Investigations for Improved Tank Characterization and Cleaning – 15454

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ABSTRACT

In the process of tank closure, significant waste characterization and chemical cleaning occur. Costly characterization is performed at numerous processing stages. A need exists for new strategies and technologies to understand, optimize, scale, and speed up tank waste characterization with the end goal of reducing costs through implementation of programmatic changes that accelerate tank waste processing and tank closure schedules. This paper will explore a possible alternative to waste characterization methodologies and cleaning technologies to reduce costs and improve processing efficiency.

INTRODUCTION

Approximately 320 million liters of radioactive waste is being safely stored across the DOE Hanford, Savannah River, and Idaho sites. The material, widely varying in chemistry and form, is stored in 226 tanks. It has long been recognized that processing this radioactive tank waste and ultimately closing the tanks is one of the most technologically complicated efforts in the Department. While the details of processing the waste may vary, the process is comprised of common basic steps: retrieve the waste, separate waste into low- and high- radioactivity waste streams, close tanks according to compliance agreements. A significant amount of characterization and cleaning is involved in these steps leading up to the closure of a tank, which equates to significant cost and time requirements. With the remaining tank closure duration estimated at more than 40 years, improvements in characterization and cleaning could result in substantial reductions in the overall cost and schedule for the closure effort.

Chemical cleaning may include a series of processes. The chemical cleaning process used on Tank 12 at the Savannah River Site (SRS) included a cleaning strategy of low temperature aluminum dissolution, washing, bulk oxalic acid cleaning, and neutralization. While these cleaning steps provide an effective and thorough cleaning of the tank, the subsequent byproducts may be undesirable in other processing steps. For example, insoluble oxalate salts accumulate within the SRS Tank Farm and waste processing facilities from bulk oxalic acid cleaning. A need exists to evaluate alternative chemical cleaning techniques and methods to identify best cleaning practices for the overall processing facility.

Two efforts underway to improve the overall process for tank closure are: 1) a modeling approach evaluating effective characterization based on the dose risk to a member of the public (MOP) and 2) an experimental approach evaluating alternative chemical cleaning methods for high level waste tanks.

COST EFFECTIVE CHARACTERIZATION

In an effort to develop a more cost effective approach to waste characterization, a risk based approach has been considered that would focus waste characterization on those radionuclides that pose a potential risk for significant dose to an MOP. Refining the screening process already performed prior to Performance Assessment (PA), Composite Analysis (CA), and Special Analysis (SA) could potentially lead to a reduction in the number of radionuclides that need to be evaluated. Based on current ICRP data, 1252 radionuclides have been identified that pose some risk for internal and/or external dose to an MOP. Typically, the number of radionuclides considered for an analysis is reduced by conservatively assuming a large inventory for each radionuclide and performing simple transport and decay calculations from the source to receptor location and eliminating those radionuclides that do not yield a significant dose from

further analysis. This process typically reduces the number of radionuclides that must be consideration to less than 100 species.

To improve the screening process, a more realistic calculation would be performed, while still remaining conservative. The associated costs and time requirements needed to perform the risk analysis would be reduced by limiting waste characterization to those species that pose significant risk to a MOP. The GoldSim™ (Version 10.50 SP 3) software, which has been used to model radionuclide transport and perform dose calculations for several applications at SRS, was utilized to develop the risk screening model. GoldSim™ is a convenient modeling tool for simple applications because it offers the capability of modeling 1-dimensional radionuclide transport and decay and is also capable of performing probabilistic simulations for uncertainty analysis.

As illustrated in Figure 1, features taken into account in the radionuclide transport model include a closure cap placed over the waste disposal unit, transport of material from the waste source through the vadose zone to the aquifer, and transport in the aquifer to the receptor source. For cleaned SRS waste tanks following closure, residual contamination is assumed to be concentrated in the last model cell representing a 3.78 liter thick layer at the bottom of the tank. Other cells in the waste zone are assumed to be clean grout, see Figure 2.

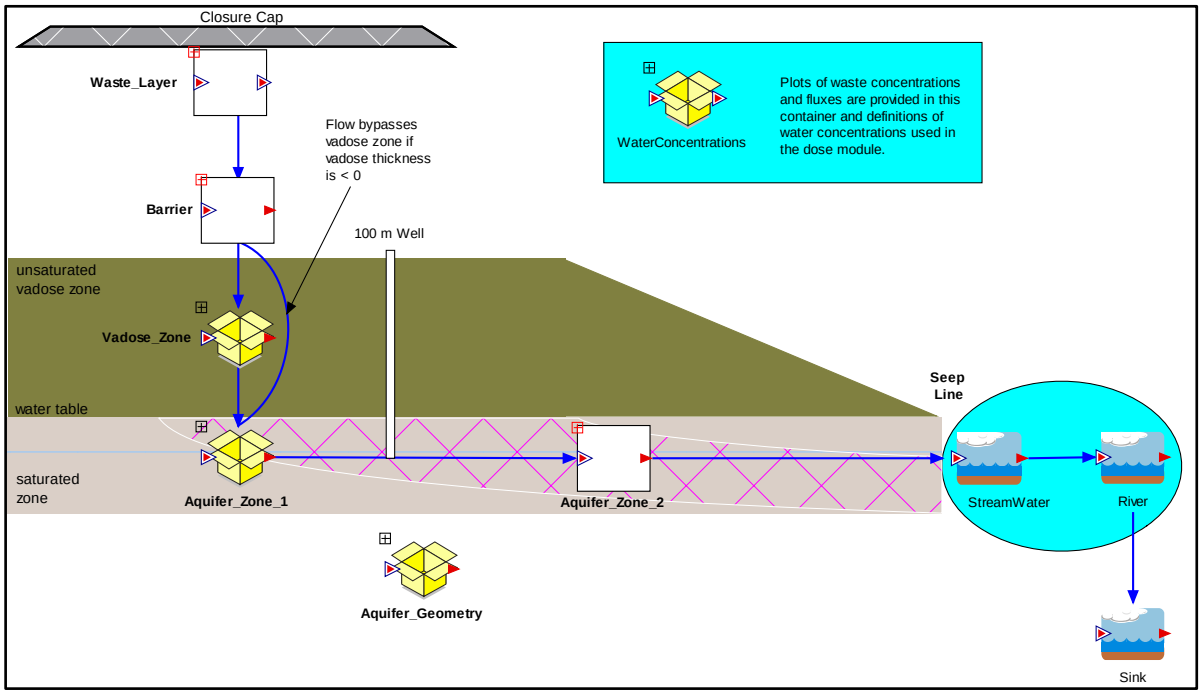

Fig. 1. GoldSim model of radionuclide transport in waste characterization model.

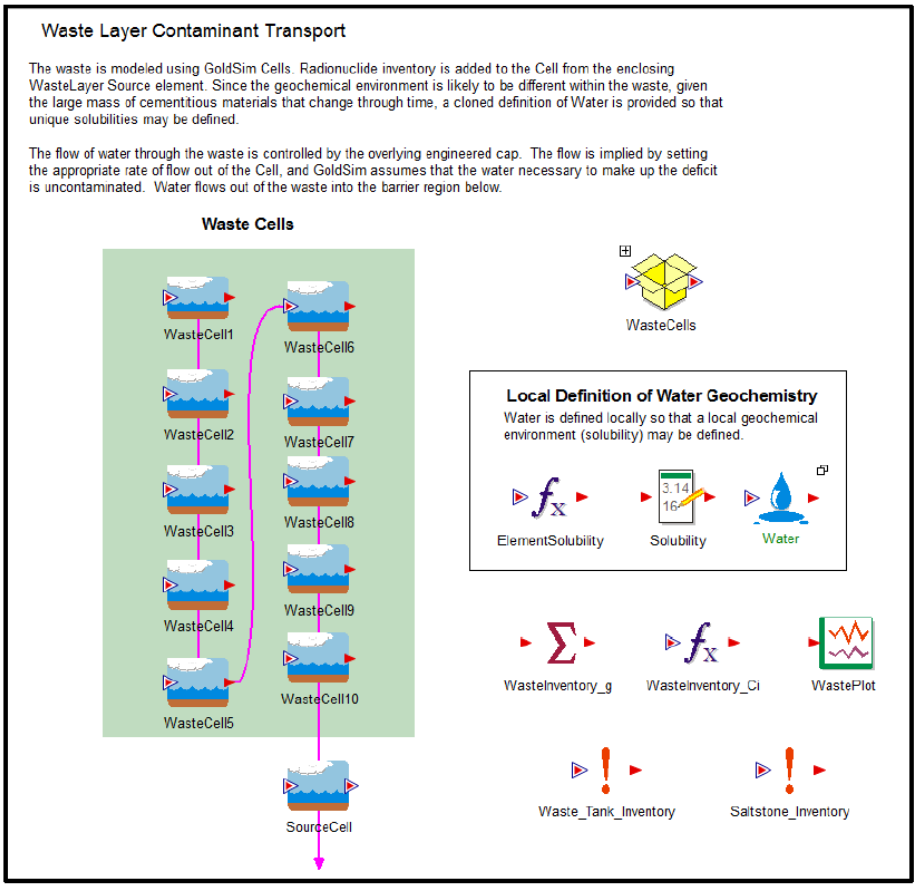

Fig. 2. GoldSim model of waste disposal facility.

The aquifer model is divided into two separate zones, which are further subdivided into regions as shown in Figure 3. To model different waste disposal sites, the length of the zones are variable, defined through inputs to the model, however the number of cells is fixed. The cells of Zone 1 contain only sandy soil. The first group of cells in Zone 2 can represent sandy soil, clayey soil, or a mixture of the two. The dose calculations reported in this paper are based on the concentration of radionuclides in the groundwater at the 100 meter well location.

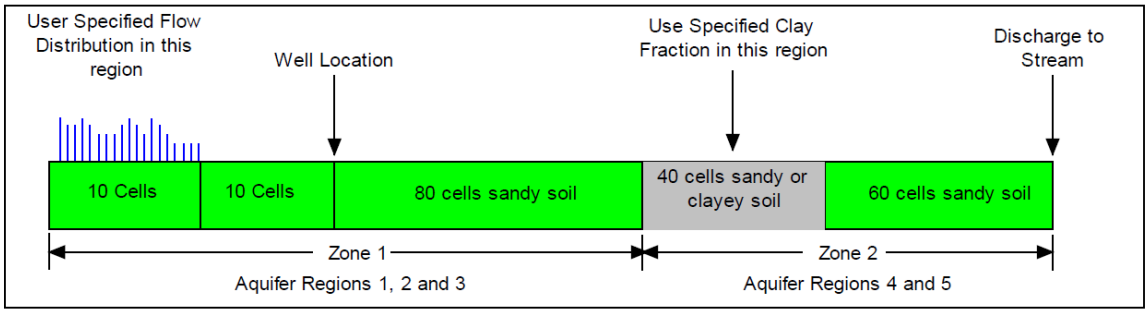

Fig. 3. Schematic representation of aquifer transport model.

To estimate the source inventory to model, inventories assumed in the Tank Farm PAs (SRS-REG-2007-00002, SRR-CWDA-2010-00128) and the SRS CA (SRNL-STI-2008-00380) were compared to the residual inventory estimated from characterization studies. In general, inventories assumed in the

performance analyses were very conservative. To evaluate the risk to a MOP more realistically, results from the waste characterizations were used to estimate tank inventories.

The scenario used to evaluate dose to a MOP is that institutional control over the disposal site is lost 100 years after site closure. At this time a resident farmer occupies the site and uses groundwater from a well located 100 m from the waste disposal area (SRS Tank Farm) for personal use and agriculture. For these scoping calculations, it was assumed that the residual inventory was placed in the waste in 2010 and a geo-synthetic cap was placed over the grouted tanks in 2025. After 500 years of placement, the cap was assumed to fail. The vadose zone thickness and distance from the tank to the 100 meter well were obtained from the Tank Farm PAs. The aquifer pore velocity was obtained from the CA. The GoldSim™ model was run for a 10,000 year evaluation period to determine the all-pathways dose at the 100 meter well for 62 radionuclides identified through previous screening.

ALTERNATIVE CHEMICAL CLEANING METHODS

Two chemical cleaning technologies that have been implemented at SRS are Low Temperature Aluminum Dissolution (LTAD) and Bulk Oxalic Acid Cleaning (BOAC). The chemical cleaning effort was successful in Tank 12 with regard to sludge heel and beta/gamma radionuclide removal. The cleaning strategy employed followed the processing sequence of LTAD, washing, BOAC, and neutralization. While effective, a disposition path has not been identified for the oxalate added during BOAC, and insoluble oxalate salts are accumulating within the SRS Tank Farm and waste processing facilities such as evaporators. Extensive washing is required to remove the moderately soluble sodium oxalate salts from the sludge prior to vitrification at the Defense Waste Processing Facility (DWPF). As a result, it is desirable to minimize oxalate addition to the tank farm, and improved strategies and technologies are needed to chemically clean radioactive High Level Waste (HLW) tanks prior to tank closure.

Primary activities underway to evaluate improvements to chemical cleaning technologies and strategies include non-radiological simulant testing, radioactive simulant testing, corrosion evaluations, and real waste testing. Chemical cleaning options other than BOAC are under consideration, including oxidation of actinides with permanganate in either strong caustic or dilute acid solutions. Utilization of permanganate-based methods lead to the addition of manganese oxide solids in the waste, therefore minimization of permanganate additions is needed. Solubility of primary and secondary sludge phases characteristic of those found in SRS and Hanford tanks will be evaluated using individual metal phase reagents or waste heel simulants. The potential cleaning agents under evaluation include oxalic acid, oxalic acid blended with supplementary acids, permanganate solutions (both acidic and caustic).

In addition to the evaluation of the cleaning agents, testing is also underway to develop an optimized chemical cleaning flow sheet. The experimental matrix is based upon information reported from the SRS tank cleaning campaigns and focuses on minimizing cleaning agent additions, optimizing tank heel washing and acidification steps, and identifying phases not suitable for retrieval with current methods.

RESULTS AND DISCUSSION

Some results from the GoldSim™ 10,000 year evaluation for SRS Tanks 5, 18 and 19 are shown in Figures 4-6. The total all-pathways dose is plotted for each tank along with the 14 individual radionuclides yielding the highest doses. The maximum doses to a MOP vary by tank. Figure 4 shows a maximum dose of about 1.0E-4 mSv/yr from Tank 5, with a significant peak of Np-237 around 3000 years into the calculation. Figure 5 suggests that the maximum dose from Tank 18 is approximately 0.1 mSv/yr at about 1,500 years. Figure 6 for Tank 19 shows two significant peaks, one at slightly less than 2000 years from Ra-226 and one at slightly less than 5000 years from C-14. The maximum overall total

dose is predicted to be about 4.0E-3 mSv/year. While the results cover a period of 10,000 years, some of the radionuclides identified had concentrations that are continuing to increase at the 10,000 year mark, suggesting that the highest concentration levels have yet to be evaluated. Examples include Ra-226 for Tanks 18 and 19.

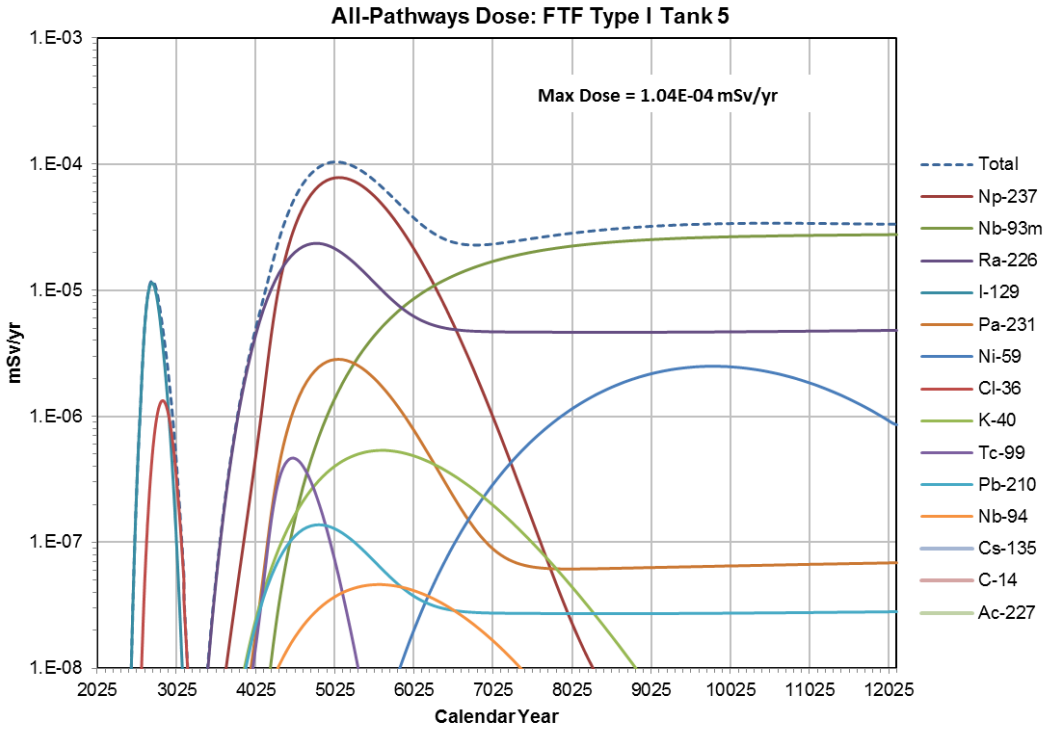

Fig. 4. All-Pathways dose from Tank 5-F based on characterization.

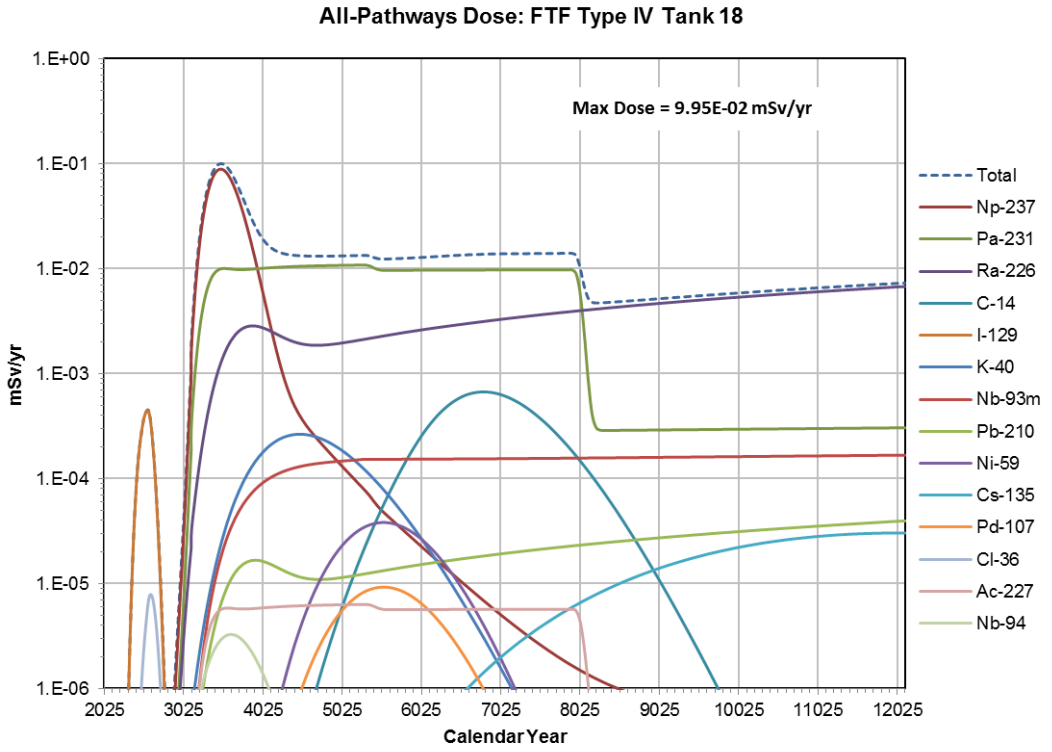

Fig. 5. All-Pathways dose from Tank 18-F based on characterization.

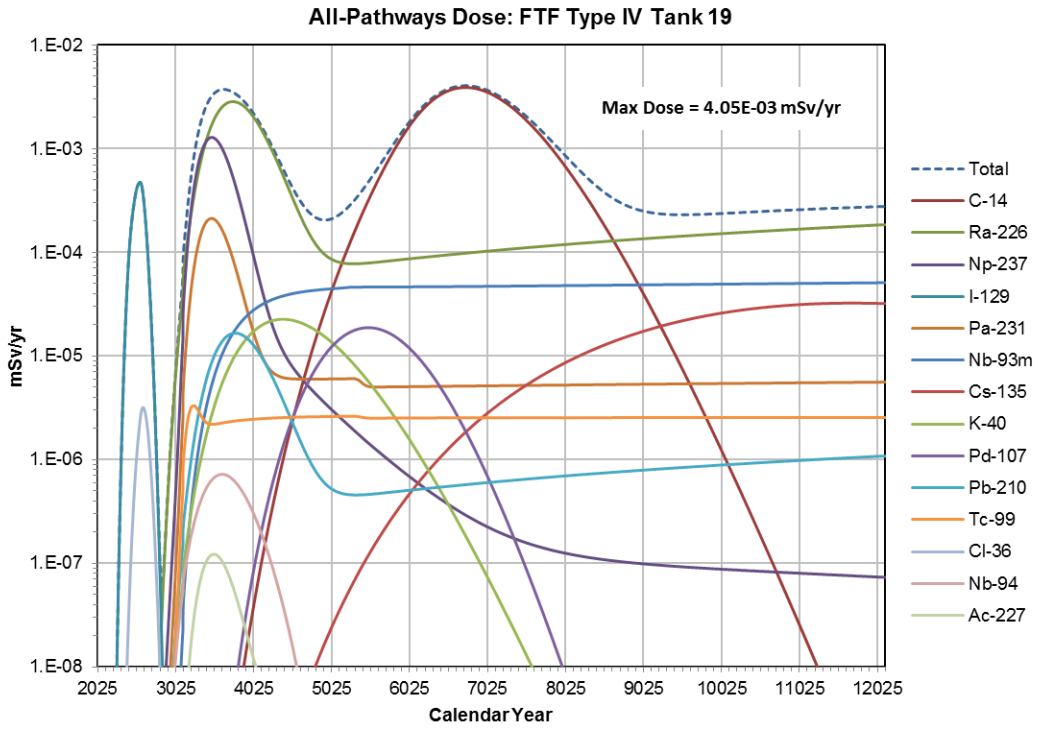

Fig. 6. All-Pathways dose from Tank 19-F based on characterization.

CONCLUSION

As the results in Figures 4-6 show, for each tank analyzed a relatively small number of radionuclides account for greater than 1% of the total dose and, for all three tanks evaluated, only 10 radionuclides gave doses greater than 1.0E-04 mSv/yr. A true evaluation of dose risk would require estimating dose probability from the uncertainty in factors such as residual inventory, human behavior, and soil properties. A full risk assessment was not made in this preliminary study. However, the consistency of the response from individual radionuclides as well as the overall maximum dose calculated between the tanks suggests that applying risk screening is a promising approach to reducing waste characterization requirements. In contrast to the PA results, the characterization analysis did not predict a significant dose from Tc-99. Three factors contributed to this result: 1) The PA used conservative estimates of residual radionuclides whereas the characterization study evaluated measured amounts; 2) The PA considered the dose from the entire tank farm whereas the characterization study considered individual tanks; 3) The characterization study tested a revised dose model that has not been fully evaluated.

When considering chemical cleaning methods, the effectiveness of the cleaning technique as well as the impact on downstream processes must be taken into account. Experiments are in progress to minimize the use of cleaning agent, optimize tank heel washing and acidification, and identify phases not suitable for retrieval with current methods.

REFERENCES

1. "Performance Assessment for the F-Tank Farm at the Savannah River Site", SRS-REG-2007-00002, Revision 0, Washington Savannah River Company (2008).
2. "Performance Assessment for the H-Area Tank Farm at the Savannah River Site", SRR-CWDA-2010-00128, Revision 1, Savannah River Remediation (2012).
3. Hiergesell, R.A., M.A. Phifer, J.R. Cook, K. Young, M. Birk, and B. Dean, "Inventory of Projected Residual Radioactivity at the Savannah River Site End State", SRNL-STI-2008-00380, Revision 0, Savannah River National Laboratory (2008).