Performance Assessment Inventory Considerations at Savannah River Site - 11322

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ABSTRACT

To support the closure documentation process, the Savannah River Site (SRS) has developed Performance Assessments (PA) for F-Tank Farm (FTF) and is presently developing Revision 0 of the PA for H-Tank Farm (HTF). The residual material inventory within the waste tanks and ancillary equipment is the source used for the transport modeling performed as a part of the PA. Therefore estimating the residual inventories is an important factor in the overall modeling process to determine groundwater concentrations and doses. Since these inventory projections for the PAs are forward looking, i.e. the waste has not yet been removed from the tanks, estimating the inventory of residual material can present unique issues. The projected inventories remaining at closure in the various waste tanks and ancillary equipment is a function of both the volume at closure and concentrations for both specific radionuclides and non-radionuclides.

Since there are a limited number of sample analyses currently available that are directly applicable to the residual inventories remaining in the tank after cleaning, other methods of estimating concentrations were required to be used in order to determine some radionuclide specific concentrations at closure. These methods include fission yield data, equilibrium relationships, analytical capabilities and limitations, and process knowledge. Estimates are particularly challenging due to the various types of waste present in the tank farms. There were different types of materials and separation processes that led to the various waste forms; liquid, salt, and sludge.

INTRODUCTION

The SRS is a U.S. Department of Energy (DOE) facility located in south-central South Carolina, approximately 161 kilometers (100 miles) from the Atlantic Coast. The major physical feature at SRS is the Savannah River, approximately 32 kilometers (20 miles) of which serves as the southwestern boundary of the site and the South Carolina-Georgia border. The SRS includes portions of Aiken, Barnwell, and Allendale Counties in South Carolina. The SRS occupies an almost circular area of approximately 803 square kilometers (310 square miles) and contains production, service, and research and development areas.

The F-Area and H-Area are in the north-central portion of the SRS and occupies approximately 1.5 square kilometers (364 acres) and 1.6 square kilometers (395 acres) respectively. The F-Area and H-Area Tank Farms (FTF, HTF) are active liquid waste storage facilities operated by Savannah River Remediation, LLC (SRR), the Liquid Waste Operations contractor. The FTF consists of 22 carbon steel waste tanks. The HTF consists of 29 carbon steel waste tanks in varying degrees of service or liquid waste removal operations. The FTF and HTF carbon steel waste tanks store (or once stored) liquid radioactive waste generated primarily from chemical separations processes associated with production of nuclear materials from defense reactors.

Tank 17 and Tank 20 in the FTF have already been filled with grout and closed via a South Carolina reviewed and approved Closure Plan and Closure Modules [1, 2].

The PAs are used to assess the long-term fate and transport of residual contamination in the environment and provide the DOE with reasonable assurance that the removal from service of the SRS tank farm underground radioactive waste tanks and ancillary equipment will meet defined performance objectives for the protection of human health and the environment into the future.

These PAs are prepared to support the eventual closure of the FTF and HTF underground radioactive waste tanks and ancillary equipment The PA provides the technical basis and results to be used in subsequent documents to demonstrate compliance with the pertinent requirements identified for final closure of FTF including those in DOE Order 435.1, the *Ronald W. Reagan National Defense Authorization Act (NDAA) for Fiscal Year 2005* Section 3116, and South Carolina Department of Health and Environmental Control (SCDHEC) industrial wastewater regulations.

One important matter when developing these inventory estimates was that they are forward looking estimates. The PAs are developed prior to closing the waste tanks. Given this uncertainty it is necessary to provide conservatism to ensure the appropriate risks are identified.

It is important to note as part of the closure process and following the completion of material removal from the waste tanks, the residual material will be sampled and analyzed to determine the tanks' final residual inventory.

CONSTITUENT SELECTION

First, the set of candidate contaminants requiring assessment had to be determined and then a list of potential contaminants was developed based on specific screening criteria. The inventory projections for the contaminants identified through the multiple step screening process were then developed using a methodical approach to construct the inventory of each contaminant for each tank. The screening methodology addressed both radiological and non-radiological constituents.

For radiological constituents, a screening was performed to determine which needed to be modeled and therefore estimated. An initial radionuclide screening process, developed to support characterization efforts applicable for PA modeling, evaluated over 800 isotopes. Of the original isotopes, approximately 700 were excluded from further consideration using the following information:

- Physical properties of each radioisotope (e.g., half-life and decay mechanism)
- Source and handling of the waste was used based on isotope production mechanisms and time since the isotope was produced
- Screening factors for ground disposal of radionuclides developed in NRC-123, which convert a quantity of each radionuclide to a dose
- Radioisotopes with less than 5 year half life

Many of the remaining isotopes from the initial screening were not created in SRS reactors and therefore further evaluation determined which isotopes could be screened from the inventory estimates. Also, differences between the types of material treated in each area were considered. This meant a slightly different list of isotopes for each area. The remaining isotopes were included in the inventory estimate process (see Table I).

Table I. Radionuclides of Concern for the FTF and HTF PAs

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

The list of non-radiological constituents that were included in the PA modeling was derived from the *State Primary Drinking Water Regulations* (SCDHEC R.61-58) for inorganic contaminants. Constituents were screened based on the existence in the waste tanks. The resulting list was compared to the list of inorganic characteristic hazards specified in 40 CFR 261 and constituents added. Constituents were also included due to the process knowledge that they were potentially present in the waste (see Table II).

Table II. Non-Radiological Inventory of Concern for the FTF and HTF PAs

Ag	F	NO_3
As	Fe	Pb
Ba	Hg	Sb
Cd	Mn	Se
Cr	Ni	U
Cu	NO_2	Zn

ESTIMATE METHOD

The next step in the process was to estimate the residual tank inventories. The estimate of residual inventory was determined by applying a radionuclide or chemical concentration to the assumed residual volume per tank. Then adjustments were made as appropriate on the inventory estimates. Radiological inventories (activity) and non-radiological inventories (mass) are important to the PA analyses, and not the specific estimated volume or concentration of the residual solids. The specific details of the estimate methodology are documented [3, 4].

Note, at closure each tank's residual inventory will be determined through measurements. These determined inventories, not volumes or concentrations, will be compared to the estimates generated by the process described in this paper. The specific volume and concentrations after cleaning do not have individual target values, since only final inventory (activity or mass) was modeled. The determined inventory will be compared to the calculated potential dose and risk impacts.

Concentrations

The concentration of each constituent was estimated based on a variety of factors listed below in order of priority.

- Sample analysis data
- Tracking system based on fission yield calculations
- Physical relationships (e.g. equilibriums)
- Detection limits

Considering the number of waste tanks contained within FTF and HTF and the fact that most of the waste tanks are still being used to store and process their contents, determining the concentration of the final closure residual material is uncertain. The first assumption that was made was that the residual material will resemble the current solid content within the tanks and tank farms. It is believed that this is a conservative assumption. This presumes that any future processes will not concentrate any constituent. For these reasons, waste tank solids sample results are the preferred estimate method. Comparison of the concentrations of limited sampling prior to and following recent chemical cleaning show no appreciable concentrating of material [5, 6]

A majority of the radionuclide concentrations and the entire inventory non-radiological constituents (chemicals) in the residual material are tracked in the Waste Characterization System, an electronic information system that tracks waste tank data, including projected radiological and non-radiological inventories, based on liquid and solid sample analyses, process histories, composition studies, and theoretical relationships. The system (initially developed in 1995) tracks the dry sludge concentrations of radionuclides and non-radiological waste compounds in each of the SRS waste tanks.

Physical relationships were used as the third method of estimating concentrations. For radiological constituents, equilibrium relationships were used. For those radiological constituents in secular equilibrium, our PA transport modeling does not include these constituents. Since the time frames used are on the order of tens to thousands of years, isotopes with relatively small half lives are not included in the transport models although they are accounted for in the dose calculations by using their parent concentrations).

For those isotopes involved with transient equilibrium, estimating their concentrations can be more complicated. The parent and source of the parent influences the basis for the estimate. For example, uranium daughters can be difficult to estimate. Following uranium through the complete process from mining to disposal will illustrate this difficultly. Uranium daughters can be considered at equilibrium at the time of mining. Once the processing of the uranium in the fuel preparation steps, the daughters can be assumed to be removed. At this point the in-growth of daughters begins. The fuel then proceeds through the reactors to the chemical separation processes. At this point the age of the waste would allow an estimate of the amount of daughter in-growth. Although, in the chemical separation process, the relationship between parent and daughters can be broken as certain elements are recovered and others discharged to the waste tanks.

Adjustments

Not only will a majority of the material in each tank be removed prior to tank closure, the material in one tank could be transferred through other tanks as part of the closure process. The

complete duration of the closure process is expected to take approximately 20 years and the current planned sequences are subject to changes. To address this risk, a conservatism was built into the inventory estimates. This conservatism was accomplished by grouping the tanks with other tanks containing similar materials and with similar histories and/or futures. Once grouped, the maximum concentration for each constituent within that grouping for any one tank was applied to all the tanks within the group.

The tank type generally had an effect on the type of waste received and therefore guided the group selection. In general, each waste tank was built at approximately the same time as others of the same type. In addition, the tanks generally contain largely one of two waste types, metal hydroxides (commonly referred to as sludge) and a sodium nitrate/nitrite salt (commonly referred to as salt). For the tanks categorized as either salt or sludge, the predominant use was considered. This was established as the use (past and future) for the majority of the tank's life.

To allow for more efficient and cost effective means of confirming concentrations within residual materials for radionuclides with a limited potential impact to dose, inventories were increased. At a minimum, any constituent's inventory, on a tank by tank basis, less than the analytical detection was adjusted to the minimum detection limit.

Also, for a majority of the radionuclides with an adjusted inventory less than 1 Ci, their inventories were adjusted to 1 Ci. The choice of 1 Ci was based on selecting a value of an easily identifiable and measurable inventory. These constituents not adjusted have been observed (through previous analyses or scoping studies) to have greater potential impact on the overall dose.

Note, those radionuclides with adjusted inventories greater than 1 Ci were not adjusted in this step. This adjustment only applied to the radionuclide inventories and not the chemical inventories.

Another adjustment to the residual material inventories was based on recent experience with the cleaning processes and its effect on specific constituent's concentrations. Sample results before and after the cleaning process within one tank showed significant decreases in concentrations for several elements (see Table III) [3, 4]. Based on this observation, the inventories of these elements (both radiological and non-radiological) were reduced by one order of magnitude to reflect the cleaning efficiency expected by chemical cleaning these tanks.

Table III. Consitutents that showed decreased concentrations during chemical cleaning

	2007 (avg) Prior to cleaning	2009 (avg) following cleaning
Cs-137 (mCi/kg)	1.09E+03	4.2E+01
Sr-90 (mCi/kg)	3.70E+04	5.6E+03
Tc-99 (mCi/kg)	<1.1E-02	<2E-03
U (mg/kg)	9.9E+04	<1.6E+03

ESTIMATE UNCERTAINTY

The uncertainty used with the inventory estimates are based on amount of conservatism in the estimate. For the inventory estimates based on sample analyses, in general less uncertainty was used.

The process used to estimate the waste tank residual material at operational closure created differing levels of conservatism with the estimates; bounding and reasonable. Estimates were developed for all chemicals and radionuclides expected to occur in HTF, but those components expected to affect dose are closely scrutinized, and the values selected are intended to provide conservatism over what is expected to remain at operational closure.

FEEDBACK

Knowledge has been gained on the effect of the relative inventory levels of the residual components through the development of these various PAs. This knowledge can be used to identify which inventory projections have negligible impact on dose and allow allocation of analytical resources in a risk informed manner for future tank closures. This experience has also been used to better understand the uncertainty and variability inherent in the inventory projections.

The equilibrium estimating method is an example of where care needs to be taken with respect to conservatism. In the FTF PA Rev. 0, the Ra-226 waste tank residual inventory was assumed to be in equilibrium with the U-234 grandparent [7]. It was thought, and the latest sample results indicate, this assumption was significantly conservative [8, 9]. Subsequent modeling showed a dose due to the amount of Ra-226 present in the waste tank residual material at closure.

As material is removed from waste tanks and tanks are closed, samples will be collected and analyzed. These analyses will allow comparisons to the estimates and will be applied to future estimates.

CONCLUSION

Estimating the residual inventories is an important factor in the overall modeling process to determine groundwater concentrations and doses. Since these inventory projections for the PAs are forward looking, i.e. the waste has not yet been removed from the tanks, estimating the inventory of residual material can present unique issues. The projected inventories remaining at closure in the various waste tanks and ancillary equipment is a function of both the volume at closure and concentrations for both specific radionuclides and non-radionuclides.

Since there are a limited number of sample analyses currently available that are directly applicable to the residual inventories remaining in the tank after cleaning, other methods were used to estimate constituent concentrations at closure.

REFERENCES

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