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Pursuing Cost Effective Tank Waste Characterization at the Savannah River Site – 16172

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ABSTRACT

The U. S. Department of Energy Office of Environmental Management has tasked the Savannah River National Laboratory (SRNL) with developing strategies and technologies to understand, optimize, scale, and speed up tank waste characterization. This scope is part of a multi-year project with the end goal of implementing programmatic changes that accelerate tank waste processing and tank closure schedules, and at the same time reduce characterization costs, while maintaining data integrity. The project is currently in its second year.

During the first year of the project, SRNL completed a series of activities focused on understanding the range of current characterization practices, needs, gaps, risks, and potential alternative approaches. Based on the results, conclusions were drawn regarding aspects of the programs that are most costly and time consuming; particular waste constituents that drive costs, schedule, and program risks; alternative characterization approaches (other than sampling and analysis) that are less expensive and more rapid; new laboratory methods holding the greatest promise for reducing characterization costs and schedule; and in whole, the primary areas where current characterization practices can be improved.

SRNL's current scope is focusing on three primary activities: 1) development of a technical basis/strategy for improving the cost effectiveness and schedule of SRNL's tank closure characterization program; 2) design/assembly of hardware, plumbing, and software for automating select radiochemical separation and waste removal processes; and 3) development and testing of alternative radiochemical separation protocols most likely to improve high resource demand/time consuming analysis methods (such as Ra-226, Pa-231, Tc-99/I-129, and/or Y/trivalent actinide separations).

INTRODUCTION

Extensive characterization of tank waste is performed at DOE sites in support of ongoing waste processing, waste disposition, and tank closure activities. At the Savannah River Site, characterization is routinely performed to process and disposition batches of salt waste and sludge waste, and to quantify inventories of residual material prior to closing emptied and cleaned waste tanks. In each characterization campaign, several dozen radionuclides and stable constituents are quantified to meet waste acceptance requirements and/or regulatory requirements. Because the radioactivity content of the waste is high, and because the waste matrices are typically highly variable and complex, the characterization activities are extremely resource intensive and time consuming. Correspondingly, the costs and time requirements of the characterization activities are high – so high that they can limit the progress of other site activities and impede the ability to meet regulatory commitments.

Given this situation, 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." This scope is part of a multi-year continuing project (currently in its second year) with the end goal of implementing programmatic changes that accelerate tank waste processing and tank closure schedules, and at the same time reduce characterization costs, while maintaining data integrity. The specific near-term primary project objectives are: a) to identify opportunities for improving characterization practices in the context of reducing cost and schedule; and b) to develop and evaluate potential alternative characterization methodologies.

During the first year of the project, SRNL completed the following five activities: 1) identification of SRNL's 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 laboratory characterization methods holding promise for being less costly and/or less time consuming [1]. (Note that Reference 1 contains the details of the first year activities and a comprehensive listing of the applicable reference sources utilized in the project).

Based on the results, conclusions were drawn regarding the aspects of SRNL's current characterization programs that are most costly and time consuming; the particular waste constituents that drive the costs, schedule, and program risks; the potential recommended alternative characterization approaches (other than sampling and analysis) that are less expensive and more rapid; new laboratory methods that hold the greatest promise for reducing characterization costs and schedule; and in whole, the primary areas where the characterization practices can be improved.

During the second year of the project, the focus has been on the following three activities: 1) development of a technical basis and strategy for improving the cost effectiveness and schedule of SRNL's tank closure characterization program; 2) initiation of the design and assembly of hardware, plumbing, and software for automating select radiochemical separation and waste removal processes; and 3) development and feasibility testing of at least two alternative radiochemical separation protocols holding promise for improving high resource demand/time consuming analysis methods.

Results of this project will ultimately provide the bases 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.

BACKGROUND AND APPROACH

Characterization Activities Driving Cost and Schedule

Extensive laboratory analyses of radionuclides in a multitude of SRS tank waste samples have been performed by SRNL over the past several decades. This includes characterization of numerous HLW sludge, salt, and tank closure residue samples. Typically, each sample is characterized for dozens of individual radionuclides, along with a routine series of stable constituents. Many of the radionuclide analyses involve multiple cycles of radiochemical separations, to ensure removal of interfering nuclides and to achieve low minimum detection limits. In many cases, the time requirements for completion of the radionuclide analyses are several months, and the respective costs are commensurately high. In this task activity, the costs and time requirements of the various analyses were compiled and analyzed, for the purpose of identifying average costs and durations associated with each type of analysis and each type of characterization campaign. The results were used to identify analyses and campaigns for which alternative characterization methods should be pursued.

Potential Streamlining of Characterization Requirements Based on the Relative Constituent Risks

Performance Assessment (PA) analyses are used to evaluate the expected dose to a hypothetical person from the release of radionuclides from waste disposal sites into the environment. This includes waste disposal sites associated with recently emptied and cleaned tanks (Tanks 5, 6, 18, 19, and 16), as well as four types of Saltstone vaults constructed for disposal of stabilized salt waste. In this task activity, a conservative estimate of the inventory in a waste disposal unit (e.g. residual material in a waste tank or stabilized salt waste) requiring characterization was “screened” using a simplified PA type analysis to identify radionuclides that contribute significantly to dose and therefore pose the greatest risk. PA analyses typically consider on the order of 60 different radionuclides. However, results from the analysis show that only a small number of radionuclides and daughters are responsible for most of the dose. The remaining constituents could be considered low risk allowing reduced characterization requirements. The screening approach utilized a one-dimensional transport model, similar to that employed in the SRS Composite Analysis (CA), to estimate the expected dose from a waste disposal site. The screening model was used to quickly evaluate the applicability and utility of this approach to waste characterization requirements. The end goal is to provide a basis for streamlining the characterization requirements based on the relative constituent risks.

Determination of the Relative Usefulness of Potential Characterization Bases Other than Sampling and Analysis

Historically, most characterization of SRS tank waste has been accomplished utilizing laboratory analyses of real-waste samples. Typically, the data generated through the laboratory analyses has been utilized on its own, in the absence of other waste considerations, including the tank waste receipt history, process knowledge, technically-based constituent scaling relationships, and other potential characterization bases. In this task activity, the relative usefulness of such alternative characterization bases was assessed, with the intent of establishing alternative technical bases, where justified, either as a complement to the analytical data or as a potential standalone source of data. This is particularly important in cases where the existing laboratory method is costly and time consuming or in cases where the constituent being characterized has minimal or no practical impact on the disposition decisions/requirements.

Honing of Characterization Needs as a Function of Waste Type

For sludge batches, a primary use of the characterization data is for determining which radionuclides are reportable; for salt batches, the primary uses include demonstration of compliance with waste acceptance criteria and input into the Saltstone environmental transport models. In contrast, for post-cleaning residue, the data support the Closure Modules and provide inputs to the Special Analyses. Clearly, the end uses of the data are different, and as such, the needs are different. In this task activity, existing characterization data for past sludge batches, salt batches, and post-cleaning residue, as well as the results of Performance Assessments and Special Analyses, were compiled and analyzed to identify those waste constituents which appear most impactive/important to each application. The end goal of this activity is to provide a basis for prioritizing characterization needs as a function of waste type.

Alternative Laboratory Methods with the Potential for Being Less Costly and/or Less Time Consuming

Many of the current laboratory analyses being performed depend upon conventional radiochemical separation technologies and/or historic measurement instrumentation. Such approaches are acceptable in many cases, particularly when the existing analytical approach is straightforward and efficient. However, in some cases, there is the potential for improvement, from a simplicity, cost, and/or time standpoint. Approaches to semi-automating select radiochemical separation processes were investigated. Automation would provide benefits in reducing man-power requirements, decreasing analysis times where multiple analyses are required, and in reducing personnel dose overall. Alternate approaches to some of the current radiochemical separation schemes were also investigated. From these investigations, recommendations were developed for implementation of the successes into the existing laboratory analysis program. This activity offers the greatest potential benefit by focusing on alternative characterization methods to replace costly and time consuming methods, particularly those methods applying to high impact, risk-driving constituents.

Technical Basis/Strategy for Improving Cost Effectiveness of SRNL's Tank Closure Characterization Program

SRNL's current tank closure characterization program includes laboratory analysis of a total of more than sixty radionuclides and stable waste constituents, with a nominal six month completion schedule beginning once all the samples have been received and the sample compositing protocols have been identified. The bulk of the analytical cost and schedule is dedicated to activities supporting the radionuclide analyses (as opposed to the stable constituent analyses), due to the extensive matrix preparations, radiochemical separations, and hybrid measurements utilized to achieve the high analytical sensitivities necessary for effective performance assessment modeling of the radionuclides. As such, this activity is focusing on the data needs, drivers, findings, and analysis attributes of the various radionuclides, on a nuclide-by-nuclide basis. A total of fifty-six nuclides are being addressed,[1] to cover all radionuclides requiring characterization in one or more of the most recent six SRS tank closure campaigns (Tanks 18, 19, 5, 6, 16, and 12). In each case, a series of technical and programmatic attributes feeding nuclide characterization relevancy are being assessed. Based on the results, conclusions and recommendations are being developed which will identify changes that will make SRNL's tank closure characterization activities more cost effective and timely. A path forward for implementing the changes is also being developed

Design/Assembly of System for Automating Select Radiochemical Separation and Waste Removal Processes

The majority of tank waste radionuclide analyses depend on radiochemical separations utilizing highly specific solid phase extractants. The separations associated these extractions, along with the activities required to remove the resulting extraction waste from the laboratory, pose risks to hands-on personnel, due to the associated radiological doses and the potential for material contamination. These dose issues often require that the initial phase of the separations be conducted in the SRNL Shielded Cells facility, which increases the cost of analyses and adds considerable time to the schedule. Apart from the dose issues, the separations and waste removal processes are labor intensive and time consuming, because of the high degree of extended duration hands-on tasks. In this activity, design and assembly of a flexible system for automating the key steps of the radiochemical separations and the waste removal processes are being initiated. Successful automation of these steps will enable such processes to become hands-off tasks, which will increase productivity and lower personnel risk. In specific, this activity has included the procurement and integration of the hardware, plumbing, and computer software necessary for remotely performing many of the reagent preparation, reagent addition, and waste removal steps. The automated system will introduce reagents to the resin columns as programmed, transfer the waste to a holding vessel, adjust the pH of the liquid waste, and finally discharge the waste into the radioactive drain system. The analyte of interest will remain on the columns, and will subsequently be manually transferred to a vacuum box for the final step of the separation. The automated system will have a user friendly graphical user interface, programmed in LabVIEW or an equivalent. The end goal is to program various protocols into the system, which will allow automation of a wide range of radiochemical separation methods. This initial phase of development work is limited to automating the waste handling portion of the system, which will increase productivity and reduce worker exposure considerably. The primary hardware is being designed and procured along with the associated plumbing and software. Initiation of the integration of the hardware and software has begun. Completion of the system and subsequent implementation and testing is anticipated to be performed next year.

Alternative Radiochemical Separation Protocols

Most of SRNL's existing radioanalytical methods have not yet been optimized, as they were developed over short timeframes limited by funding restrictions and the need to meet

[1] This includes H-3, C-14, Al-26, Cl-36, K-40, Ni-59, Ni-63, Co-60, Se-79, Sr-90, Y-90, Zr-93, Nb-94, Tc-99, Pd-107, Sn-126, Sb-126, Sb-126m, I-129, Cs-135, Cs-137, Ba-137m, Sm-151, Eu-152, Eu-154, Pt-193, Ra-226, Ra-228, Ac-227, Th-229, Th-230, Th-232, Pa-231, U-232, U-233, U-234, U-235, U-236, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Pu-244, Am-241, Am-242m, Am-243, Cm-243, Cm-244, Cm-245, Cm-247, Cm-248, Cf-249, and Cf-251.

aggressive reporting deadlines. This has a particular impact in cases where the existing analytical approach is highly labor intensive, highly time consuming, subject to matrix interferences, and/or incapable of meeting the targeted minimum detection limits. Examples of methods fitting into this category include the analytical approaches currently utilized for analyzing Tc-99, I-129, Ra-226, Pa-231, Th isotopes, and Am/Cm isotopes. In this activity, a focused effort is being undertaken to develop and test alternative radiochemical separation protocols holding the greatest promise for having a significant impact on the cost-effectiveness and timeliness of the program. Specifically, the near-term goals are: a) develop a single separation protocol that would allow simultaneous preparation of both Tc-99 and I-129 (as opposed to the current approach, which requires two separate Shielded Cell protocols performed individually); b) develop high yield separation protocols for Ra-236 and Pa-231, which will lower minimum detection limits and introduce the possibility of utilizing smaller sample aliquots outside of the Shielded Cells (the current protocols result in relatively low chemical yields); c) develop a new protocol for removing yttrium from the trivalent actinides, so a Y-90 decay waiting period is not required prior to removing purified Am/Cm aliquots from the Shielded Cells (the current approach typically requires a 1-2 week decay waiting period); and/or d) investigate benefits obtained from electroplating alpha spectroscopy mounts for the thorium isotope measurements and Am/Cm isotope measurements (the current approach produces low resolution, which raises minimum detection limits). In this initial phase of development work, a minimum of two improved radiochemical separation protocols are being developed and tested. Development of additional improved protocols is planned for next year.

RESULTS AND DISCUSSION

Characterization Activities Driving Cost and Schedule

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

- Tank 50 quarterly supernatant 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 elimination or 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.

Potential Streamlining of Characterization Requirements Based on the Relative Constituent Risks

Results from this study evaluating risk screening as a method of reducing waste characterization requirements are promising. Results for the emptied and cleaned waste tanks considered in this analysis consistently showed that five radionuclides contribute 99% of the maximum projected long term potential environmental dose associated with closed tanks (over a ten thousand year period into the future). Specifically, that includes Np-237 contributing a maximum projected annual dose of 88 microsieverts per year, Ra-226 contributing a maximum projected annual dose of 19 microsieverts per year, Pa-231 contributing a maximum projected annual dose of 11 microsieverts per year, C-14 contributing a maximum projected annual dose of 3.9 microsieverts per year, and I-129 contributing a maximum projected annual dose of 1.4 microsieverts per year. In general, these dose drivers were consistent with those identified through the Tank Farm PAs and CA. However, there was one significant difference – Tc-99 was identified as risk driver in the H-area PA. The reason for this discrepancy is under investigation.

In contrast, results for the four Saltstone disposal units considered in this analysis gave consistent results indicating that three primary radionuclides contributed the bulk of the maximum projected long term potential environmental dose associated with stabilized salt waste (again, over a ten thousand year time period into the future). Specifically, that includes Ra-226 contributing a maximum projected annual dose of 2.7 microsieverts per year, I-129 contributing a maximum projected annual dose of 0.40 microsieverts per year, and Np-237 contributing a maximum projected annual dose of 0.38 microsieverts. Maximum projected doses associated with all other nuclides in stabilized salt waste were at least an order of magnitude lower than that of Np-237. As in the case of the projected doses associated with emptied and cleaned tanks, the projected doses for stabilized salt waste were generally consistent with those of the Saltstone PA – however, as before, Tc-99 was not identified as a dose driver using the methodology of the screening technique, although it was identified as a dose driver in the Saltstone PA. The reason for this discrepancy is under investigation.

Determination of the Relative Usefulness of Potential Characterization Bases Other than Sampling and Analysis

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. Examples of the variation of key radionuclide concentrations in SRS sludge batches, salt feed solutions, and post-cleaning residue can be seen in Figures 1, 2, and 3, respectively. Interestingly, the usefulness 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. This is consistent with the data presented in Figures 1-3, where nuclide concentration trends over time were relatively predictable for sludge batches and salt batches, but significantly more erratic for post-cleaning residue.

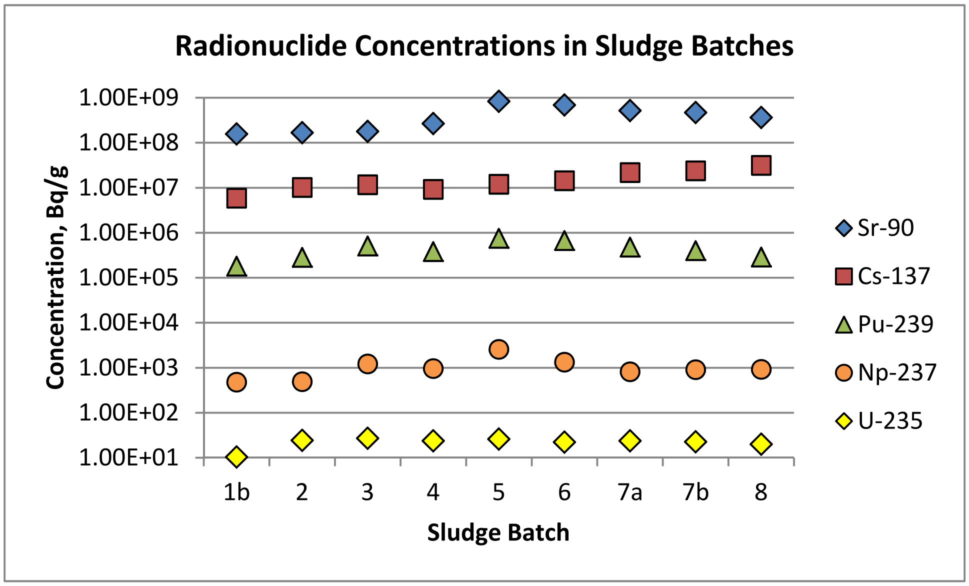

Figure 1. Concentrations of Select Radionuclides in SRS Sludge Batches 1B through 8

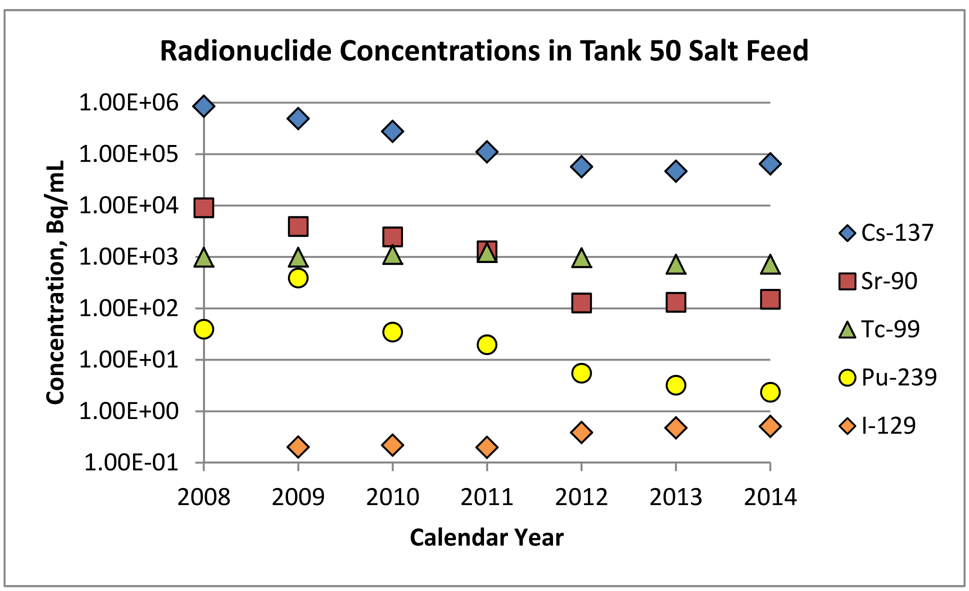

Figure 2. Concentrations of Select Radionuclides in SRS Tank 50 Salt Feed

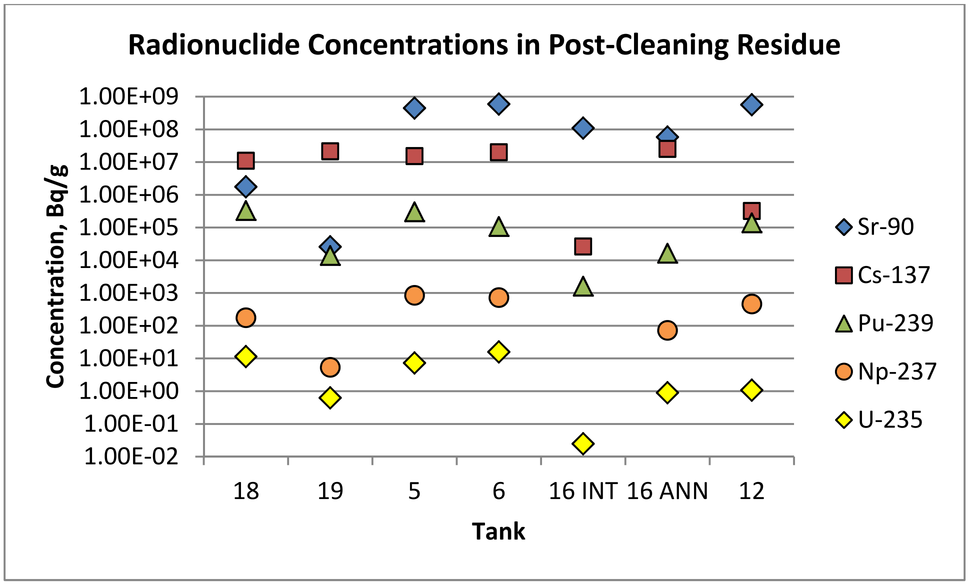

Figure 3. Concentrations of Select Radionuclides in SRS Post-Cleaning Residue

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 select wastes
  - the concentration ranges of key radioisotopes and metals in select wastes
- 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.

Honing of Characterization Needs as a Function of Waste Type

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 (only five constituents have ever exceeded 10% of the WAC limits, as shown in Figure 4) and are predictable based on historic trends, process knowledge, and qualification testing

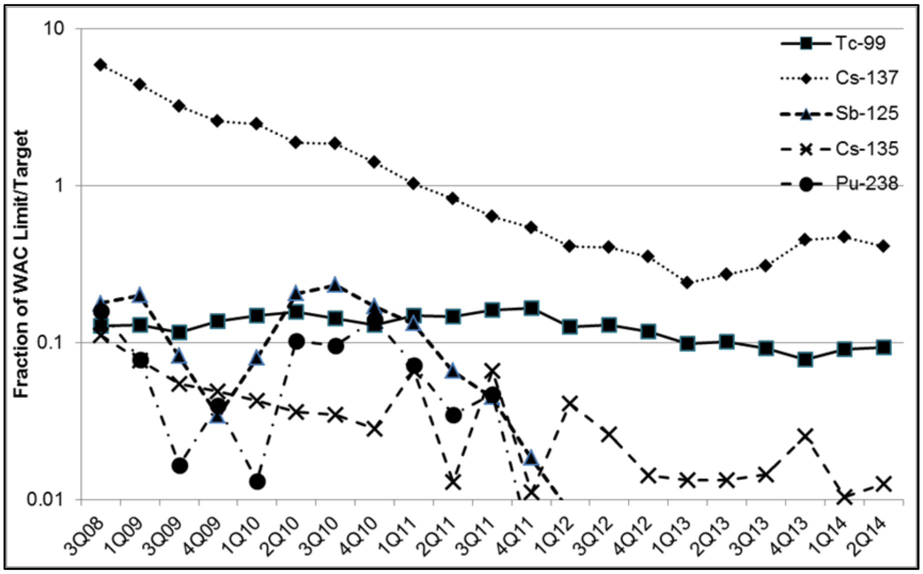

Figure 4. Radionuclides Exceeding 10% of the 2014 Saltstone WAC Limits/Targets

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

Alternative Laboratory Methods with the Potential for Being Less Costly and/or Less Time Consuming

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.

Technical Basis/Strategy for Improving Cost Effectiveness of SRNL's Tank Closure Characterization Program

Literature reviews and SRS Tank Closure data compilations are currently in progress to provide nuclide-specific bases for determining: 1) radioactivity dominance over time; 2) relative analytical cost and time requirements; 3) nuclide detectability; 4) anticipated long term environmental impacts; 5) potential application of alternative characterization approaches; and 6) primary long-term sources (decay products versus parent nuclides). Tables and plots of the compiled data are being generated and then analyzed to support conclusions identifying programmatic changes making SRNL's Tank Closure characterization program more cost effective and timely. Estimates of the expected savings are also being identified.

Design/Assembly of System for Automating Select Radiochemical Separation and Waste Removal Processes

This initial phase of design/assembly work focusses on automation of the waste removal processes associated with extraction chromatography-based radiochemical separation methods. By automating the waste removal processes, substantial time savings in executing various radiochemical protocols are expected to be achieved. More importantly, this system will minimize handling of these highly radioactive solutions, which will result in significantly less dose as well as less contamination risk to personnel.

Thus far, the following scope has been completed: 1) the software controlling the system has been developed and demonstrated; 2) worker feedback on the software operation has been received and utilized to improve system workflow and simplify the user interface; 3) the system configuration has been designed to facilitate waste disposal requirements and to fit within the radiological hoods at SRNL; and 4) hardware and plumbing has been designed, and fabrication/procurement is in progress. A prototype of the hardware can be seen in Figure 5. Assembly and testing is occurring next, with the goal of making the automated waste removal system ready for implementation in CY16. Expanding the capabilities of the system for automation of select radiochemical separation processes is planned for CY16, assuming continued funding is available.

Figure 5. Prototype of Hardware for Automating Waste Removal

Alternative Radiochemical Separation Protocols

Development of an alternative separation/measurement protocol for analyzing Ra-226 is currently in progress. The alternative Ra-226 protocol will likely utilize one or more high selectivity radium extractants coupled with a reduced background Compton suppressed gamma spectrometry measurement instrument. Current protocols allow for decontamination factors for a number of radionuclides in the neighborhood of eight orders of magnitude, but less so for others. With improved selectivity, minimum detection limits by gamma spectrometry are expected to be reduced further and alpha spectrometry may be employed to drive minimum detection limits down even lower. An objective of the new protocol is to increase radium yield and/or measurement sensitivity sufficiently, such that the sample aliquot size can be reduced, thereby eliminating the need to perform the initial labor-intensive separation steps in the Shielded Cells. (Note that the current Ra-226 protocol results in relatively low chemical yields and relatively high minimum detection limits). Several alternative extraction agents are being investigated, including an HDEHP impregnated resin, a Superlig 640 impregnated filter membrane, and MnO_2 and monosodium titanate based getters.

Alternative protocols for analysis of Am/Cm and Th isotopes will be pursued next, utilizing the following approaches: 1) separation/removal of yttrium from trivalent actinides, to eliminate Y-90 decay waiting periods (for dose reduction purposes) prior to removing purified Am/Cm fractions from the Shielded Cells; and 2) investigation of the benefits of electroplating alpha spectroscopy mounts for Th isotope measurements (to increase energy resolution, with the goal of reducing minimum detection limits).

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) The most costly nuclides to analyze include Cl-36, Ni-59/63, Tc-99, I-129, Ra-226, Th-229/230, Pa-231, and the Am/Cm isotopes. The longest duration analyses include those for Ra-226, Pa-231, and the Am/Cm isotopes.

3) 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

4) The total projected dose risks are driven by a relatively small number of nuclides

- Tank Farm: Tc-99, Ra-226, Pa-231, Np-237 (and applicable parent nuclides)
- Saltstone: Tc-99, I-129, Ra-226, Np-237 (and applicable parent nuclides)
- Only five Saltstone nuclides have ever exceeded 10% of WAC limits (Tc-99, Sb-125, Cs-135, Cs-137, and Pu-238)

5) Characterization uncertainties

- Relatively small for well-executed sampling and analysis (typically ±20%)
- Larger based on receipt histories, process knowledge, scaling factors, historic trends
  - o Dependent on heterogeneity, but often 2-3X to an order of magnitude
  - o May be acceptable for “low risk” constituents
  - o With conservatism, has been used effectively for safety and planning purposes

6) 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

7) 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)

8) 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

9) 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, the existing program seems excessive

10) Best near-term focus areas for improvement of laboratory characterization methods include:

- Automation of select radiochemistry separation protocols and waste removal processes
- Development of more effective/efficient separation techniques for high resource demand analytes

11) Applicability to other DOE sites

- Provides a baseline for PUREX and HM tank waste characterization

REFERENCE

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