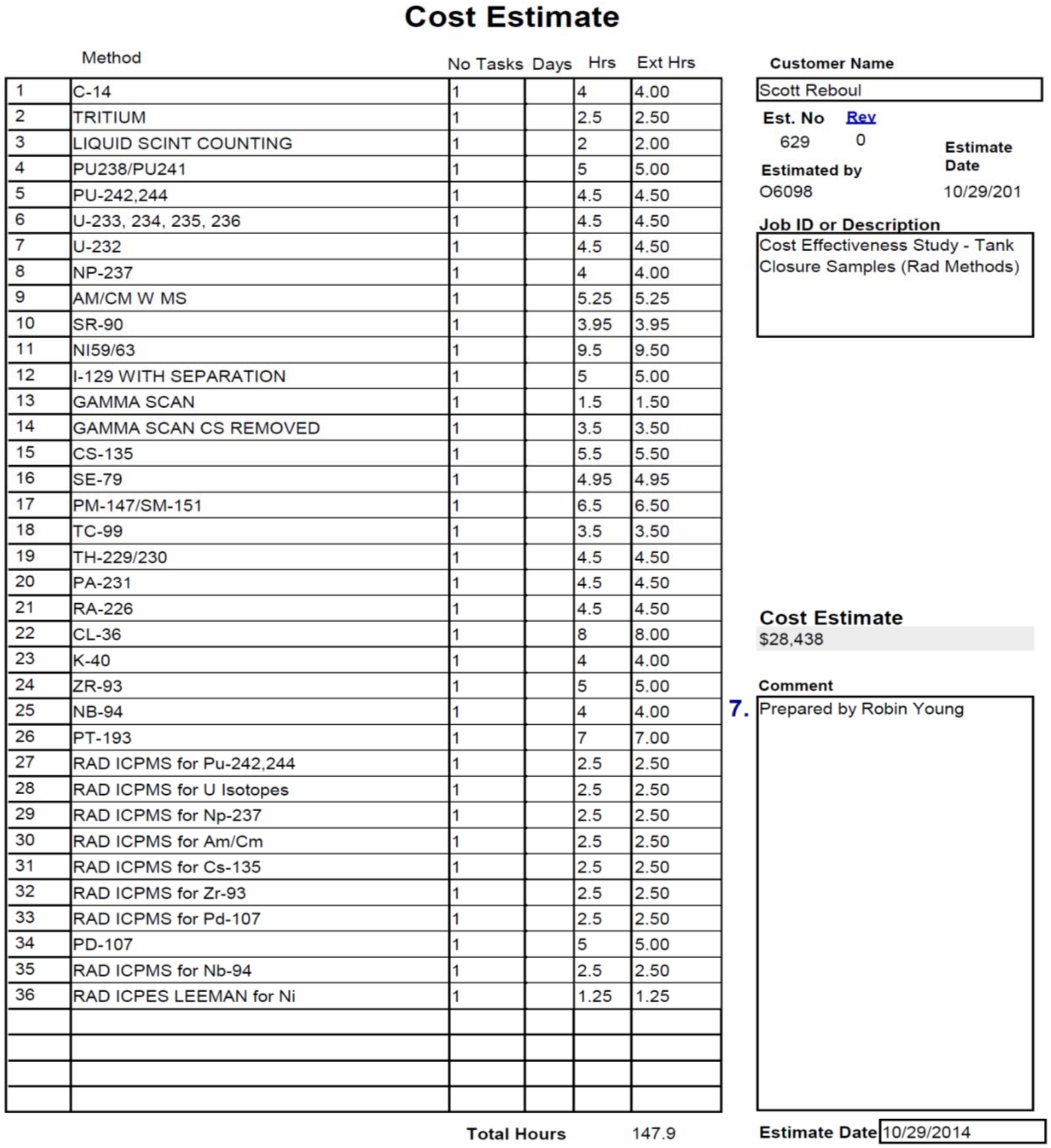
Appendix D: Tank Closure Campaigns

METHOD	Radiochemical Method	DURATION - Tank 5	DURATION - Tank 6	DURATION - Tank 16 Primary	DURATION - Tank 16 Annulus	Average for Four Tanks
AAAS	N	7	16	48	56	32
AASE	N	6	14	48	56	31
CV HG	N	7	14	48	56	31
HLC-ALKALI FUSION	N	16	3	5	12	9
HLC-AQUAREGIA*	N	18	1	6	4	7
IC ANIONS	N	22	35	12	27	24
RAD ICPES LEEMAN	N	43	51	23	28	36
RAD ICPMS*	N	34	58	88	118	75
AM/CM	Y	200	112	228	159	175
C-14	Y	184	133	231	125	168
CL-36	Y	199	NA	213	167	193
CS-135	Y	189	73	225	144	158
GAMMA SCAN*	Y	65	16	90	52	56
GAMMA SCAN CS REMOVED	Y	79	35	201	118	108
I-129 WITH SEPARATION	Y	79	51	38	111	70
K-40	Y	NA	NA	228	165	197
LIQUID SCINT COUNTING	Y	15	71	NA	NA	43
NB-94	Y	203	150	225	158	184
NI59/63	Y	97	51	199	67	104
NP-237 W MS	Y	204	112	NA	NA	158
PA-231	Y	199	127	217	173	179
PM-147/SM-151	Y	73	34	NA	NA	54
PT-193	Y	200	NA	NA	NA	200
PU238/PU241	Y	38	34	90	111	68
PU-242,244	Y	67	44	266	173	138
RA-226	Y	204	153	239	158	189
SE-79	Y	154	99	170	NA	141
SR-90	Y	63	34	83	117	74
TC-99	Y	190	45	44	90	92
TH-229/230	Y	164	129	212	144	162
TRITIUM	Y	126	37	NA	NA	82
U-232	Y	119	128	NA	NA	124
U-233, 234, 235, 236	Y	67	62	88	119	84
ZR-93	Y	189	115	NA	167	157
	For Non-radiochemical Methods:					**Grand Metrics for Average (below):**
	Minimum =	**6**	**1**	**5**	**4**	**7.25**
	Maximum =	**43**	**58**	**88**	**118**	**75**
	Average for all methods =	**19**	**24**	**35**	**45**	**31**
	For Radiochemical Methods:					
	Minimum =	**15**	**16**	**38**	**52**	**43**
	Maximum =	**204**	**153**	**266**	**173**	**200**
	Average for all methods =	**135**	**80**	**173**	**133**	**129**
			Best-case			
			Worst-case			

Appendix E: Comparison of Costs For Non-radiochemical and Radiochemical Methods Associated with Varied Sample Campaigns

Campaign	**Batch**	**# of Samples**	**LIMS Cost Estimate**	**LIMS Actuals**	**Direct Charges**	**Total Cost**	**Per-Sample Cost**	**Total # Methods**
Tank 50	3Q 2013	3	71000	68,022.65	11,110.37	79,133.02	26,377.67	35
Supernate	4Q 2013	3	71000	51,507.31	9,523.87	61,031.18	20,343.73	35
	1Q 2014	3	71000	90,362.44	20,745.91	111,108.35	37,036.12	35
	2Q 2014	3	71000	85,325.09	22,364.03	107,689.12	35,896.37	35
	3Q 2014	3	71000	27,404.10	16,987.61	44,391.71	14,797.24	35
	Averages for Tank 50		71000	64,524.32	16,146.36	80,670.68	26,890.23	
Salt Batch	SB 5	2	47000	47,737.17	28,923.89	76,661.06	38,330.53	28
Supernate	SB 6	2	47000	36,729.10	13,537.46	50,266.56	25,133.28	28
	SB 7	2	47000	38,165.88	11,633.12	49,799.00	24,899.50	28
	Averages for Salt Batches		47000	40,877.38	18,031.49	58,908.87	29,454.44	
Sludge Batch	SB 7A	5	113600	85,480.54	16,968.02	102,448.56	20,489.71	27
Solids	SB 7B	5	113600	95,333.77	69,329.90	164,663.67	32,932.73	27
	SB 8	5	113600	116,824.47	18,108.79	134,933.26	26,986.65	27
	Averages for Sludge Batches		113600	99,212.93	34,802.24	134,015.16	26,803.03	
Tank Closure	Tanks 5 &6	24	783000	800,448.05	1,121,358.51	1,921,806.56	80,075.27	34
Solids	Tanks 16 P&A	24	783000	817,517.05	392,200.97	1,209,718.02	50,404.92	34
	Averages for Tank Closure Batches		783000	808,982.55	756,779.74	1,565,762.29	65,240.10	

Appendix F: Example of LIMS Estimating Worksheet



Cost Estimate

	Method	No Tasks	Days	Hrs	Ext Hrs
1	C-14	1		4	4.00
2	TRITIUM	1		2.5	2.50
3	LIQUID SCINT COUNTING	1		2	2.00
4	PU238/PU241	1		5	5.00
5	PU-242,244	1		4.5	4.50
6	U-233, 234, 235, 236	1		4.5	4.50
7	U-232	1		4.5	4.50
8	NP-237	1		4	4.00
9	AM/CM W MS	1		5.25	5.25
10	SR-90	1		3.95	3.95
11	NI59/63	1		9.5	9.50
12	I-129 WITH SEPARATION	1		5	5.00
13	GAMMA SCAN	1		1.5	1.50
14	GAMMA SCAN CS REMOVED	1		3.5	3.50
15	CS-135	1		5.5	5.50
16	SE-79	1		4.95	4.95
17	PM-147/SM-151	1		6.5	6.50
18	TC-99	1		3.5	3.50
19	TH-229/230	1		4.5	4.50
20	PA-231	1		4.5	4.50
21	RA-226	1		4.5	4.50
22	CL-36	1		8	8.00
23	K-40	1		4	4.00
24	ZR-93	1		5	5.00
25	NB-94	1		4	4.00
26	PT-193	1		7	7.00
27	RAD ICPMS for Pu-242,244	1		2.5	2.50
28	RAD ICPMS for U Isotopes	1		2.5	2.50
29	RAD ICPMS for Np-237	1		2.5	2.50
30	RAD ICPMS for Am/Cm	1		2.5	2.50
31	RAD ICPMS for Cs-135	1		2.5	2.50
32	RAD ICPMS for Zr-93	1		2.5	2.50
33	RAD ICPMS for Pd-107	1		2.5	2.50
34	PD-107	1		5	5.00
35	RAD ICPMS for Nb-94	1		2.5	2.50
36	RAD ICPES LEEMAN for Ni	1		1.25	1.25

Total Hours 147.9

Customer Name
Scott Reboul

Est. No	**Rev**	**Estimate Date**
629	0	

Estimated by
O6098

Estimate Date
10/29/201

Job ID or Description
Cost Effectiveness Study - Tank Closure Samples (Rad Methods)

Cost Estimate
$28,438

Comment
7. Prepared by Robin Young

Estimate Date 10/29/2014

Appendix G: Comparison of Radiological Method Labor Costs for Tank Closure Campaigns - SRNL Shielded Cells and AD

METHOD	Shielded Cell Hours	Shielded Cell Costs	LIMS hours/Repl	Total LIMS Hours	LIMS Cost	Tech Hours	Tech Cost	Exempt Hours	Exempt Costs	Total Costs	Relative Cost, %
AM/CM w/ MS	180	27200	8	96	18816	3	420	65	16800	63236	100
C-14	13	2320	4	48	9408	8	1120	45	12000	24848	39
CL-36	90	13200	8	96	18816	6	840	65	16800	49656	79
CS-135 w/ MS	12	2280	8	96	18816	8	1120	50	13200	35416	56
GAMMA SCAN CS REMOVED	12	2280	3.5	42	8232	8	1120	50	13200	24832	39
GAMMA SCAN*	12	2280	1.5	18	3528	4	560	50	13200	19568	31
I-129 WITH SEPARATION	84	13760	5	60	11760	6	840	65	16800	43160	68
K-40	24	3960	4	48	9408	4	560	50	13200	27128	43
LIQUID SCINT COUNTING	12	2280	2	24	4704	4	560	40	10800	18344	29
NB-94	12	2280	4	48	9408	7	980	45	12000	24668	39
NI59/63 w/ES	10	1400	9.5	114	22344	8	1120	55	14400	39264	62
NP-237 w/ MS	12	1980	6.5	78	15288	6	840	50	13200	31308	50
PA-231	180	27200	7	84	16464	5	700	45	12000	56364	89
PM-147/SM-151	12	2280	6.5	78	15288	4	560	55	14400	32528	51
PT-193 w/ES	12	2280	8	96	18816	6	840	45	12000	33936	54
PU238/PU241	12	2280	5	60	11760	9	1260	55	14400	29700	47
PU-242,244 w/ MS	12	2280	7	84	16464	4	560	45	12000	31304	50
RA-226	148	22720	4.5	54	10584	12	1680	45	12000	46984	74
SE-79	12	2280	5	60	11760	5	700	55	14400	29140	46
SR-90	12	2280	4	48	9408	5	700	55	14400	26788	42
TC-99	100	16000	3.5	42	8232	6	840	65	16800	41872	66
TH-229/230	180	27200	4.5	54	10584	9	1260	50	13200	52244	83
TRITIUM	12	2280	2.5	30	5880	4	560	45	12000	20720	33
U-232	12	2280	4.5	54	10584	6	840	50	13200	26904	43
U-233, 234, 235, 236 w/MS	12	2280	7	84	16464	6	840	40	10800	30384	48
ZR-93 w/MS	12	2280	7.5	90	17640	6	840	45	12000	32760	52
	Totals ====>	191140			330456		22260		349200	893056	

NOTE: Table does not include any costs for materials.

Number of replicates (Repl) 12

Appendix H: Supporting Material Extracted from F-Area Tank Farm PA

5.5.1 Member of the Public at 100m Groundwater Pathway Dose Results

An overview of the modeling results indicate:

- Dose peaks prior to year 3,600 are associated with ancillary equipment releases, in particular the transfer lines, which are distributed throughout the FTF and therefore affect all sectors.
- Dose peaks near year 700 are associated with Tc-99 and Np-237 from the ancillary equipment.
- Dose peaks near year 1,800 are associated with Ra-226 from the ancillary equipment and also the tail end of the Np-237 release from the ancillary equipment.
- The dose peak near 3,750 is associated with I-129 from the Type IV tanks.
- Dose peaks between years 3,750 and 12,700 are tied primarily to releases from the Type IV tanks. The peaks after year 12,700 are tied to the tail end of releases from the Type IV tanks and the start of releases from the Type I and III/IIIA tanks. The Type IV tank liners are considered to fail at approximately year 3,750 while the Type I and III/IIIA tanks don't fail until approximately year 12,700.
- Dose peaks near year 4,300 have a contribution from C-14 in the Type IV tanks.
- Sector D and E doses between approximately year 4,000 and 20,000 have a significant Ra-226 contribution. The Ra-226 contribution is tied to the release and travel of U-234. The largest Type IV tank U-234 inventory is in Tank 18 (0.38 Ci). The Ra-226 inventories in the Type IV tanks are insignificant.
- Sector D and E dose peaks near year 6,000 are associated with Np-237 from the Type IV tanks. The largest Type IV tank Np-237 inventories are in Tank 18 (0.24 Ci). Tank 18 also has 82 Ci of Am-241, which is a parent of Np-237.
- The dose at approximately year 6,000 has a significant contribution from Cs-135 in the Type IV tanks.
- Sector D and E dose peaks at year 10,000 are associated with Ra-226 and Np-237, but also with relatively slow moving radionuclides just reaching the 100m boundary. The radionuclides that were released from the Type IV tanks that are starting to be seen at year 10,000 include U-233, U-234, and Th-229.

5.5.1.2 Individual Radionuclide Contributions to the MOP 100m Peak Annual Groundwater Pathway Dose

The peak groundwater pathway dose to the MOP at 100m during the 10,000 years evaluation period of 2.3 mrem/yr is primarily associated with Ra-226 (40%) and Np-237 (23%). The top contributors (>5% contribution) to the MOP at 100m peak groundwater pathway dose are Cs-135, Ra-226, U-233, U-234, and Np-237.

Table 5.5-4: Member of the Public at 100m Peak Dose Individual Groundwater Pathway Contributions for Sector E at 10,000 years.

Pathway	Associated Contribution at year 10,000 (mrem/yr)	Percentage of Total Peak Dose	Principal Radionuclide Pathway Dose
Water Ingestion	1.48	64%	Ra-226 (42%)
Vegetable Ingestion	0.66	29%	Ra-226 (42%)
Finfish Ingestion	0.14	6%	Cs-135 (86%)
Milk Ingestion	0.01	1%	Ra-226 (79%)
Beef Ingestion	0.01	<1%	Ra-226 (44%)
TOTAL	**2.30**	**100%**	

Radionuclides with a contribution of less than 0.1 mrem/yr were included in the sensitivity analysis because they had a significant (i.e., > 0.1 mrem/yr) impact on progeny; Am-241 (for Np-237), Am-243 (for Pu-239), Cm-244 (for Pu-240), Pu-238 (for Ra-226), Th-230 (for Ra-226), and U-235 (for Pa-231).

Appendix I: Supporting Material Extracted from H-Area Tank Farm PA

5.5.1 MOP at 100-Meter Groundwater Pathway Dose Results

An overview of the modeling results indicate:

- Early dose peaks (prior to year 2,500) are associated with the inventory from ancillary equipment (including transfer lines), from sand pads under Type II tanks, and tanks assumed to have failed steel liners at the time of closure (Tanks 12, 14, 15 and 16).
- Later dose peaks result from the loss of containment due to failure of the steel liner (Type IV tanks at year 3,638; Type I tanks at year 11,397; Type II tanks at year 12,687; and Type III and IIIA tanks at year 12,751).
- Peak doses to the MOP within 10,000 years at the 100-meter boundary are primarily from Tc-99, Pa-231, and Ra-226 from the groundwater pathways in Sectors A, B, and C.

Table 5.5-2: MOP at 100-Meter Peak Groundwater Pathway Dose Individual Radionuclide Contributions at Peak Years – Sectors C (1,000 years) and A (10,000 years).

Radionuclide	**Contribution to Sector C Peak dose at year 700 (mrem/yr)**	**Percentage of Total Peak Dose**	**Contribution to Sector A Peak dose at year 8,790 (mrem/yr)**	**Percentage of Total Peak Dose**
I-129	< 0.01	< 0.5 %	< 0.01	< 0.5 %
Nb-93m	0.01	1.70%	< 0.01	< 0.5%
Nb-94	< 0.01	< 0.5 %	0.02	0.60%
Np-237	0.03	10%	0.04	0.90%
Pa-231	< 0.01	< 0.5 %	0.05	1.20%
Pu-239	< 0.01	< 0.5 %	< 0.01	< 0.5 %
Ra-226	< 0.01	< 0.5 %	0.06	1.50%
Tc-99	0.27	88%	3.8	96%
Others	< 0.01	< 0.5 %	< 0.01	< 0.5 %

Table 5.5-3: MOP at 100-Meter Peak Groundwater Pathway Dose Individual Source Contributions at Peak Years - Sectors A and C Peak.

Waste Source[a]	Contribution to Sector C Peak Dose at year 700 (mrem/yr)	Percentage of Total Peak Dose	Contribution to Sector A Peak Dose at year 8,790 (mrem/yr)	Percentage of Total Peak Dose
Tanks 9, 10, and 11	< 0.01	< 0.5 %	3.8	95 %
Tank 12	< 0.01	< 0.5 %	0.08	2.0 %
Tank 13	< 0.01	< 0.5 %	< 0.01	< 0.5 %
Tanks 14 and 15	0.05	10 %	0.01	< 0.5 %
Tank 16	0.13	28 %	< 0.01	< 0.5 %
Tank 22	< 0.01	< 0.5 %	0.01	< 0.5 %
Tanks 21, 23, and 24	< 0.01	< 0.5 %	0.07	1.9 %
Transfer Line, Group 2 (Type I and Type II)	0.09	18%	< 0.01	< 0.5 %
Transfer Line, Group 3 (West Hill)	< 0.01	< 0.5 %	< 0.01	< 0.5 %
All Other Sources	0.19	40 %	< 0.01	< 0.5 %
TOTAL	0.47	100 %	4.0	100 %

[a] The Type III and IIIA tanks (Tanks 29, 30, 31, 32, 35, 36, 37, 38, 39, 40, 41, 42, 43, 48, 49, 50, and 51) do not fail prior to 10,000 years and are excluded from this table because their contribution to peak doses is 0 %.

Table 5.5-6: MOP at 100-Meter Peak Water Ingestion Doses by Sector.

Sector	Peak Water Ingestion Dose in 1,000 years (mrem/yr)	Principal Radionuclide	Peak Water Ingestion Dose in 10,000 years (mrem/yr)	Principal Radionuclide
A	0.17 (year 860)	Tc-99 (95 %)	2.3 (year 8,790)	Tc-99 (93 %)
B	0.14 (year 720)	Tc-99 (99 %)	1.2 (year 9,330)	Ra-226 (32 %) Pa-231 (29 %) Tc-99 (19 %)
C	0.19 (year 700)	Tc-99 (83 %)	1.9 (year 10,000)	Ra-226 (58 %) Pa-231 (20 %)
D	0.023 (year 880)	Tc-99 (96 %)	0.091 (year 10,000)	Ra-226 (81 %)
E	0.081 (year 870)	Tc-99 (97 %)	0.081 (year 870)	Tc-99 (97 %)
F	0.087 (year 870)	Tc-99 (91 %)	0.081 (year 870)	Tc-99 (91 %)

In order to fully evaluate the contribution of the sensitivity run radionuclides, some radionuclides with a contribution of less than 0.25 mrem/yr at the 100-meter boundary were included because they had a significant (i.e., > 0.25 mrem/yr) impact on progeny; Am-241 (for Np-237), Pu-238 (for Ra-226), Pu-239 (for Pa-231), Th-230 (for Pb-210 and Ra-226), U-234 (for Pb-210 and Ra-226), and U-235 (for Pa-231).

Appendix J: Supporting Material Extracted from Saltstone Disposal Facility PA

5.2.2 Key Radionuclide Determination

The key radionuclides were determined based on the peak all-pathways doses calculated using the 100m groundwater concentrations listed in Section 5.2.1. The peak doses for each individual radionuclide are not necessarily in the same year. Any radionuclide with a peak individual all-pathways dose (assuming Base Case pathways and assumptions) greater than 0.05 mrem/yr was considered a key radionuclide. The key radionuclide determination was conducted based on the peak all-pathways doses within 20,000 years. The screening conclusions are provided in Table 5.2-21. The resulting key radionuclides are Tc-99, I-129, Ra-226, Np-237 and Pa-231. The 0.05 mrem/yr screening threshold was considered sufficiently low enough that no radionuclides that were screened out would contribute appreciably to the peak dose results. The screening evaluation indicated that there were also no radionuclides screened with a peak dose greater than 0.02 mrem/yr but less than 0.05 mrem/yr. In order to evaluate the contribution of the key radionuclides, not only was transport of the key radionuclides modeled, but also transport of the parents of the key radionuclides; U-235 (for Pa-231), Th-230 (for Ra-226), U-234 (for Ra-226), and Pu-238 (for Ra-226) that contributed significantly to the peak dose.

Table 5.2-21: Determination of Key Radionuclides

Peak all-Pathways Dose Contribution in 20,000 Years (mrem/yr)					
Sector	**Tc-99**	**I-129**	**Ra-226**	**Np-237**	**Pa-231**
A	0.8	1.3	1.9	0.1	< 0.1
B	0.9	1.4	2.2	0.1	0.1
C	0.3	1.3	1.2	0.1	< 0.1
D	0.2	1.1	0.8	< 0.05	< 0.05
E	0.6	1.3	1.6	< 0.05	< 0.05
F	0.1	1	0.6	< 0.05	< 0.05
G	0.2	2.4	0.8	< 0.05	< 0.05
H	0.2	2.4	0.8	< 0.05	< 0.05
I	0.3	2.7	0.8	< 0.05	< 0.05
J	0.2	2.3	0.8	< 0.05	< 0.05
K	0.2	2.4	0.8	< 0.05	< 0.05
L	0.2	1.9	0.7	< 0.05	< 0.05

Distribution:

M. J. Barnes, 773-A
T. B. Brown, 773-A
J. R. Cantrell, 705-1C
M. J. Cercy, 773-42A
D. A. Crowley, 773-43A
D. P. DiPrete, 773-42A
S. D. Fink, 773-A
M. S. Hay, 773-42A
C. C. Herman, 773-A
E. N. Hoffman, 999-W
W. D. King, 773-42A
M. J. Mahoney, 705-1C
S. L. Marra, 773-A
D. H. McGuire, 999-W
J. M. Pareizs, 773-A
F. M. Pennebaker, 773-42A
G. R. Peterson, DOE HQ
S. H. Reboul, 773-42A
S. R. Ross, 704-S
F. G. Smith, III 703-41A
P. G. Suggs, 704-S
S. A. Thomas, 705-1C
W. R. Wilmarth, 773-A
R. H. Young, 773-A
Records Administration (EDWS)