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Technetium Inventory, Distribution, and Speciation in Hanford Tanks

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Executive Summary

This report was revised in September 2014 to add detail and correct inaccuracies in Section 5.0 on the fate of technetium (Tc) recycle from the off-gas systems downstream of the low-activity waste (LAW) melters back to the melters, based on several reports that were not found in the original literature search on the topic. The newly provided reports, from experts active in the Hanford Tank Waste Treatment and Immobilization Plant (WTP) glass studies, the Vitreous State Laboratory at The Catholic University of America (VSL) melter and off-gas system demonstrations, and overall WTP systems analysis, were not originally found on electronic databases commonly searched. The major revisions to Section 5.0 also required changes to Section 7.0 (Summary and Conclusions) and this executive summary.

The purpose of this report is threefold: 1) assemble the available information regarding Tc inventory, distribution between phases, and speciation in Hanford's 177 storage tanks into a single, detailed, comprehensive assessment; 2) discuss the fate (distribution/speciation) of Tc once retrieved from the storage tanks and processed into final waste forms; and 3) discuss/document in less detail the available data on the inventory of Tc in other "pools" such as the vadose zone below inactive cribs and trenches, below single-shell tanks (SSTs) that have leaked, and in the groundwater below the Hanford Site.

A thorough understanding of the inventory for mobile contaminants is key to any performance or risk assessment for Hanford Site facilities because potential groundwater and river contamination levels are proportional to the amount of contaminants disposed at the Hanford Site. Because the majority of the total ^{99}Tc produced at Hanford (~$3.26 \times 10^4 \times 10^4$ Ci) is currently stored in Hanford's 177 tanks (~2.65×10^4 Ci), there is a need to understand the fate of this ^{99}Tc as it is removed from the tanks and processed into final solid waste forms. Current flowsheets for the WTP process show most of the ^{99}Tc will be immobilized as LAW form(s), with glass being the current baseline LAW waste form. The LAW that cannot be accommodated in the current LAW Facility may be vitrified in a second LAW vitrification facility that would be built, or a supplemental solidification process may be chosen. A third alternative being investigated is to increase the throughput of the WTP LAW Facility such that it can process the entire LAW inventory. Whatever approach is taken, the current baseline is that solid LAW waste forms will remain on the Hanford Site and be disposed at the Integrated Disposal Facility (IDF). Only a small fraction of Hanford's ^{99}Tc will be shipped to a geologic repository within the immobilized high-level waste. Past performance assessment (PA) studies, which focused on groundwater protection, have shown that ^{99}Tc within various waste forms disposed in the IDF would be the primary dose contributor to the IDF performance. It also should be noted—based on the 2001 IDF PA (Mann et al. 2001), the Supplemental ILAW risk assessment (Mann et al. 2003), and the Tank Closure and Waste Management Environmental Impact Statement (TC&WM EIS; DOE 2012)—that the ^{99}Tc inventory not immobilized in glass (i.e., off-gas secondary wastes that currently would be solidified in a low-temperature waste form such as Cast Stone) was the only ^{99}Tc-bearing waste fraction that challenged the requirements for the long-term protection of the environment.

The best basis inventory (BBI) is the official estimate of the current contents in SSTs and double-shell tanks (DSTs). The BBI data are stored in an electronic database, called the Tank Waste Information Network System (TWINS), that is regularly updated. A query of the TWINS database in late January 2014 and subsequent analysis of the data led to the following observations:

- The 28 DSTs contain more total ^{99}Tc (1.51×10^4 Ci) than the 149 SSTs (1.14×10^4 Ci).

- The DSTs contain 57% (1.51×10^4 Ci) of the total ^{99}Tc inventory (2.65×10^4 Ci) in Hanford tanks, with the bulk (1.33×10^4 Ci) being in four DST farms (AP, AN, AW, and AZ) in the 200-E Area.

A key issue addressed in this literature review was to estimate the amount of ^{99}Tc present as non-pertechnetate (n-Tc) species. This is important because this species has been shown to be difficult to separate from treated liquid tank wastes, should ^{99}Tc removal prior to vitrification become the preferred path forward. Further, there are no data on how n-Tc species will interact in the melters or whether they will partition into the glass similar to the pertechnetate form, which has been studied.

The review of all available studies found that the n-Tc species have not been definitively identified. However, based on all investigations and detailed measurements, n-Tc species in the Hanford DST supernates are tentatively identified as Tc(I) carbonyl complexes and it is speculated that they are derived from either $Tc(CO)_3^+$ or $Tc(CO)_2(NO)^{2+}$ precursor molecules that may lead to final species such as $Tc(CO)_3(gluconate)^{2-}$ as a result of the radiolytic decomposition of organics and nitrite in those DSTs that contain relatively high concentrations of dissolved organic carbon.

A summary of the n-Tc percentages in the supernates from DSTs that have had samples analyzed (10 DSTs) or that can be estimated from similarity of supernates (5 DSTs) shows between 963 and 1,160 Ci of n-Tc species may be present in these DSTs. These 15 DSTs for which we can estimate the mass of n-Tc species contain 5,530 Ci of ^{99}Tc. There is a total ^{99}Tc content of 10,900 Ci in all 28 DST supernates. The 28 DST tanks also contain 4,200 Ci of ^{99}Tc as saltcake and sludge solids. The 13 DST tank supernates for which there is no information on what the n-Tc species content might be contain 5,370 Ci of ^{99}Tc. Therefore, our estimate that there are between 963 and 1,160 Ci of n-Tc species does not include the possibility that there are more n-Tc species in the supernates in the 13 DSTs for which no information is available. Because the BBI tables downloaded from the database do not give any information on what source or type of waste is in the 13 DSTs supernates that have not been directly analyzed for n-Tc species, we cannot speculate on what percentages of n-Tc species might be present in the 13 DSTs. It is possible that the historical tank transfer records could be used to piece together some estimates of the SST source and waste types in these 13 DSTs. However, it was beyond the available resources for this literature review to pursue historical tank transfer records. Further discussion on the n-Tc estimates is found in Section 7.0.

Based on the current WTP process flowsheets, almost all of the ^{99}Tc in retrieved tank wastes after processing in the Pretreatment (PT) Facility will be found in the low-activity portion of the liquid waste sent to the LAW melters. In the LAW melters, Tc is semi-volatile, causing most of it (estimates average around two-thirds) to partition to the off-gas systems. Off-gas treatment equipment downstream of the melters includes a film cooler, submerged bed scrubber (SBS), wet electrostatic precipitator (WESP), heater, HEPA filters, a carbon bed to remove mercury, thermal selective catalytic oxidizer and reducer units, and a caustic scrubber. Most of these off-gas units are used to capture suspended particulates, to condense steam, scrub acid gases, and convert nitrous oxides into nitrogen. The current WTP flowsheets plan to continually recycle the liquid effluents from the SBS and WESP to a receiver tank in the PT Facility, where it will be mixed with fresh LAW feed and evaporated. This combined evaporator concentrate will then be sent to the LAW Facility (or the supplemental LAW (SLAW) treatment facility, to be built in the future) and vitrified.

Matlack et al. (2012)[1] performed the first LAW off-gas fluids recycle testing of a prototype off-gas system using the DM10 melter at the VSL. They used seven different LAW simulants, each spiked with known amounts of ^{99m}Tc, a short-lived gamma-emitting isotope of Tc, in the form of pertechnetate in solution. They performed a mass balance for the ^{99m}Tc throughout the DM10 melter and off-gas treatment system configured in a continuous recycle loop modeled after the WTP process. Key findings from this work include the following (text in quotes is taken directly from the cited document):

- "With recycle, retention of Tc and Re in the glass product is increased by factors of at least 2 to 3 over the corresponding single-pass values for almost all glasses made. For all but two LAW compositions, Tc retention in glass ranged from 68% to 84% of the total."
- "The increase in Tc retention in LAW glass was limited by holdup of material, including Tc in the system, particularly in the WESP internals, the film cooler, and transition line[2]. Mobilization of this material in order to make it available for recycle would likely further increase the retention in glass."
- In the DM-10 off-gas recycle system, there were two places where volatile species could exit the recycle system: 1) as overheads from the evaporator that treated SBS condensates and WESP fluids, and 2) as an exhaust downstream of the WESP and HEPA filters. The percentage of ^{99m}Tc in the melter feed that escaped through these two recycle exits "was less than 0.03% during normal operations and much lower for many tests" for the overheads from the evaporator and "from 0.01 to 0.5%" for the WESP exhaust during normal operations of the WESP. "The fraction of feed Tc exiting the recycle loop through the WESP exhaust was critically dependent on the performance of the WESP and increased to above 10% (i.e., by a factor of about 500 or more) when the WESP was not functioning."
- Likely because of the material hung up on interior metal surfaces and transition lines, the overall mass balance closure for "technetium reached as high as 95% but averaged about 90% over the later tests, which is about 10% lower than that observed for rhenium (Re). This difference likely contributed to the observed difference in the retentions in glass. In comparison, in 72 previously conducted single-pass DM10 tests (without recycle) the average mass balance closures were 97% for technetium and 102% for rhenium."
- Assuming that the Tc hung up on the interior metal surfaces of the off-gas recycle equipment and transition lines can be flushed during the periodic cleaning cycles and that the WESP down time is minimized, the small amount (0.04% to 0.5%) of Tc that was directly measured exiting the overhead of the evaporator and the WESP exhaust suggests that LAW off-gas recycle will continuously return up to 99% of the Tc to the melter until Tc becomes incorporated in the glass.

One potential issue remains in the Tc off-gas capture and continual recycle back through the PT Facility to the LAW and SLAW melters. When the WESP is not operational, the Tc will likely be caught on the heated HEPA filter, which is next in line after the LAW WESP. Currently, the HEPA filters are planned to be placed into drums and sent to the IDF. We recommend that a HEPA filter leaching step,

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[1] Matlack KS, H Abramowitz, M Brandys, IS Muller, RA Callow, N D'Angelo, R Cecil, I Joseph, and IL Pegg. 2012. *Technetium Retention in WTP LAW Glass with Recycle Flow-Sheet: DM10 Melter Testing.* VSL-12R2640-1, Vitreous State Laboratory, The Catholic University of America Washington, DC 20064. This report is found in the Hanford Site IDMS report database as Abramowitz H, M Brandys, R Cecil, N D'Angelo, KS Matlack, IS Muller, IL Pegg, and RA Callow. 2012. *Technetium Retention in WTP LAW Glass with Recycle Flow-Sheet: DM10 Melter Testing.* RPP-54130, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.

[2] The transition line is piping between the film cooler and the SBS.

with the leach water recycled to the PT Facility for return to the melter, be added to the flowsheet if it is critical to reduce the amount of Tc going into secondary wastes.

Regarding the speciation of ^{99}Tc in the off-gas and the resultant fluids, several reports suggest that when the hot gases contact the water in the SBS and WESP systems, ^{99}Tc should become pertechnetate. This assumption seems reasonable given the likely oxidizing environment in the SBS and WESP systems. The Matlack et al. (2012) study did not address any ^{99}Tc speciation issues because only the total ^{99m}Tc activity could be measured.

The main focus of the Tc Management Program is to improve the understanding of the fate of the ^{99}Tc currently stored in the 177 storage tanks at Hanford. Based on the analysis in this review, 26,500 Ci currently reside in the tanks. At a high level there is a good mass balance for how the ^{99}Tc produced at the Hanford Site is currently distributed. About 80% of the ^{99}Tc produced at Hanford currently resides in the 177 storage tanks, ~17% to 20% (based on discussion presented in Section 4.2) was shipped off-site with reprocessed uranium that was recycled/purified for other uses, and ~2% to 3% has been released to the Hanford sediments. The partitioning of the "pool" of ^{99}Tc released to Hanford sediments in the past and in the future after the ^{99}Tc currently in the storage tanks has been dispositioned is and will remain important in the future for making site remediation decisions and site risk assessments, but is not the main scope of the Tc Management Program that is funding this report.

Final observations and recommendations for determining the amount and identity of n-Tc in Hanford tank wastes from this report and the companion document[1] are provided. Unfortunately, there is no clear method for identifying n-Tc species in Hanford tank supernate or the resultant liquids from dissolving tank saltcake and sludges. It is possible to use the pertechnetate-specific "exchange" resins to evaluate the amount of n-Tc species in a sample, but this does not identify what the n-Tc species is/are. Identification of the n-Tc species would be useful to understand how it may be formed in the tanks and how best to convert it to a form that is amenable to separation from liquid wastes or to convert it to pertechnetate, for which removal/separation processes are available. Of the commonly used methods for inorganic compound identification, UV-vis and vibrational spectroscopy are perhaps the most routine, but to date these methods have not provided any useful information or any characteristic signal that can be associated with the presence of n-Tc. To date, only two methods have given information as to the oxidation state and structural features of n-Tc: 1) X-ray absorption spectroscopy (XAS) of one form or another and 2) nuclear magnetic resonance (NMR). Unfortunately, XAS analysis is both time- and labor-intensive and ^{99}Tc NMR cannot observe Tc compounds when in several of the available oxidation states. Still, to the extent possible, continued analysis of new DST supernates for n-Tc—once its presence has been indicated by other methods–by X-ray adsorption near edge structure and/or extended X-ray absorption fine structure is recommended. Size exclusion chromatography may also have some merit on separating ^{99}Tc species from each other prior to analysis (in hopes of simplifying the sample matrix) by the recommended NMR and XAS methods.

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[1] Rapko BM. 2014. *Protocol for Identifying the Presence of and Understanding the Nature of Soluble, Non-pertechnetate Technetium in Hanford Tank Supernatants. PNNL-23180,* EMSP-RPT-020, Pacific Northwest National Laboratory, Richland, WA.

Acronyms and Abbreviations

BARD	Flowsheet Bases, Assumptions, and Requirements Document (a frequently revised WTP document) used to describe current assumptions, flowsheets, and requirement currently Rev. 7
BBI	best basis inventory (a frequently revised electronic database containing estimates of inventory of many components within Hanford tanks)
CSM	conceptual site model
detn	diethylenetriamine; tri-dentate organic ligand
DOE	U.S. Department of Energy
DST	double-shell tank
DTPA	diethylene triamine pentaacetic acid; formula $C_{14}H_{23}N_3O_{10}$; chelating agent
EDDA	ethylenediiminodiacetic acid; formula $C_6H_{12}N_2O_4$ degradation product of EDTA
EDTA	ethylenediaminetetraacetic acid; formula $C_{10}H_{16}N_2O_{8;}$ chelating agent
en	ethhylenediamine; chelator and precursor ligand to make EDTA
EPR	electron paramagnetic resonance
ETF	Effluent Treatment Facility
EXAFS	extended X-ray absorption fine structure; XAS technique
HDW	Hanford Defined Waste; categorization of Hanford reprocessing waste streams
HEDTA	N-(2-hydroxyethyl) ethylenediamine-N,N',N'-triacetic acid; $C_{10}H_{18}N_2O_7$; chelating agent
HEME	high-efficiency mist eliminator
HEPA	high-efficiency particulate air (filter)
HLW	high-level waste
HTWOS	Hanford Tank Waste Operations Simulator; computer code
ICP-MS	inductively coupled plasma mass spectrometry
IDA	iminodiacetic acid; formula $C_4H_7NO_4$; chelating agent
IDF	Integrated Disposal Facility
IHLW	immobilized high-level waste
ILAW	immobilized low-activity waste
LANL	Los Alamos National Laboratory
LAW	low-activity waste
LSC	liquid scintillation counting
MRT	Molecular Recognition Technology
MWd/MTU	megawatt-days per metric ton of uranium; measure of how much energy is extracted from a primary nuclear fuel source
n-Tc	non-pertechnetate species found in Hanford tank supernates
NMR	nuclear magnetic resonance

NOx	generic term for nitrogen oxide gases (primarily mono-nitrogen oxide NO and nitrogen dioxide NO_2)
NTA	nitrilotritacetic acid; formula $N(CH_2CO_2H)_3$; chelating agent
ORP	Office of River Protection
PA	performance assessment
PBS	packed-bed scrubber
PNNL	Pacific Northwest National Laboratory
PT	Pretreatment (Facility)
RA	risk assessment
SBS	submerged bed scrubber
SCO	selective catalytic oxidation unit (part of off-gas treatment equipment)
SCR	selective catalytic reduction unit (part of off-gas treatment equipment)
SLAW	supplemental LAW (treatment facility)
SIM	Soil Inventory Model
SOx	sulfur products; generally refers to SO_2 and SO_3
SRNL	Savannah River National Laboratory
SST	single-shell tank
TCO	thermal catalytic oxidation unit
TC&WM EIS	Tank Closure and Waste Management Environmental Impact Statement
tetn	triethylenetetraamine; chelating agent
TWINS	Tank Waste Information Network System; electronic database
VOC	volatile organic carbon
VSL	Vitreous State Laboratory at The Catholic University of America
WESP	wet electrostatic precipitator; off-gas treatment equipment
WRPS	Washington River Protection Solutions
WTP	Hanford Tank Waste Treatment and Immobilization Plant
XANES	X-ray absorption near edge structure; XAS technique
XAS	X-ray absorption spectroscopy

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1.0 Introduction

This report was funded by the Technetium Management Program. The Technetium Management Program represents an integrated effort that includes work funded directly by the Office of River Protection (ORP), U.S. Department of Energy (DOE) subcontractors, and the DOE Office of Environmental Management. Scientists from Pacific Northwest National Laboratory (PNNL), Savannah River National Laboratory (SRNL), Hanford Site operations contractor Washington River Protection Solutions (WRPS), and several subcontractors (synchrotron support—Stanford Radiation Laboratory, used by collaborators at Lawrence Berkeley National Laboratory), along with experts in Tc sensor development from the University of Cincinnati, are all engaged in performing the work. This report is part of this integrated effort.

The objectives of this report are as follows:

1. Assemble the available information regarding technetium (Tc) inventory, distribution of soluble and insoluble Tc between the three phases in the tanks (supernate, saltcake, and sludge), and Tc speciation in each phase into a single, comprehensive assessment.
2. Discuss anticipated amounts and fate of Tc during melter operations, and recovery and recycle of the fluids from various off-gas units.
3. Document and discuss available information on Tc inventory and distribution in other "pools" on the Hanford Site such as releases to inactive cribs, trenches, and ponds, and releases from single-shell tanks (SSTs) and their infrastructure and in the aquifer.

There remain several significant uncertainties in the understanding and modeling of the fate and speciation of ^{99}Tc currently in the Hanford tanks, and in the glass and low-temperature waste forms that are contemplated as final waste forms destined for permanent disposal in the subsurface. A particularly vexing issue arises should significant Tc removal be required from the waste after it is sluiced from the tanks and sent to "pretreatment" facilities. A significant (2% to 25%) fraction of the ^{99}Tc currently stored in some of the 177 tanks appears to be present as a supernatant-soluble, non-pertechnetate (n-Tc) species that has only been partially identified and, based on experimentation to date, cannot be effectively separated from the tank waste or easily converted to the pertechnetate species that is readily removable from the various liquid wastes.

In Section 4.3 the available information on the n-Tc species is discussed. To summarize, taken in total, the available evidence for existence of soluble n-Tc species in several of the double-shell tank supernates is strong. The evidence includes direct spectroscopic evidence using XANES and ^{99}Tc NMR. Multiple instances and types of indirect evidence also exist involving differing behaviors in the amount of ^{99}Tc in DST supernates sorbed onto pertechnetate-specific resins when compared to the amount of pertechnetate radiotracers spiked into the supernate. Finally, and as the weakest indirect evidence, batch contacts for ^{99}Tc added as pertechnetate in simulants and ^{99}Tc in actual tank waste differ.

In total, approximately 30 studies have been published by researchers from four DOE national laboratories on these findings. Interestingly, good agreement between laboratories as to the amount of soluble n-Tc species deduced by these methods is generally observed. However, **the number and identities of the n-Tc species have not been conclusively determined**. Available data suggest that the soluble n-Tc species have a Tc valence of +1 [Tc(I)] and may be complexed with carbon monoxide or nitrous oxide generated in the high radiation fields within the Hanford tanks.

While there has been significant work on the removal of the pertechnetate anion from tank liquid wastes, there has been limited work in areas such as whether the n-Tc species incorporates into glass similar to pertechnetate, and whether the n-Tc species is captured similarly in off-gas systems downstream from the melters like pertechnetate. One preliminary test has been performed to explore capturing pertechnetate from off-gas streams, in equipment that removes volatiles and entrained solids from the glass melters, and to quantify the recycle of the pertechnetate back to the LAW melter. This test is summarized in Section 5.0.

2.0 Quality Assurance

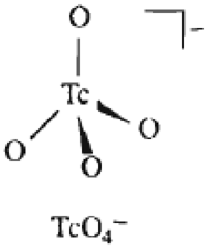
This work was performed in accordance with PNNL's Quality Management System Description and associated Quality Assurance Program Description (QAPD), which are maintained electronically as part of the HDI system. Details of this project's approach to assuring quality are contained in the *Environmental Management Support Program Quality Assurance Plan* (QA-EMSP-001). Preparation of this report was conducted in accordance with QA-EMSP-1102, *Scientific Investigation for Applied Research.* All staff members contributing to the work received proper technical and quality assurance training prior to commencing quality-affecting work.

3.0 Background

Tc is one of the most difficult contaminants to address at the DOE Hanford Site because of its complex chemical behavior in tank waste, limited incorporation in mid- to high-temperature immobilization processes (vitrification, steam reformation, etc.), and high mobility in subsurface environments. Approximately 32,600 to 34,000 Ci of ^{99}Tc were produced at the Hanford Site (see Section 4.1). Of this, a small amount either has been released purposefully to cribs/trenches or has leaked from the SSTs, and an even smaller amount is found in the unconfined aquifer below the Hanford Site. Thus, most (~26,500 Ci; see Section 4.2) of the ^{99}Tc inventory remains in the Hanford storage tanks (149 SSTs and 28 double-shell tanks [DSTs]) and must be dispositioned into approved waste forms.

Because the majority of the ^{99}Tc will be immobilized in the final LAW form(s), with a smaller fraction immobilized as a secondary waste, it will remain on the Hanford Site and be disposed at the Integrated Disposal Facility (IDF); only a small fraction will be shipped to a geologic repository as part of the immobilized high-level waste (IHLW). Past performance assessment studies, which focused on groundwater protection, have shown that ^{99}Tc would be the primary dose contributor to the IDF performance. Due to its solubility and potential volatility during high-temperature vitrification processes, effective Tc management is important to the success of the overall ORP mission. Removal of Tc from LAW and/or off-gas secondary wastes would eliminate a key risk driver for the IDF performance assessment (as long as the separated Tc was shipped off-site or immobilized in a robust waste from with performance equivalent to glass).

Tc in its typical Tc(VII) [pertechnetate; TcO_4^-] form (see Figure 3.1) is problematic in LAW due to its long half-life (213,000 years), complex redox chemistry, high solubility, and volatility at high temperatures. While most of the long-lived radionuclides in tank waste (e.g., plutonium and uranium) are largely insoluble under storage conditions, such as high pH and low temperature, Tc is predominantly in the aqueous phase and is highly mobile in Hanford's subsurface environments—vadose zone and aquifer sediment pore-water/groundwater. In addition, Tc volatility at LAW vitrification melter temperatures creates the potential for Tc concentrations in the secondary waste streams formed when treating the melter off-gas to be a long-term risk unless judiciously processed or recycled and incorporated into the glass. To reduce the technical uncertainty associated with the long-term environmental impact of Tc, the Tc Management Program is addressing these ^{99}Tc management issues by evaluating and developing science and engineering options to treat, immobilize, and dispose of the ^{99}Tc contained in tank wastes.



bond length	(Tc–O) = 1.711(3)Å
ΔG_{hydr}	-60 Kcal/mole
reduction potential	TcO_4^-/TcO_2 $E^\circ = 0.74$ V

Figure 3.1. Structure of the Pertechnetate Anion and Selected Thermodynamic Properties (from Schroeder et al. (1995))

Over the past two decades, the effectiveness of anion exchange-like resins has been examined for their ability to remove soluble Tc from Hanford tank supernatants. As discussed in Rapko et al. (2013a,b) and elsewhere, in most cases, excellent removal of pertechnetate Tc has been observed. However, in several instances, there appeared to be a source of Tc that behaves differently than pertechnetate, [TcO_4^-]. The fraction of soluble Tc that is not pertechnetate appears to vary significantly between tanks as well, ranging from effectively none to being the majority of the soluble Tc present in select tank supernates (see Tc Management Program Task 1 companion report, Rapko 2014 and references within).

This report is organized into eight sections and four appendices that document available information of the three objectives described in the introduction.

- Section 1.0 is the introduction and Section 3.0 includes background.
- Section 4.0 covers objective 1, which summarizes information on Tc inventory, distribution of soluble and insoluble Tc between the three phases in the tanks (supernate, saltcake, and sludge), and Tc speciation in each phase into a single, comprehensive assessment.
- Section 5.0 covers available information on objective 2, anticipated Tc recycle and recovery during melter operations, and off-gas recycle testing data.
- Section 6.0 covers objective 3, which briefly reviews available information on Tc inventory and distribution in other "pools" on the Hanford Site such as releases to inactive cribs, trenches, and ponds and releases from SSTs and their infrastructure.

- Section 7.0 summarizes the information presented in the report and Section 8.0 lists references cited in the main report.

The appendices provide additional details on several of the objectives:

- Appendix A presents additional data on estimated inventory of ^{99}Tc currently stored in the 177 Hanford tanks.
- Appendix B lists available reports and a short summary of findings on ^{99}Tc leachability from residual sludges left in SSTs after retrieval.
- Appendix C describes the Hanford Tank Waste Operations Simulator (HTWOS) computer code, which is used to estimate the partitioning of radionuclides and chemicals retrieved from the Hanford tanks as they are processed through the Hanford Tank Waste Treatment and Immobilization Plant (WTP).
- Appendix D briefly discusses and lists many available documents with information on the distribution and fate of ^{99}Tc not currently stored in the 177 Hanford tanks.

4.0 Tc Inventory, Distribution, and Speciation in Hanford Tanks

An understanding of the total Tc inventory currently stored in Hanford SSTs and DSTs is useful for several reasons. Human health and environmental impacts are proportional to the amount (or inventory) of the key contaminants present at a given disposal/storage site. Thus, knowing the inventory for key contaminants is crucial to risk assessments. At the Hanford Site, the final disposition of LAW will be on site in the IDF, a shallow land burial facility. Groundwater is the most probable pathway to the biosphere for contaminants presently 1) stored in SSTs and DSTs, 2) currently in the vadose zone sediments from past disposal to trenches, cribs, and ponds, and 3) released inadvertently from SSTs. Given that Tc is quite mobile when present in its pertechnetate form and that the majority of Tc currently on site is stored in the 177 Hanford tanks, a thorough understanding of the current inventory in the Hanford tanks is key to any risk assessment of the impacts of Tc present at the Hanford Site. Past long-term groundwater impact predictions (Mann et al. 2001 [2001 IDF PA]; Mann et al. 2003 [Supplemental ILAW risk assessment]; DOE 2012 [TC&WM EIS]) show that ^{99}Tc reigns as the radioactive waste constituent contributing the highest impacts.

4.1 Total Tc Production (Historical Inventory)

A key to performing a mass balance on the ^{99}Tc at the Hanford Site is an estimate of the total mass (kilograms) or activity (curies) that was produced during the irradiation of fuel used to produce plutonium. A single source of inventory data does not exist for the 60-plus years of Hanford operations. However, records on the amounts and types of fuel (percentage of ^{235}U enrichment, Th-based fuel) and reactor burn-up were fairly well documented in comparison to records of liquid and solid waste discharges and disposals.

Detailed records show that ~99,000 metric tons of uranium were processed in separations facilities at the Hanford Site (Kupfer et al. 1997, Appendix B). Table 4.1 (from Kupfer et al. 1997) provides details on the dates and facilities used for reprocessing the irradiated fuels, including two campaigns where thorium oxide was irradiated. Figure 4.1 (from DOE/ORP 2010) shows the reprocessing timeline and states ~100,000 metric tons of irradiated fuel were processed. These two sources generally agree on the mass of fuel that was irradiated and processed to extract plutonium. Detailed records of the dates, fuel types, and fuel cladding loaded into each Hanford reactor and the fuel exposure (megawatt days per metric ton of fuel [MWd/MTU] for monthly and for some shorter periods were documented. In all, ~1300 distinct fuel batch records are available that document the dates, fuel and cladding types, fuel tonnage placed in reactor, burn-up (exposure), date irradiated fuel was removed from reactor, cooling time before separation, and dates of reprocessing to remove the plutonium (and later also uranium in the REDOX and PUREX processes). The type of cladding, especially impurities within, has an important impact on the amounts and types of activation products that are formed during the irradiation of the fuel elements. Because ^{99}Tc is a fission product, its production is not sensitive to the cladding type. The fuel reprocessing began in 1944 and ended in 1989.

Most recently, the ORIGEN2 code (Croff 1980) has been used to calculate the curie amounts of fission and activation products created during the fuel irradiations using the detailed historical records (just mentioned) available at the Hanford Site. To limit the number of ORIGEN2 runs needed to calculate the production of fission and activation products, the ~1300 fuel batch records were categorized into 15 bins of similar conditions and only 15 distinct ORIGEN2 runs were performed to estimate the masses and activities of fission and activation products. Note that some fission products are stable nuclides. A second computer code, DKPRO, was created at Hanford to generate output files that conveniently list the curie values for 46 key radionuclides associated with each of the ~1300 batches of irradiated fuel that was processed through the various reprocessing plants. DKPRO splits the calculated activities of the 46 radionuclides into two categories: 1) curies in the cladding and 2) curies in the fuel "cores."

Table 4.1. Hanford Site Fuel Reprocessing Plant Summary (from Kupfer et al. 1997)

	Aluminum Cladding		Zirconium Cladding
Separations Plant (operating periods)	Natural Uranium (MT)	Low-Enriched Uranium (MT)	Natural Uranium (MT)
T Plant (1944 to 1956)	5,034	0	0
B Plant (1945 to 1952)	2,766	0	0
REDOX (1952 to 1966)	11,609	7,852	245
PUREX (1956 to 1972)	58,748	7,176	1572.3
PUREX (1983 to 1989)	0	0	3,890.4
Thorium Oxide Purex (1966, 1970)	629 (ThO_2)	--	--

Table 4.2 shows the range in total curie inventory of ^{99}Tc produced at the Hanford Site—as found in various technical reports and environmental impacts statements and decay corrected to January 1, 2000—created from irradiating fuel to produce plutonium. The slight difference in total curies appears to be caused by the earlier estimates using a different code (RIBD-II; Gumprecht 1968) than ORIGEN2 to

calculate fission product production rates per fuel burn-up (MWd/MTU). Because of the long half-life of ^{99}Tc, making decay corrections to the present (January 1, 2014) does not change the inventory estimates. The ~32,600 Ci of ^{99}Tc produced at Hanford equates to 1922 kg of ^{99}Tc.

Table 4.2. Range in Total Tc Inventory (curies) Produced at the Hanford Site

^{99}Tc Inventory (curies) Decay corrected to 01/01/2000	Source
3.43×10^4	Puigh and Wood 2005; Table 2-1 based on Watrous 2002
3.26×10^4	Kupfer et al. (1999) (Table ES-2); Kupfer et al. (1997) (Table ES-2)
3.21×10^4	DOE 1996 (Appendix A), DOE 1997 (Table 2.11)
3.13×10^4	Kincaid et al. (2006)
^{99}Tc Inventory (curies) Decay corrected to 01/01/2014	
3.2598×10^4	Kupfer et al. (1999) (Table ES-2); Kupfer et al. (1997) (Table ES-2)
3.2098×10^4	DOE 1996 (Appendix A), DOE 1997 (Table 2.11)
3.13×10^4	Kincaid et al. (2006)

The only other sources of ^{99}Tc not present in spent fuel in inactive and active reactors (Columbia Generating Station) on the Hanford Site would be imported from other sites. The commercial low-level waste burial ground located south of the 200-E Area, currently operated by US Ecology, Inc., has ^{99}Tc inventory not related to Hanford fuel reprocessing. Kincaid et al. (2006) estimated that the US Ecology commercial burial ground contained ~50.3 Ci of ^{99}Tc. The Final Environmental Impact Statement for the US Ecology site in Table 2.D states that between the start of operations through 2002, an estimated 50.1 Ci of ^{99}Tc have been disposed at the site, and it is projected that the average ^{99}Tc curies expected each ensuing year is 0.093 Ci/yr. The Final Environmental Impact Statement for the US Ecology site can be accessed at http://www.doh.wa.gov/Portals/1/Documents/Pubs/320-031_vol1_w.pdf. Records of waste inventories and volumes disposed at the US Ecology burial grounds are kept at the Washington State Department of Health. Through calendar year 2012, the Washington State Department of Health Waste Management Division documents a total ^{99}Tc inventory at the US Ecology site at 50.2 Ci.[1]

The current Radioactive Materials License (WN-I019-2 Amendment 40) issued by the Washington State Department of Health states that the maximum amount of ^{99}Tc that can be disposed at the US Ecology site is 55.10 Ci, so this would appear to set an upper bound on the ^{99}Tc that could be in the commercial burial ground and this quantity is insignificant compared to the ^{99}Tc produced during the fuel irradiation to generate plutonium. The Radioactive Materials License (WN-I019-2 Amendment 40) is available at http://www.usecology.com/richland_docs_forms.htm.

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[1] Phone conversation with Kristen Schwab (Washington Department of Health, Waste Management Division) on March 5, 2014. Also can be found in Table 1 of *Facility Utilization Report -2012* prepared by US Ecology but not available on the internet and provided by Kristen Schwab.

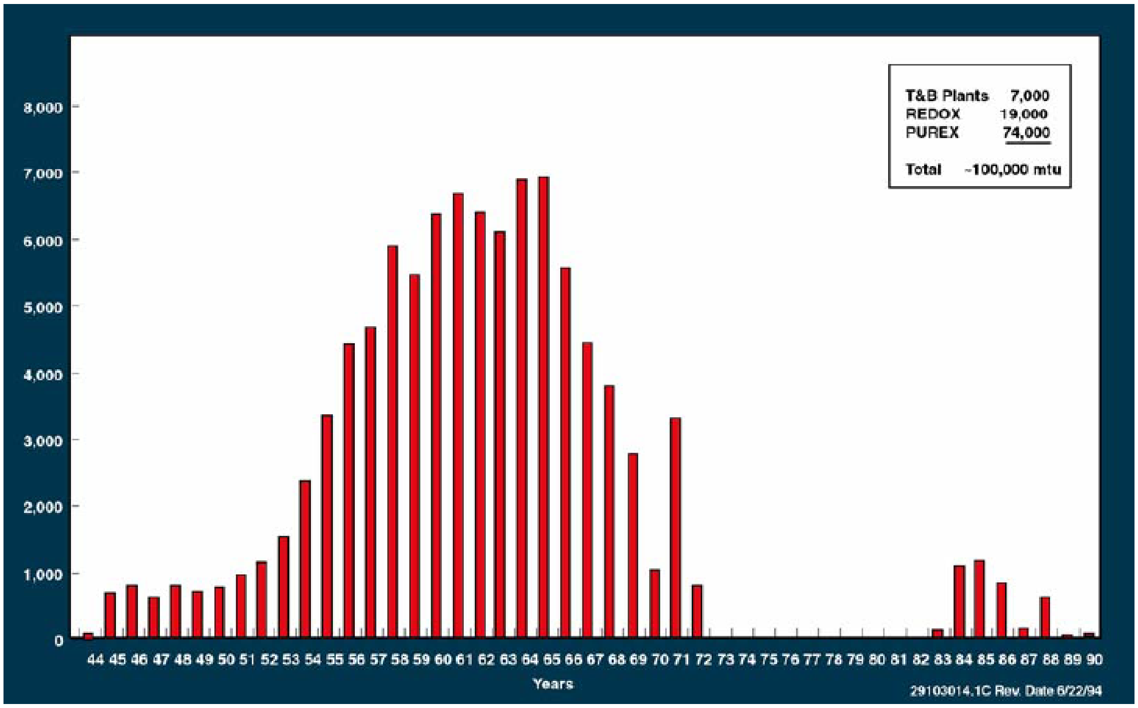

Figure 4.1. Timeline of the Hanford Site Uranium Fuel Reprocessing (Metric Tons of Uranium); from DOE/ORP 2010

A second off-site source of ^{99}Tc that may in the future be shipped to the Hanford Site is discussed in the *Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site, Richland, Washington* (TC&WM EIS; DOE 2012). Appendix D of the TC&WM EIS discusses the potential shipment of nuclear waste from other DOE sites for final disposal at Hanford. Whether or not such off-sites wastes will be allowed to be shipped to Hanford is an ongoing legal debate. Appendix D, Table D-87 of DOE 2012 estimates that the maximum off-site ^{99}Tc inventory that might be shipped to Hanford for final disposal is 1460 Ci (86 kg) [see http://www.hanford.gov/page.cfm/FinalTCWMEIS]. The total ^{99}Tc produced or shipped onto the Hanford Site can be used as the starting inventory for comparing with ^{99}Tc inventories in other "pools" such as in SSTs and DSTs, present in the vadose zone from purposeful disposals to cribs, trenches, and ponds, and inadvertently released/leaked from SSTs, etc. A brief discussion of ^{99}Tc mass balance and the inventories in these other "pools" is presented in Section 6.0. Finding a good mass balance on ^{99}Tc at the Hanford Site would be of great value to future decision makers responsible for selecting remediation alternatives and to convince stakeholders that we understand what quantities and where this problematic contaminant resides at the Hanford Site.

4.2 ^{99}Tc Inventory Currently Stored in Hanford Tanks

For several decades there has been a concerted effort to estimate the inventory of key radionuclides and chemicals within the 177 storage tanks (149 SSTs and 28 DSTs) at the Hanford Site. The key reason estimates of the inventories of radionuclides and chemicals and the volumes of waste in the tanks are needed is to provide basic information to the complicated task of removing the wastes from the tanks and determining the size and logistics of the processing facilities needed (e.g., WTP) to convert the wastes into acceptable final solid waste forms and to project the volumes of final solid wastes and types (IHLW, immobilized low-activity waste [ILAW], and transuranic waste). Each of these three waste types likely will need to be disposed at different facilities and locations.

Over the years there have been several attempts to estimate how much of the total ^{99}Tc produced at Hanford remains in the storage tanks and to determine the inventory in each tank. Estimating the individual tank ^{99}Tc inventories relies on actual sample analyses when available and waste processing knowledge and waste transfer (both into and out of each tank) records. The waste processing knowledge relies on the same ~1300 distinct fuel batch records mentioned in Section 4.1, but relies on different computer codes and methodologies to calculate how much of the produced fission and activation products that formed in the irradiated fuel ended up in the waste streams created during fuel dissolution and reprocessing.

Because some of the reprocessing wastes were sent directly to cribs and trenches, not all reprocessing wastes ended up in the Hanford storage tanks. Several different conceptual models and computer algorithms were created over the years to estimate the split in the mass/activity of chemicals and radionuclides that ultimately went to the storage tanks. One of the earlier tank inventory protocols was named TRAC (see for example Jungfliesh and Simpson 1993).

As time progressed, efforts to refine the methodology and computer codes for predicting inventories in the storage tanks continued such that by about 1999 (see Kupfer et al. 1999) a suite of computer codes that collectively produced a best basis inventory (BBI) of the waste volumes, separated into three phases (supernate, saltcake, and sludge), and inventories (curies for radionuclide and kilograms for chemicals) was adopted. Historical tank inventories thus varied with time both because of transfers in and out of the tanks and because different "split" factors were used to estimate the amount of each constituent in each of the many waste streams generated by the fuel reprocessing steps. In addition, wastes from the bismuth phosphate fuel reprocessing were later removed from SSTs and reprocessed to recover uranium. Other isotope recovery campaigns using yet additional processes removed ^{90}Sr, ^{137}Cs, and for a small volume of tank waste other nuclides/chemicals deemed of economic or scientific importance. A good summary of the three main fuel reprocessing operations and the later U, ^{90}Sr, ^{137}Cs, and other element separation/recovery processes is found in Appendix B of DOE/ORP 2010.

In summary, the history of various fuel reprocessing and isotope recoveries, utilization of and transfers between the 177 storage tanks, and difficulties obtaining and analyzing samples from the tanks presents a complicated chemical, logistical, and bookkeeping challenge to estimating the inventories in each tank. This challenge has received much attention and for decades efforts have been made to improve and refine both conceptual and mathematical models to establish defensible estimates of the inventory of chemicals and radionuclides within the storage tanks. The reconciliation process used to produce the global (all tanks) and individual tank inventories has been described in detail by Kupfer et al. (1997, 1999). One key component of the inventory process is determining the fate of each component (element, nuclide, etc.) during the dissolution and reprocessing of the fuel to extract plutonium and later both plutonium and uranium. One model used early in the inventory estimation exercises in conjunction with DKPRO (the bookkeeping algorithm) was called SPLIT. Later, scientists at Los Alamos National Laboratory (LANL) created a different and independent conceptual and computer algorithm, called the Hanford Defined Waste (HDW) model, to follow the fate of key constituents. The HDW model simulates the paths of the irradiated fuel and cladding into the process plants, where the chemical operations took place. Chemicals added to process the irradiated fuel and treat the resultant liquid streams are also tracked. Improvements to HDW over time included treating fuel and cladding separately as discrete entities, incorporation of process losses, improved chemical process models, redefining selected physical and chemical assumptions, and an improved solubility model. HDW has undergone five revisions over the years in attempts to accurately determine the chemical fate (solubility-precipitation reactions) and waste stream association (how much ends up in which waste stream) for each key chemical and radionuclide for each fuel reprocessing or isotope recovery operation. The current version, HDW version 5, is discussed in detail in Higley and Place 2005. Estimating the solubility of key constituents in each process is a key to determining how much of the starting inventory goes to each waste stream, and is an important consideration because cascading of wastes was a common practice in the 1950s.[1] The SPLIT code did not account for inventory losses from the cascading activities.

SPLIT assumed that 24% of the ^{99}Tc present in reprocessed fuels using the PUREX process and 37% in the REDOX process remained with the uranium product and thus was not sent to storage tanks. SPLIT assumed that only <1% of the ^{99}Tc in the bismuth phosphate fuel reprocessing campaign remained with the plutonium product after the first two precipitation/purifications steps and did not get sent to storage tanks. Serne et al. (2007) corroborate the fact that ≈ <1% of the ^{99}Tc in the bismuth phosphate fuel reprocessing campaign was **not** sent to the SSTs. After accounting for the larger quantity of fuel reprocessed using the REDOX and PUREX processes, the HDW version 5 model estimates that approximately 20% to 23% of the ^{99}Tc produced in the reactors was shipped off-site with recovered uranium, not sent to the Hanford storage tanks[2]. In contrast, the early versions of the HDW model did not consider ^{99}Tc remaining with the uranium product in the REDOX and PUREX reprocessing campaigns but did consider ^{99}Tc "losses" from tank inventories via the cascading activities. HDW version 5 (see Higley and Place 2005) does account for ^{99}Tc sent off-site with uranium product.

Table 4.3 lists some early-to-present total ^{99}Tc inventory estimates in the Hanford storage tanks. A more complete listing of ^{99}Tc total inventories in the storage tanks and a description of how they were derived is found in Puigh and Wood 2005. Figure 4.2, taken from Puigh and Wood 2005, shows a slow reduction in the ^{99}Tc total inventory in the storage tanks with time, likely because of a combination of more sampling and chemical analyses availability and refinements in the HDW model that estimates the fate of ^{99}Tc in the overall reprocessing chemistry. The only high ^{99}Tc inventory value (4.7×10^4 Ci) is based on limited DST sampling data and HDW version 1, which did not allow any loss of ^{99}Tc to waste streams not sent to tanks.

The BBI is the current official estimate of the current contents (46 radionuclides and 25 chemicals) in SSTs and DSTs and is based on Kupfer et al. (1999). Separate inventories are provided for supernate, saltcake, and sludge waste. Supernate, especially in the DSTs, is an aqueous solution of sodium nitrate, nitrite, and hydroxide, and various organic compounds including citrate, gluconate, formate, oxalate, ethylenediaminetetraacetate (EDTA), and nitrilotritacetate (NTA). Most of the supernate in the SSTs has been removed over the past several decades to promote SST stabilization. Saltcake consists of

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[1] Waste cascading was a process where fuel reprocessing liquid wastes containing suspended solids were piped into a chain (usually three) of connected SSTs. As the waste filled the first tank, suspended solids settled and the relatively clarified liquid would then flow into the second tank (buried at a slightly lower elevation), with the prospect that any residual suspended solids would settle out before the supernate flowed into the third tank (again buried lower than the second tank). Ultimately, very-clarified supernate from the third tank would flow directly to cribs, where the clarified liquid seeped into the ground.

[2] It was beyond the scope of this report to determine how these values were chosen and, as discussed in Section 6.0, based on other inventories of ^{99}Tc still present on the Hanford Site, this range in percentage shipped off-site with uranium seems too high.

water-soluble salts that have precipitated during reduction of supernate volume by evaporation and consists mainly of sodium nitrate and nitrite. Sludge consists of the waste components that are insoluble under strongly alkaline conditions and includes most of the fission products and actinides plus large quantities of aluminum and iron oxides and aluminosilicates. Whenever possible, measured analytical values for each particular phase are used to develop the BBI. If such actual measurements for any of the three phases are not available, then measured values in a similar phase in another tank with similar waste history are used. Finally, if no analogous measurements are available, then values from the HDW model are used. In most cases, inventory estimates for radionuclides, including ^{99}Tc, are based on estimated values from the HDW model.

Table 4.3. Selected ^{99}Tc Inventory Estimates Within Hanford's 177 Storage Tanks

^{99}Tc Inventory (curies) and Approximate Date for Estimate	Split Between SSTs and DSTs	Source
2.99 x 10^4 (1980)		Jungfliesh and Simpson (1993)
3.00 x 10^4 (1990)	2.00 × 10^4 in SSTs 1.00 × 10^4 in DSTs	Morgan et al. (1988)
2.83 x 10^4 (1997-99)	Total ^{99}Tc reprocessed	Kupfer et al. (1999) (Table ES-2; Table 6.1-3)
2.47 x 10^4 (1997)	Used SPLIT model to estimate ^{99}Tc that stayed with U product	Kupfer et al. (1999) (Table 6.1-3)
2.97 x 10^4 (2002)	1.55 × 10^4 in SSTs 1.42 × 10^4 in DSTs	DOE TC&WM EIS; DOE 2012
2.64 x 10^4 (2010)	Used HDW version 5, removes ^{99}Tc that went off-site with U product	DOE TC&WM EIS; DOE 2012
2.68 x 10^4 (2004)	Includes future Hanford tank receipts (see Kirkbride et al. 2005)	Puigh and Wood (2005), (Table ES-1)
2.64 x 10^4 (2012)	BBI in TWINS database July 2012	Robbins and May (2013)
2.65 x 10^4 (2014)	Current BBI in TWINS database 01-24-2014	This report

The requirements for the BBI process are given in Place 2006. A very detailed protocol (see Nguyen 2010) for evaluating analytical data from cores, grab samples, and archived samples is followed. This protocol accounts for potential sample evaporation during storage, potential matrix interferences, and how to average data from multiple samplings of a particular phase. If more than one analytical method can be used for measurement, the one that has shown more accurate results is chosen, and other considerations are reviewed by a team of experienced tank experts. Today the BBI is updated quarterly for each of the 177 storage tanks for which changes in volume and inventories (caused by retrieval activities or tank-to-tank transfers) have occurred or new data becomes available, such as new sampling and analyses activities. The BBI data are stored in an electronic database called the Tank Waste Information Network System (TWINS), which can be found at the following URL[1] for those with access to the Hanford intranet https://twins.labworks.org/twinsdata/Forms/About.aspx.

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[1] TWINS is operated and maintained by the Pacific Northwest National Laboratory for Washington River Protection Solutions. Anyone who has a PNNL or HLAN account has access to this site. Others who require access to TWINS data may request authorization to do so via an external website. Requests may be made to matthew_rodgers@rl.gov or by calling (509) 376-2993.

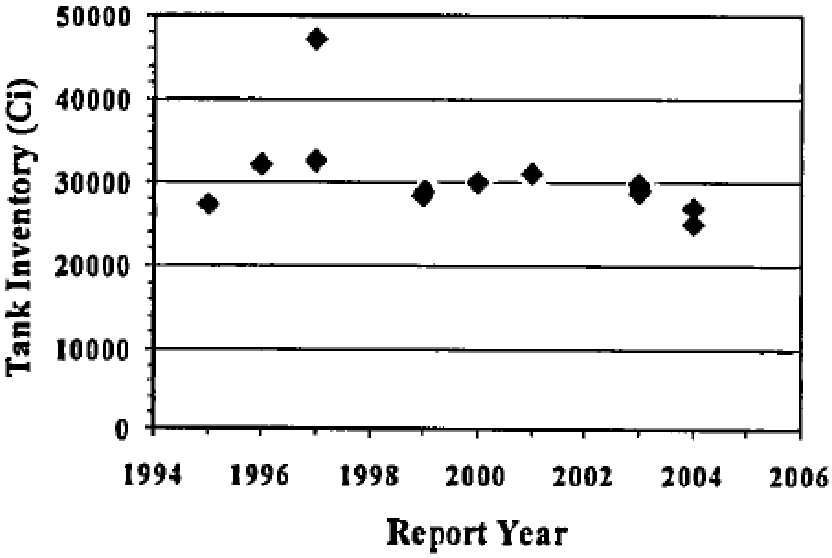

Figure 4.2. Trend in ^{99}Tc Inventory Estimates in Hanford Storage Tanks as Function of Time (from Puigh and Wood 2005)

Table 4.5 and Table 4.6 were constructed using data for the 177 Hanford storage tanks downloaded from TWINS on January 27, 2014, and are the current BBI for the 177 Hanford storage tanks. More details from TWINS are presented in Appendix A. Assuming that the BBI is an accurate tabulation of the current inventory in the Hanford storage tanks, Table 4.4 shows the total ^{99}Tc inventory in each tank farm ranked from highest to lowest total ^{99}Tc and differentiated between SST and DST farms. Appendix A shows the ranking of highest to lowest ^{99}Tc inventories for all 177 tanks. Observations from the data in Table 4.4 show that there is more total ^{99}Tc (1.51×10^4 Ci) in the 28 DSTs than in the 149 SSTs (1.14×10^4 Ci). The ^{99}Tc present in three of the 200-W farms (TX, S, and SX) accounts for 54% of the ^{99}Tc in all SSTs (and 23.4 % of the total ^{99}Tc inventory) and in 200-E the BY SST farm contains ~14% of the total ^{99}Tc in all SSTs (or 6% of the total ^{99}Tc inventory). The DSTs contain 57% of the total ^{99}Tc inventory in Hanford tanks, with the bulk (1.33×10^4 Ci) within four DST farms (AP, AN, AW, and AZ) in the 200-E Area.

Table 4.5 shows that the supernate in the SSTs contains insignificant (32.8 Ci or 0.1% of the total ^{99}Tc in Hanford storage tanks) amounts of ^{99}Tc. This is expected because the SSTs have been rigorously pumped to remove drainable liquids, leaving only very small volumes of supernate. On the other hand, the SST saltcake (combined solids and un-pumpable liquids) contains the bulk (1.02×10^4 Ci) of the ^{99}Tc in SSTs. The saltcake ^{99}Tc may be more present as liquid within the saltcake solids that could not be removed by salt-well pumping as opposed to being present as discrete ^{99}Tc-bearing evaporated salts. In Appendix A, the ^{99}Tc distribution between the three major phases is split further into 5 categories: 1) supernate, 2) saltcake liquids, 3) saltcake solids, 4) sludge liquids, and 5) sludge solids for many tanks when samples were obtained and the saltcake and sludge phases were either manipulated (e.g., centrifuged or filtered) or allowed to gravity separate into a liquid phase and solid phase. Within the SSTs only about 5% (1230 Ci) of the total ^{99}Tc inventory is associated with sludge. In contrast, within the DSTs 1.09×10^4 Ci, or 41%, of the ^{99}Tc is found in the supernate and is thus considered “soluble.” About 13% (3520 Ci) of the total ^{99}Tc inventory is found in DST saltcake and 652 Ci, or 2.5%, in DST sludge. Again, Appendix A shows more detailed data on the ^{99}Tc distribution between liquids and solids in the DST saltcake and DST sludge. Given the current BBI values, overall, 41.2% of the total ^{99}Tc in the 177 storage tanks is found in supernate, 51.6 % is associated with saltcake, and 7.2 % is associated with sludge.

Table 4.4. Total ^{99}Tc Inventory (Ci) in Each Tank Farm

SST Farm	^{99}Tc (Ci)	SST Farm Ranking	Overall Ranking	DST Farm	^{99}Tc (Ci)	DST Farm Ranking	Overall Ranking
TX	2.52E+03	1	4	AP	4.79E+03	1	1
S	2.20E+03	2	5	AN	3.95E+03	2	2
U	1.66E+03	3	6	AW	2.90E+03	3	3
BY	1.55E+03	4	8	AZ	1.63E+03	4	7
SX	1.48E+03	5	10	SY	1.55E+03	5	9
A	7.15E+02	6	11	AY	2.38E+02	6	14
AX	3.87E+02	7	12				
BX	3.66E+02	8	13				
B	2.08E+02	9	15				
T	1.51E+02	10	16				
C	1.02E+02	11	17				
TY	9.57E+01	12	18				
SST Total	**1.14E+04**			DST Total	**1.51E+04**		
		Total ^{99}Tc	**2.65E+04**	**curies**			

Table 4.5. Total ^{99}Tc Inventory (Ci) in Each Phase by Tank Farm

SST Tank Farm	Supernate	Saltcake	Sludge	Total
A	8.48E+00	5.70E+02	1.38E+02	7.15E+02
AX	0.00E+00	3.62E+02	2.54E+01	3.87E+02
B	3.38E+00	1.11E+01	1.95E+02	2.08E+02
BX	4.20E+00	1.65E+02	1.96E+02	3.66E+02
BY	0.00E+00	1.55E+03	1.40E+00	1.55E+03
C	5.67E-03	5.77E-02	1.02E+02	1.02E+02
S	1.04E+00	2.11E+03	9.16E+01	2.20E+03
SX	0.00E+00	1.34E+03	1.46E+02	1.48E+03
T	9.91E+00	3.47E+01	1.06E+02	1.51E+02
TX	2.23E+00	2.36E+03	1.57E+02	2.52E+03
TY	2.55E-01	2.75E+01	6.80E+01	9.57E+01
U	3.33E+00	1.63E+03	2.65E+01	1.66E+03
SST (TOTAL)	3.28E+01	1.02E+04	1.25E+03	1.14E+04
% of Total	**0.1%**	**38.3%**	**4.7%**	**43.2%**
DST Tank Farm	Supernate	Saltcake	Sludge	Total
AN	2.18E+03	1.55E+03	2.23E+02	3.95E+03
AP	4.47E+03	3.13E+02	1.34E+01	4.79E+03
AW	1.96E+03	8.72E+02	6.49E+01	2.90E+03
AY	1.97E+02	0.00E+00	4.07E+01	2.37E+02
AZ	1.49E+03	0.00E+00	1.37E+02	1.63E+03
SY	5.96E+02	7.84E+02	1.73E+02	1.55E+03
DST (TOTAL)	1.09E+04	3.52E+03	6.52E+02	1.51E+04
% of Total	**41.1%**	**13.3%**	**2.5%**	**56.8%**

% of Total is total ^{99}Tc in all 177 storage tanks = **2.65E+04 Ci**. Rounding errors impact the last significant figure in sum of the individual phases in the SSTs and DSTs.

Another important consideration is how each tank's ^{99}Tc inventory was estimated. The BBI protocol uses several "bases" or categories to represent how the inventory value was derived. The bases/categories are shown in Table 4.6, briefly explained, and ranked by technical robustness or certainty.

Table 4.6. BBI Categories Used to Described How Inventory Values Were Derived

Category	Category Description	Level of Certainty
S	Sample based; values from different samplings of the same layer in a specific tank are averaged; sometimes samples from two or more tanks that contain the same waste type are averaged and designated "S."	Highest
E	Process knowledge based; generally derived from sample-based results; concentrations generated from new data such as 1) weighted average values for waste transfers, 2) reconstituted values for centrifuged samples or precipitated solids, 3) pre-1989 sample data that are not in TWINS because QA/QC was not as good as today, and 4) sample data from another similar tank are applied.	Second highest
TS	Template sample based; tank-specific sampling or process knowledge values not available; TS values are average values for the waste type (designated for the tank in question) derived from analytical results from two or more tanks that contain the designated waste type.	Third highest
TE	Template engineering based; tank-specific sampling or process knowledge values not available; TE values are from version 5 of the HDW model (RPP-19822). Values are based on fuel activity estimates, separation plant process records, and waste transfer records.	Lowest

Table 4.7 lists tanks and their ^{99}Tc inventories that have at least one of the phases in the tank actually sampled and then the ^{99}Tc directly measured (earning and "S" basis) or had the ^{99}Tc inventory estimated based on process knowledge (earning the "E" basis). These two basis categories are of higher quality or certainty. It can be seen in Table 4.7 that some portion of the tank contents (supernate, saltcake, or sludge) was actually sampled and the ^{99}Tc directly measured for 49 tanks, containing 1.00×10^4 Ci of ^{99}Tc. This represents 37.8% of the total ^{99}Tc estimated to reside currently within all the tanks. An additional 26 tanks have some portion of their tank content ^{99}Tc mass known from "process knowledge," the second-best quality basis. These 26 tanks contain an additional 6.51×10^3 Ci of ^{99}Tc, or an additional 24.6% of the total ^{99}Tc inventory estimated to reside currently in the Hanford tanks. Thus, about 62.4% of the total ^{99}Tc inventory estimate is based on more-certain information, leaving 37.6% of the total estimated ^{99}Tc inventory as more-uncertain.

Table 4.7. Tanks and ^{99}Tc Inventory (Ci) that Have Better Certainty

Tank	Total ^{99}Tc	Basis	Tank	Total ^{99}Tc	Basis
AN-101	2.01E+02	S/E/TS/TE	A-102	3.70E+01	E/TE
AN-102	5.80E+02	S/E	A-103	2.59E+02	E/TE
AN-103	6.81E+02	S	A-106	1.48E+02	E
AN-104	7.94E+02	S	AN-106	1.44E+02	E/TE
AN-105	1.12E+03	S	AP-101	7.55E+02	E
AN-107	4.34E+02	S/E	AP-102	7.03E+02	E
AP-107	1.77E+02	S	AP-103	9.59E+02	E/TS
AW-101	8.18E+02	S	AP-104	1.55E+02	E
AW-103	3.97E+02	S/E/TS	AP-105	9.51E+02	E/TS
AW-106	5.93E+02	S/TS	AP-106	3.68E+02	E
AX-102	6.30E+00	S/TE	AP-108	7.24E+02	E
AX-104	2.06E+01	S	AW-102	3.21E+02	E/TE
AY-101	5.55E+01	S/E	AW-104	7.59E+02	E/TS/TE
AY-102	1.82E+02	S/E	B-110	2.13E+01	E/TE
AZ-101	1.23E+03	S/E	BX-101	1.03E+01	E
AZ-102	4.01E+02	S/E	BX-102	1.19E-01	E/TE
B-111	1.32E+02	S/TE	C-101	4.34E-02	E/TE
B-202	6.85E-01	S	C-105	8.14E+01	E
BX-107	6.98E+01	S	C-201	2.63E-03	E
BY-105	4.63E+01	S/E/TE	S-102	2.00E+01	E/TE
C-103	4.48E-02	S	T-112	2.12E+00	E/TE
C-104	1.73E-01	S/E	TY-101	5.26E+00	E/TE
C-106	1.64E-01	S	TY-102	9.37E+00	E/TS/TE
C-107	4.00E+00	S/E	TY-103	2.71E+01	E/TS
C-108	3.96E-02	S/E	TY-104	8.86E+00	E
C-109	7.50E-03	S	TY-105	4.40E+01	E
C-110	5.77E-02	S	Total	6.51E+03	
C-112	1.29E+01	S/TS			
C-202	2.50E-03	S/E			
C-203	2.32E-03	S/E			
C-204	3.18E-03	S/E			
S-101	1.06E+02	S/E/TE			
S-104	5.12E+01	S/TE			
S-112	1.37E-01	S/E			
SY-101	2.98E+02	S/E			
SY-102	2.44E+02	S/E			
SY-103	1.01E+03	S/E			
T-102	7.13E+00	S/TE			
T-104	9.74E-01	S/E			
T-105	3.50E+01	S/TE			
T-107	4.83E+01	S			
T-111	1.66E+01	S			
TY-106	1.08E+00	S			
U-107	2.35E+02	S/TS/TE			
U-110	1.10E+01	S/TS			
U-201	1.22E-01	S			
U-202	1.10E-01	S			
U-203	9.94E-02	S			
U-204	3.56E-02	S			
Total	1.00E+04				

It is realistic to assume that ^{99}Tc in the supernate and drainable liquids within the saltcake and sludge are soluble forms/species of Tc. It is also probable that some of the ^{99}Tc in saltcake that was not differentiated between drainable liquid and saltcake solids, and even the saltcake solids for tanks where the split between drainable saltcake liquid and saltcake solids is available, would be solubilized during retrieval operations, which in general use some water. Given this assumption (that supernate, drainable liquids, and saltcake categorized Tc are soluble), the split between soluble and not soluble ^{99}Tc is shown in Table 4.8, based on data in Table 4.5 and Appendix A. Thus, the values of "soluble ^{99}Tc" shown in row 6 of Table 4.8 are simplistic estimates of the Ci and percentages of ^{99}Tc that would be readily water-soluble in the retrieved tank wastes sent to the WTP Pretreatment (PT) Facility, prior to being further processed for vitrification or sent to supplemental waste solidification facilities. The values in row 10 of Table 4.8 for "insoluble" assume that all the ^{99}Tc in saltcake (undifferentiated and/or solids) is soluble, but that any the ^{99}Tc in the sludge solids would not be soluble prior to any pretreatment manipulations. Thus, the estimated total ^{99}Tc that is readily water-soluble in the 177 storage tanks today ranges from greater than 47% to likely less than 93%.

Table 4.8. Potential Split of Soluble versus Non-soluble ^{99}Tc in Hanford Storage Tanks Before Pretreatment

Tank "Phases" or Categories	SST ^{99}Tc (Ci)	% of SSTs	% All 177 Tanks	DST ^{99}Tc (Ci)	% DSTs	% All 177 Tanks	% of Total ^{99}Tc Inventory
Supernatant	3.28E+01	0.3%	0.1%	1.09E+04	72.3%	41.1%	
Saltcake (Liquid & Solid)	4.60E+01	0.4%	0.2%	3.28E+03	21.8%	12.4%	
Saltcake Interstitial Liquid	1.50E+03	13.1%	5.7%	5.18E+01	0.3%	0.2%	
Saltcake Solids	8.62E+03	75.3%	32.5%	1.57E+02	1.0%	0.6%	
"Soluble"	**1.02E+04**	**89.1%**	**38.4%**	**1.44E+04**	**95.5%**	**54.2%**	**92.7%**
Sludge (Liquid & Solid)	1.15E+03	10.0%	4.3%	4.14E+02	2.7%	1.6%	
Sludge Solids	1.02E+02	0.9%	0.4%	2.10E+02	1.4%	0.8%	
Sludge Interstitial Liquid	0.00E+00	0.0%	0.0%	6.16E+01	0.4%	0.2%	
"Insoluble"	**1.25E+03**	**10.9%**	**4.7%**	**6.86E+02**	**4.5%**	**2.6%**	**7.3%**
Sum of all categories (Ci) or %	**1.15E+04**	**100.0%**	**43.2%**	**1.51E+04**	**100.0%**	**56.8%**	

Total ^{99}Tc in 177 tanks (Ci) = **2.65E+04.**
Rounding errors occur in the last significant figure in the sums.

There is some water solubility data available on ^{99}Tc for leach tests on two archived sludges from BX-101 SST and AY-102 DST (Krupka et al. 2004; Lindberg and Deutsch 2003). The focus of these reports was to generate data that could be used to address the post-tank-farm-closure fate of contaminants left in residual sludge. These water solubility results should not be confused with sludge washing solubility within the PT Facility. These two reports show that ~25% of the ^{99}Tc in the AY-102 sludge was leached in water over a 30-day period and 100% of the ^{99}Tc in the BX-101 sludge was water leached in the same period. More ^{99}Tc water leach studies have been done on residual sludge left in tanks that have been retrieved. The retrieved tanks for which sludge has been studied are C-103, C-106, C-108, C-202, C-203, and S-112. A detailed listing of available reports and journal articles describing the studies on residual sludges after retrieval is found in Appendix B. Retrieving the waste from the tanks with water or an oxalic acid solution (as was done for tank C-106) removes the readily soluble solids, leaving behind relatively insoluble sludge solid phases. In general, all the studies listed in Appendix B suggest that the percentage of Tc that dissolved from residual sludge from tanks in C tank farm ranged from approximately 6% to 10%, and for the S-112 residual sludge 17% of the ^{99}Tc was readily water leachable. Solid-phase characterization results indicate that the recalcitrant forms (non-water leachable ^{99}Tc) are associated with iron oxides. X-ray absorption near edge structure (XANES) analysis of Tc in one C tank farm residual sludge indicates that the ^{99}Tc (and other generally mobile contaminants such as chromate) occur in Fe oxide particles as their lower, less-soluble oxidation state [Tc(IV)]. The form of these generally mobile contaminants, ^{99}Tc included, in the residual (post-retrieval) sludge is likely as lower valence oxides or hydroxides incorporated within the structure of the Fe oxide. The key useful information generated by these residual sludge studies is that ^{99}Tc in sludge appears to be found as co-precipitates with ferric oxides that can be readily dissolved by the caustic sludge washing process that will be used in the PT Facility. Recall that soluble waste will be sent to the LAW portion of WTP for vitrification as LAW glass and/or some other supplemental waste form.

Therefore, based on the water leaching tests on residual tank sludge, it appears that the ^{99}Tc in some tank sludges is readily solubilized in water and thus the estimate of 93% of the ^{99}Tc in Table 4.8 may be low. Further, it is known that within the PT Facility vigorous sludge washing with strong caustic will solubilize almost all the ^{99}Tc (see for example Geeting et al. 2003).

4.3 ^{99}Tc Speciation in Hanford Tanks

The TWINS database and BBI protocol do not track ^{99}Tc speciation in the various materials (supernate, saltcake, and sludge) within the storage tanks. Further, the HDW model that estimates the fate of ^{99}Tc as it is released from the dissolved fuel elements and cladding during the reprocessing of irradiated fuel makes no attempts to speciate the ^{99}Tc. The aforementioned companion document (Rapko 2014) and an earlier report (Rapko et al. 2013a) introduce the ^{99}Tc speciation issue with emphasis on the unexpected finding of soluble n-Tc species in supernate within DSTs. Both Rapko reports discuss the importance of the n-Tc species after waste retrieval and most of the available ^{99}Tc speciation data. In this subsection we will briefly summarize the work that discovered the n-Tc species and subsequent studies that confirm n-Tc species do exist in DST supernate. By the end of this subsection 4.3 it will be concluded that the identity of the n-Tc species has not yet been established but based on detailed laboratory synthesis studies there is evidence that plausible candidates are all Tc(I) moieties likely associated with carbon monoxide or nitrous oxide that further complex with hydroxide, inorganic anions, or dissolved organic anions such as gluconate.

4.3.1 Précis on Discovery and Confirmation of Existence of n-Tc Species in DST Supernates

When ^{99}Tc removal testing from Hanford tank waste supernates began in the mid-1990s, it soon became apparent that a fraction of the soluble Tc, variable from tank waste supernate to tank waste supernate, was not being captured by materials generally considered to be efficient at pertechnetate removal from high-sodium, high-nitrate solutions. Three pertechnetate “ion exchange” materials that received early attention were ABEC-2000 and Reillex HPQ, anion exchange resins studied primarily at LANL and PNNL, and SuperLig-639, a material that extracts the sodium/potassium pertechnetate ion pair, studied primarily at SRNL and PNNL. Specific references with all the details are cited in Rapko et al. (2013a) and in subsections 4.3.2 through 4.3.4. Another comprehensive review of Tc-specific sorption media was performed by Duncan et al. (2011). Useful information on the characteristics of these three resins, including performance properties for removing Tc from various types of wastes, is tabulated in Table 3-6 within the Duncan et al. (2011) report. The Duncan et al. (2011) report summarizes Tc removal literature for these three “ion exchange” materials in Sections 3.2.1, 3.2.5, and 3.2.7, respectively. These sections within Duncan et al. (2011) cite several more references and give short abstracts on the cited reports’ findings. We mention this to show that much work has been done on Tc removal from many waste streams using these three materials. Duncan et al. (2011) identifies several reports besides citations discussed in this report and Rapko et al. (2013a) that observe the n-Tc species phenomenon. Duncan et al. (2011) does not delve into identifying what the n-Tc species may be.

After the initial discovery of the unexpected poor Tc removal by the pertechnetate-specific “ion exchange” media, subsequent work attempted to determine the cause of the unexpected early breakthrough of a portion of the ^{99}Tc. Several possibilities were considered: 1) that it was due to channeling or some other artifact of column testing, 2) that it was due to another soluble component in the tank supernates that was either a beta emitter (for tests that used scintillation counting as the Tc monitoring method or another mass 99 element for tests that used ICP-MS as the detection method), or 3) that it was due to an alkaline-waste-soluble, n-Tc form of Tc. The general conclusion after numerous investigations (a few described next and others discussed below in subsections 4.3.2 through 4.3.4) has been that this phenomenon is due to the third possibility, i.e., that n-Tc form(s) of Tc that do not respond to standard pertechnetate capture technologies exist in some Hanford DST tank waste supernates.

The first relevant report, Blanchard et al. (1996), describes both batch and column testing of two pertechnetate removal resins, ABEC-2000 and Reillex HPQ, using AW-101 tank supernatant. The testing protocol presented in the report is shown in Figure 4.3.

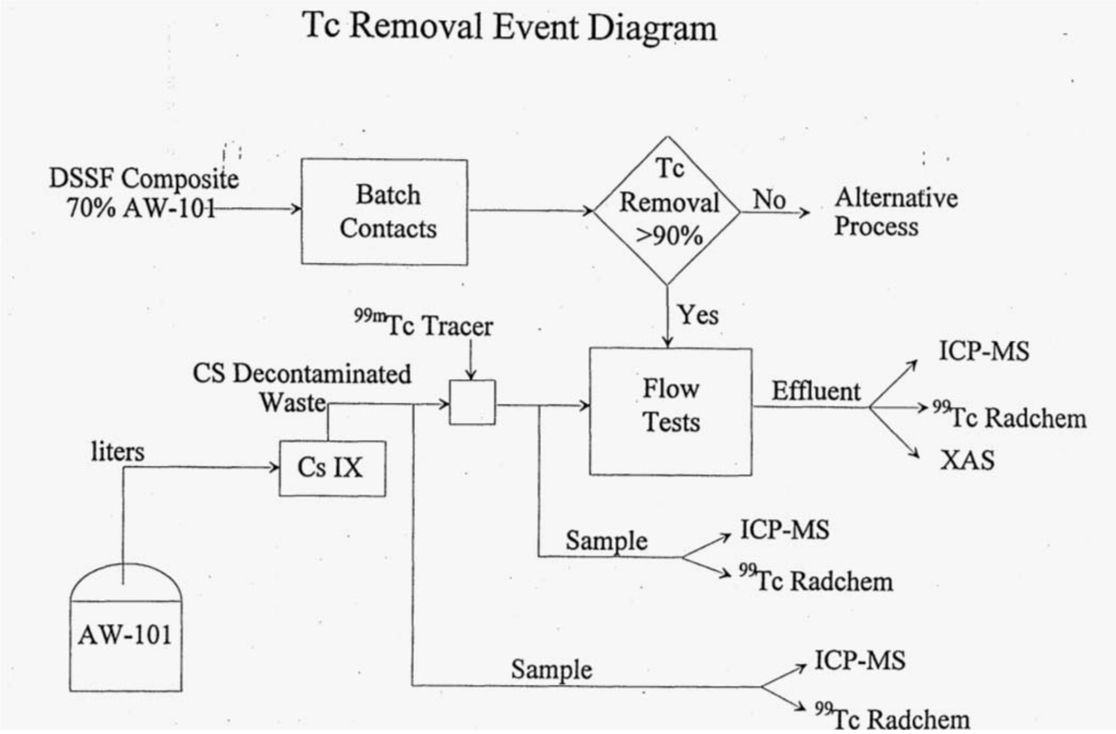

Figure 4.3. Protocol Used to Evaluate ^{99}Tc Removal from DST Liquid Wastes

Note that the AW-101 DST waste was first processed to remove ^{137}Cs and then a pertechnetate radiotracer, ^{99m}Tc, was added solely as pertechnetate to the DST supernate. The use of ^{99m}Tc allows for a distinction between pertechnetate behavior and that of the "bulk" Tc-99 present in the AW-101 supernate to be determined. The ^{99m}Tc tracer is conveniently and accurately determined by high purity germanium (HPGe) gamma spectroscopy with energy resolution that precludes other gamma emitters in the DST waste from confounding the pertechnetate radiotracer analyses. The radiotraced DST liquid waste was then contacted with two pertechnetate-specific ion exchange resins (portrayed in the Figure 4.3 flow diagram by the box named Flow Tests) in separate packed column tests. Two small columns were packed with each of the resins and set up in series, the first column was the "lead" and the second was the "lag" column. The flow tests were then conducted in a fashion that simulates the full-scale process for removal of ^{99}Tc from DST supernate: 1) resin conditioning, 2) resin loading, 3) caustic wash to remove residual feed and prevent the precipitation of $Al(OH)_3$, and 4) ^{99}Tc elution. Figure 4.4 shows a schematic of the flow tests. Small aliquots of the liquid effluent from the lead column and all the effluent from the lag column in each flow test were collected (which were described by the cumulative number of packed column bed volumes) from the start to the end of the Tc loading phase. Influent samples of the DST supernate before and after adding the ^{99m}Tc tracer but before contacting the resins were also collected and Tc measured. All three liquid sample types were then analyzed for ^{99m}Tc and ^{99}Tc content using gamma counting for the ^{99m}Tc, and for ^{99}Tc, ICP-MS and beta counting after vigorous sample oxidation. The ^{99m}Tc and ^{99}Tc concentrations in the effluent as a function of time (measured as cumulative bed volumes) were then compared with the influent concentrations to calculate the typical C/C_0 breakthrough curves. The Figure 4.3 flow chart also shows that some of the effluents from the ion exchange columns were analyzed by synchrotron XAS. The flow chart also shows that before flow through packed column testing was performed, simpler and less costly batch tests were performed to be certain that the two ion exchange resins were capable of removing ^{99}Tc from the DST supernate. The results of the batch sorption tests were positive such that the packed column tests were performed. The results of the packed column tests are shown in Figure 4.5.

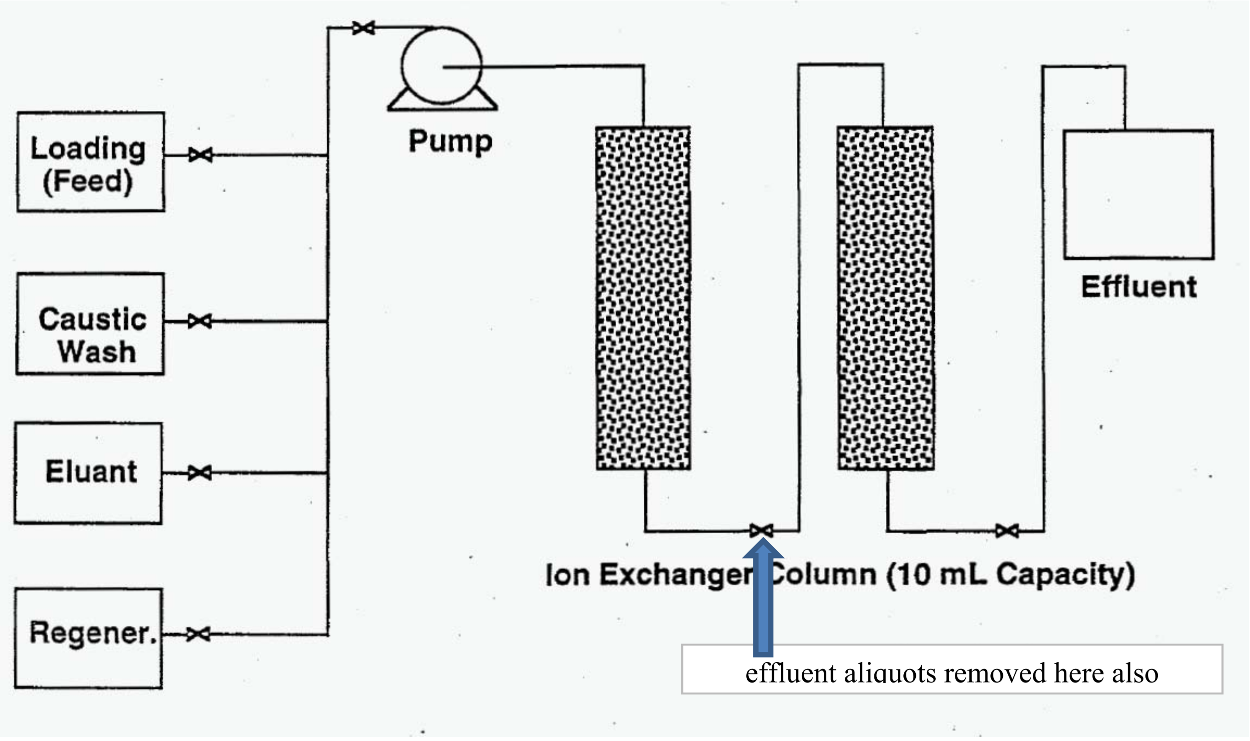

Figure 4.4. Schematic of Tc Removal Flow Tests (modified from Blanchard et al.1996)

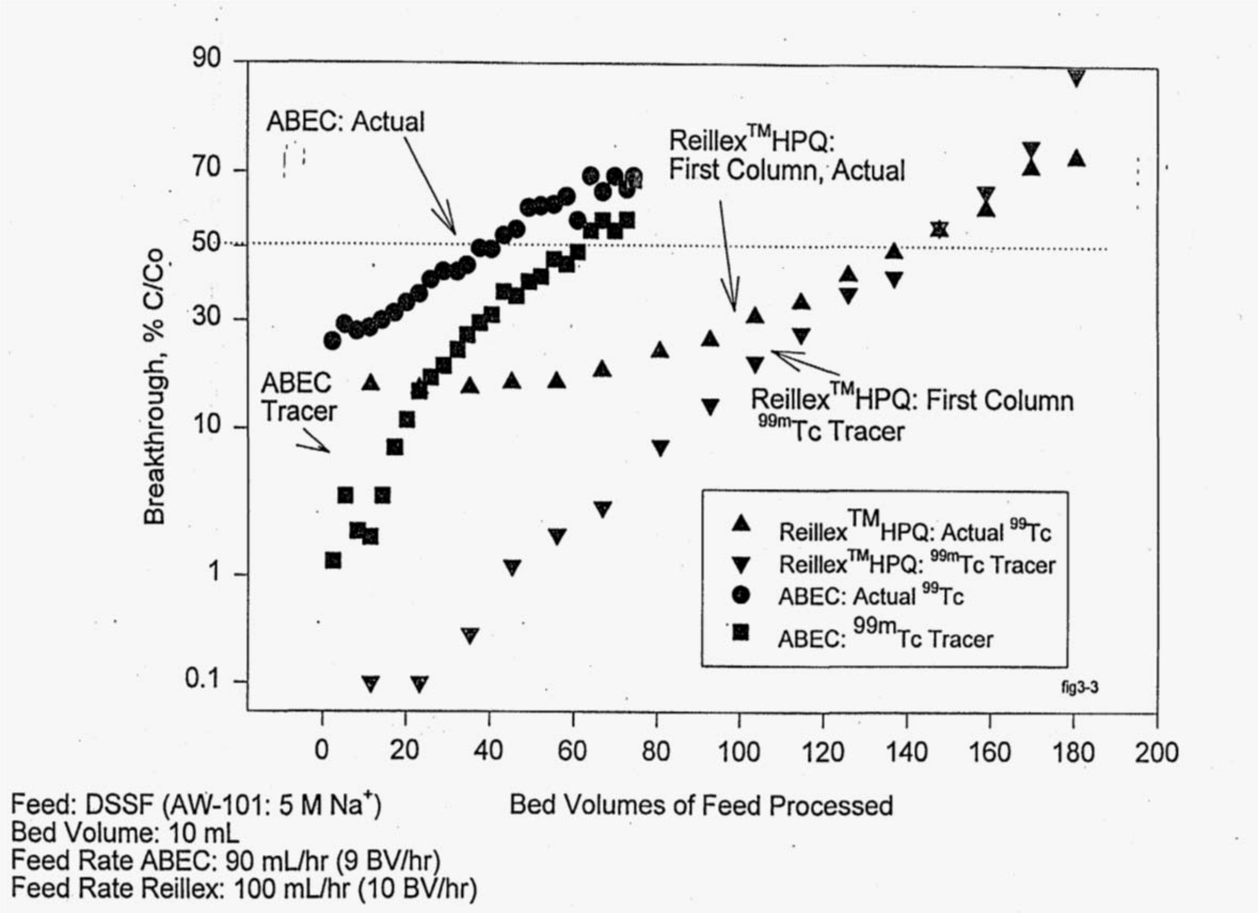

Figure 4.5. Packed Column Test Breakthrough Results (from Blanchard et al. 1996)

Note the dramatically different breakthrough behavior for both resins between pertechnetate, as represented by the ^{99m}Tc and the “bulk” ^{99}Tc in the AW-101 DST effluents from the first (lead) column. The two breakthrough curves for ^{99m}Tc show low normalized (C/C_0) values for the first 60 to 80 bed volumes of cumulative effluent in comparison to the ^{99}Tc breakthrough curves. The two breakthrough curves (^{99m}Tc vs ^{99}Tc) start to converge after about 80 and 140 bed volumes of effluent are collected for the ABEC and Reillex HPQ resins, respectively. These breakthrough results show that both resins remove significantly higher percentages of the pertechnetate (^{99m}Tc) than the “bulk” ^{99}Tc from the DST supernate. Further, the Reillex resin removes more Tc before the resin starts to become loaded with Tc than the ABEC resin. The fact that all four breakthrough curves show the typical progression towards full breakthrough, eliminates the possibility of column mechanical artifacts, such as channeling, from being responsible for the early breakthrough observed for the “bulk” Tc-99. There was a good comparison of the ^{99}Tc concentrations in the effluents between ICP-MS and beta counting results. Figure 4.5 shows only the loading portion of test for the lead column. Other data for the lag column and for the wash and Tc elution phases of the resin testing are found in Blanchard et al. (1996) and further support the presence of n-Tc species in the AW-101 supernate. The XAS results from characterizing the valence state of Tc in selected effluents from the packed columns and the original AW-101 supernate (after ^{137}Cs removal) are discussed in subsection 4.3.4.

Similar early breakthrough of “bulk” ^{99}Tc in DST supernates from several other Hanford tanks has been observed when the resin used is SuperLig-639. For example, Burgeson et al. (2004a) performed a column test using a mixture of AN-102 supernate and C-104 filtrate, wash, and leach solution. In these tests no pertechnetate radio-spike was used but the same two columns in series protocol shown in Figure 4.4 was used. The breakthrough curves for effluent from both the lead and lag columns are shown in Figure 4.6.

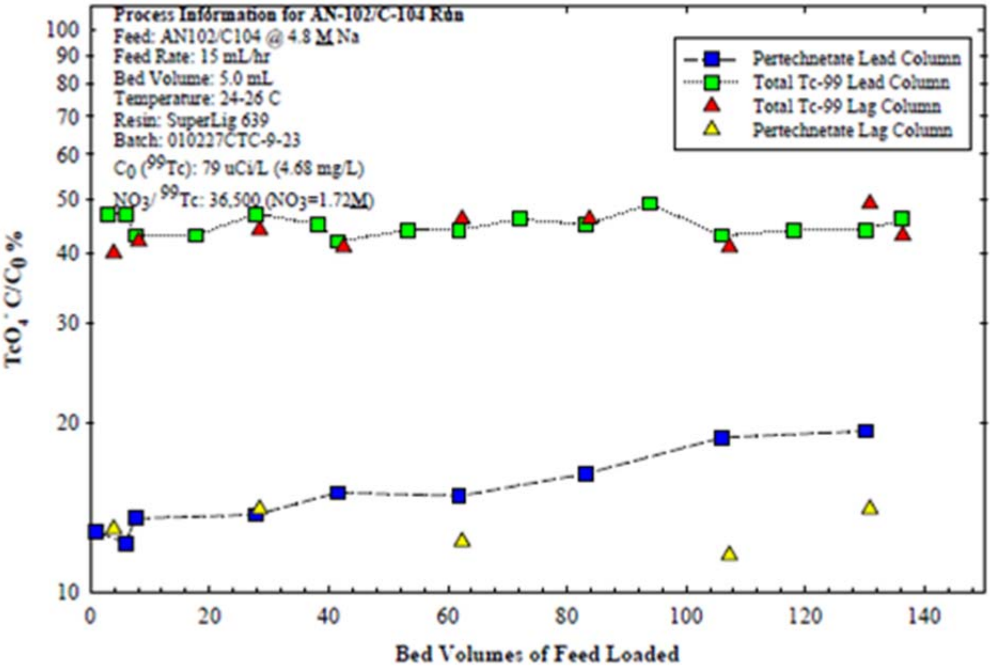

Figure 4.6. Breakthrough Curves for SuperLig 639 Columns Contacted with AN-102 Supernate

In Figure 4.6, from Burgeson et al. (2004a), total Tc was determined by ICP-MS at mass 99 and pertechnetate was determined by Eichrom TEVA resin (pertechnetate-specific) extraction methods for the column effluents followed by liquid scintillation counting. The influent or C_0 value for ^{99}Tc was determined by both ICP-MS and after vigorous oxidation of the influent by liquid scintillation. Both methods gave similar concentrations for Tc in the influent. The large differences between total Tc-99 and pertechnetate breakthrough behavior observed in both the lead and lag column effluents are consistent with the hypothesis that this DST supernate contains a significant amount of a n-Tc species. Figure 4.6 shows only the first portion of the SuperLig 639 resin loading phase. After stopping influent loading and performing resin washing and Tc elution, Burgeson et al. (2004) performed a mass balance calculation. They found excellent recovery (97 to 101%) of the total ^{99}Tc injected into the dual-column test. This suggests that their breakthrough data presented in Figure 4.6 are accurate and that the hypothesis of the presence of n-Tc species in the AN-102 supernate is confirmed.

Additional measurements that support the assignment of an alkaline-soluble, n-Tc source of Tc in various Hanford waste tanks include: the direct measurement by Tc-99 NMR of an increased pertechnetate concentration in the supernatant following exhaustive oxidation by contact with hot concentrated nitric acid/Ce(IV) (Schroeder et al. 1995) and by capture of additional activity by a pertechnetate-selective resin following microwave/persulfate oxidation of four (4) different Hanford tank supernatants (Egorov et al. 2004; Egorov et al. 2012). Other supporting evidence includes very low batch K_d values for the pertechnetate-specific resins such as SuperLig 639 and Reillex HPQ for actual DST supernates as compared to pure pertechnetate tracers (^{95m}Tc or ^{99m}Tc) added to the DST supernates or spiked simulants of similar composition. More discussion on some of these findings follows in subsections 4.3.2 through 4.3.4.

In summary, Table 4.9 adapted from Rapko et al. (2013a) lists the percentages of n-Tc that have been directly measured or inferred from studies of the ^{99}Tc contents in DST supernates. Table 4.9 also lists the methods used to support the hypothesis that n-Tc species are present. Methods include batch and flow-through column tests using pertechnetate-specific resins, pre- and post-vigorous-oxidation Tc analyses of DST supernates, and NMR and XANES direct spectroscopic analyses of DST wastes. In total, researchers at four national labs have published approximately 30 reports or journal articles supporting the presence of n-Tc species in Hanford DST supernates. What remains to be determined is the identity or identities of the n-Tc species and how they impact the processing of retrieved tank waste and the final disposition of treated wastes within the WTP and/or supplemental waste facilities.

Table 4.9. Reported Distribution of n-Tc Tc Present in Various Hanford Tank Supernatants (modified from Rapko et al. 2013a)

Tank	Method	% n-Tc (Max)	% n-Tc (Min)	Reference
AN-102	SL-639 lag breakthrough	70	60	King et al. (2001, 2003)
AN-102	SL-639 column	80	70	Hassan et al. (2000b)
AN-102	SL-639 column	70		King et al. (2000)
AN-102	Multiple SL-639 batch contacts	63		Hassan et al. (2001b, 2002)
AN-102	MP-1 captured TcO_4^- before/after oxidation	57		Egorov et al. (2012)
AN-102	SL-639 column	48		Egorov et al. (2012)
AN-102 /C-104 mix	SL-639 batch K_d	50		Burgeson et al. (2004a)
AN-103	SL-639 lag breakthrough	8	7	King et al. (2001, 2003)
AN-103	SL-639 column	8	3	McCabe et al. (2000)
AN-103	SL-639 column	2.4	2.3	Hassan et al. (2000a)
AN-103	Multiple SL-639 batch contacts	1.6		Hassan et al. (2001b, 2002)
AN-107	XANES fit	62		Blanchard et al. (1997)
AN-107	SL-639 batch K_d	78	75	Kurath et al. (2000)
AN-107	SL-639 column	80		Blanchard et al. (2000b)
AN-107	Fit to batch K_d Reillex HPQ	63	48	Schroeder et al. (1998)
AN-107	Fit to batch K_d Reillex HPQ	66±9		Schroeder and Ashley (2005)
AN-107	MP-1 captured TcO_4^- before/after oxidation	57		Egorov et al. (2004)
AN-107	SL-639 column	50		Egorov et al. (2012)
AP-101	% Tc in feed by inductively coupled plasma mass spectrometry (ICP-MS) (total) vs TcO_4^- by rad separation	13 suspect result		Burgeson et al. (2002)
AP-101	SL-639 column	<0.5		Burgeson et al. (2002)
AP-104	SL-639 column	69		Burgeson et al. (2005)
AP-104	MP-1 captured TcO_4^- before/after oxidation	72		Egorov et al. (2004)
AP-104	SL-639 column	72		Egorov et al. (2012)
AW-101	SL-639 column	0.06	0	Hassan et al. (2003)
AW-101	Reillex HPQ column	15		Blanchard et al. (1996, 2000a)
AW-101	SL-639 batch K_d	2.9		Kurath et al. (2000)

Tank	Method	% n-Tc (Max)	% n-Tc (Min)	Reference
AW-101	% Tc in feed vs SL-639 column effluent	4.5		Hassan et al. (2003)
AZ-101	SL-639 column	0	0	Burgeson et al. (2004b); Egorov et al. (2012)
AZ-102	SL-639 column	0	0	Egorov et al. (2012)
AZ-102	% Tc in feed vs SL-639 column effluent	33		Hassan et al. (2003)
AZ-102	SL-639 column	0.04	0	Hassan et al. (2001a)
AZ-102	Multiple SL-639 batch contacts	<0.1		Hassan et al. (2001b, 2002)
DSSF	Reillex HPQ batch K_d	7		Blanchard et al. (1997)
SY-101	Reillex HPQ batch K_d	53		Blanchard et al. (1997)
SY-101	Fit to batch Kd Reillex HPQ	63		Schroeder et al. (1998) Schroeder et al. (1995)
SY-101	Fit to batch Kd Reillex HPQ &TcO_4^- by nuclear magnetic resonance (NMR)	70	63	Schroeder et al. (2001)
SY-103	Reillex HPQ batch K_d	54		Blanchard et al. (1997)
SY-103	XANES fit	78		Blanchard et al. (1997)
SY-103	Fit to K_d Reillex HPQ	70		Schroeder et al. (1998) Schroeder et al. (1995)
SY-103	Fit to batch K_d Reillex HPQ &TcO_4^- by NMR	70	64	Schroeder et al. (2001)

4.3.2 Speciation Inferred from Ion Selective Resin Tests

In this subsection we provide some more details on using pertechnetate-specific materials to identify and confirm that n-Tc species exist in Hanford DST supernates. Numerous reports describe the use of organic-based resins that preferentially sequester pertechnetate anions out of Hanford liquid waste streams. Most of the studies used pertechnetate-specific resins called SuperLig® 639 (IBC Advanced Technologies, Inc., American Fork, Utah), which is a proprietary product that uses molecular recognition theory to design a substrate highly selective to sequestering the species of interest[1], or Reillex® HPQ (Vertellus Health & Specialty Products LLC, Indianapolis, Indiana 46204). SuperLig 639 functions by extracting the sodium pertechnetate salt-pair out of liquid waste at fairly high sodium concentrations (generally ~4 - 5 M Na). Reillex HPQ resin is a co-polymer of divinylbenzene and 4-vinylpyridine that has been subsequently methylated at the pyridine nitrogen to give pyridinium [$-C_5H_4N(CH_3)^+$] strong base anionic sites (Figure 4.7). The pyridinium functionality of Reillex HPQ resin is unique compared to most other strong base anion resins. Reillex HPQ is ~25% cross-linked and 70% methylated at the pyridine

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[1] SuperLig proprietary products are solid phase particles (~0.5 mm) generally based on silica gel or polymer substrates to which a selective ligand has been chemically attached. The choice of ligand uses Molecular Recognition Technology (MRT)—a highly selective, non-ion-exchange process, using specially designed organic chelating agents or ligands. The MRT process utilizes supramolecular, “lock and key” or “host guest” chemistry as the basis for its high selectivity. Feed solution is passed through a column packed with the SuperLig particles and the target species is removed selectively from the solution. SuperLig products are designed to bind selectively with ions based on multiple parameters such as size, coordination chemistry, and geometry. In contrast, separation methods used in conventional ion exchange, solvent extraction, or precipitation processes generally recognize differences between ions based only on a single parameter (e.g., charge, solubility, size). SuperLig products bind ions selectively even when the ions are present in highly acidic or basic solution and/or in solutions containing high concentrations of competing ions. High species selectivity, high binding factors, and rapid reaction kinetics result in very efficient separations. The elution chemistry for SuperLig products facilitates a wide choice of eluent formulations to ensure compatibility with particular separation requirements. Since small volumes of eluate are used, highly concentrated eluent solutions are produced.

nitrogen to give pyridinium anionic sites. The total exchange capacity provided by the manufacturer for 30-60 mesh resins in the chloride form is 4.6 meq Cl/g (dry) and 1.2 meq Cl/mL (wet). Reillex HPQ, compared to other resins, has superior stability to radiolysis and nitric acid.

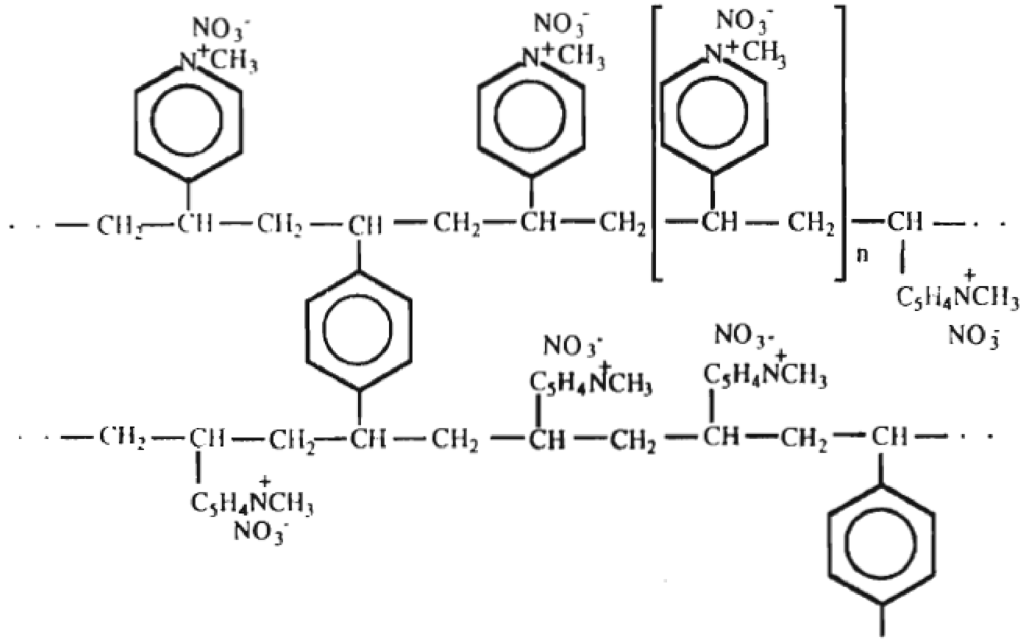

Figure 4.7. Structure of Reillex HPQ Anion Exchange Resin (from Schroeder et al. 1995)

As briefly described in subsection 4.3.1, while processing Hanford liquid wastes (generally supernate from various DSTs) through columns packed with SuperLig 639, it was observed for some of the supernates that there was an immediate breakthrough of some of the ^{99}Tc present. For other Hanford DST supernates (e.g., AZ-101) and Hanford liquid waste simulants, which had been spiked with known amounts of ^{99}Tc and/or ^{95m}Tc, both in the pertechnetate form, the SuperLig 639 resin retained all of the Tc for tens to a few hundred pore volumes of effluent before any Tc was found to break through. A second methodology, batch sorption tests, has been used to identify the presence of n-Tc . For the batch tests, known amounts of SuperLig 639 or another pertechnetate-specific resin, (e.g., Reillex HPQ) were contacted with actual Hanford DST supernatant liquids, usually at 100:1 (liquid volume to resin volume ratio) for 4-day contacts. The batch slurry was then separated by centrifugation and/or filtration and the concentration of ^{99}Tc in the effluent was compared to the concentration in the influent using the traditional K_d construct. The Tc K_d values for DST supernatants that were suspected to contain n-Tc species were significantly lower than the K_d values for DST supernatants that contained only the pertechnetate species and for simulants that had been spiked with known amounts of pertechnetate, often using the convenient gamma-emitting ^{95m}Tc isotope.

The best hypothesis that explains the immediate breakthrough of some Tc from the DST supernatant in the packed SuperLig 639 columns and the low K_d values found in the batch sorption tests is that soluble species of ^{99}Tc were present in the DST supernates that were not pertechnetate. Because all of the DST supernates that exhibited the early breakthrough of ^{99}Tc also contained significant concentrations of dissolved organic matter, ^{99}Tc-organic complexes were thought to cause the unexpected Tc behavior. Table 4.9 as mentioned originates in Rapko et al. (2013a) and has been updated herein with a few more values found in additional reports. Rapko et al. (2013a) also mention a correlation between the percentage of n-Tc and DST supernate calculated dose (mostly ^{137}Cs content). But the correlation is negative (high n-Tc correlates with low dose or ^{137}Cs), which is counterintuitive if radiolysis influences the formation of the dissolved n-Tc content in the Hanford storage tanks.

4.3.3 Additional Studies that Probe Tc Speciation

The second method used to help identify the speciation of the n-Tc species found in DST supernates relied on carefully planned laboratory studies that attempted to create n-Tc compounds using irradiation or after creation to use size exclusion chromatography to isolate the species. Shuh et al. (2001) and Lukens et al. (2002) performed radiolysis experiments (using a 600 Ci ^{60}Co irradiation facility) on 0.002 M $^{99}TcO_4^-$ spiked 2M NaOH solutions that contained various dissolved organics at concentrations between 0.1 and 0.5 M. The organic molecules studied individually were formate, glycolate, glyoxylate, ethylene glycol, ethanol, EDTA, nitrilotriacetate (NTA), iminodiacetate (IDA), diphenylphosphate, dibutylphosphate, citrate, acetate, and formaldehyde. The solutions were irradiated for up to 16 hours (absorbed doses varied between 3.4 and 19.6 kilo-grays or 10^5 rads).

The major findings of Shuh et al. (2001) and Lukens et al. (2002) follow. Reduction of TcO_4^- requires the presence of organic materials to scavenge the oxidizing O^- radical produced during radiolysis. Based on Lukens et al. (2001), radiolytic reduction of TcO_4^- is also efficient in the presence of excess nitrate, which is a major soluble component in Hanford supernates. In the presence of most organic molecules, the reduced Tc species was an insoluble solid, $TcO_2 \bullet xH_2O$ (identified by extended X-ray absorption fine structure [EXAFS] and electron paramagnetic resonance [EPR]). However, when the organic molecules are diols or polyols,[1] soluble Tc radiolysis products are observed. The polyols studied that formed soluble Tc radiolysis products were ethylene glycol, glyoxylate, and formaldehyde. These soluble Tc radiolysis products were found to be soluble Tc(IV) diolates (see next paragraph). In addition, none of the organic compounds that yielded the insoluble $TcO_2 \bullet xH_2O$ is a diol. The soluble Tc(IV) compounds were prone to re-oxidation to TcO_4^- over a period of days to one week in the presence of air at the caustic pH in the 2 M NaOH background electrolyte. The insoluble $TcO_2 \bullet xH_2O$ also oxidizes to TcO_4^-, but over a longer period.

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[1] A molecule with two hydroxyl groups is a **diol**. A **polyol** is an alcohol containing multiple hydroxyl groups.

Shuh et al. (2001) and Lukens et al. (2002) used UV-Visible spectra and EXAFS on the soluble Tc-organic complexes and found these radiolysis-induced Tc-diol complexes are Tc(IV) with binuclear $Tc_2(\mu\text{-}O)_2$ core structures similar to a well-known $(H_2EDTA)_2Tc_2(\mu\text{-}O)_2$.[1] However, relatively high Tc concentrations (1 mM) were necessary to obtain the EXAFS spectrum. At lower Tc concentrations, it is likely that monomeric alkoxide complexes would be formed rather than binuclear complexes. The authors also caveat their findings in that few of the polyol ligands used in the radiolysis tests are present in Hanford tank waste in such high concentrations as were used in the radiolysis experiments to account for the formation of the soluble n-Tc species. The one potential alkoxide ligand present in large enough quantities in high organic DSTs is gluconate.

These radiolysis experiments support the finding of reduced soluble Tc species in DST supernates because the tanks contain a myriad of molecules, and thermal and radiation vectors that in combination could produce unexpected chemical changes in the Tc species present. Shuh et al. (2001) and Lukens et al. (2002) also opine that, given the air sensitivity of the reduced Tc species formed in the radiolysis tests, archived supernate samples from the Hanford tanks will not accurately reflect the proportion of reduced Tc species. Thus, experiments to measure accurately the proportion of reduced Tc species in a given tank must be carried out on freshly sampled material. Outside the tank's radiolysis environment, the reduced Tc species could be removed, albeit slowly, by aeration.

Another interesting study, Bernard et al. (2001), showed that a soluble n-Tc species could be produced by heating (to 65 °C for 24 hr) a DST caustic supernate simulant containing ^{99}Tc spiked as pertechnetate, 0.05 M gluconate, and soluble noble metal (Rh, Ru, and Pd) catalysts. Other organic complexants also promote reduction of pertechnetate, but the product is insoluble black precipitate suspected to be $TcO_2 \bullet xH_2O$. When using gluconate as the organic, more than 99% of the Tc remained in solution, but only 76% of the soluble ^{99}Tc was extracted by the pertechnetate-sequestering Reillex HPQ resin. Thus, some (~23%) unidentified soluble n-Tc species was apparently formed without the need for radiolysis processes. Bernard et al. (2001) also claim that the noble metal catalysts are an essential component of the pertechnetate reduction process in the presence of organic complexants—at least for a thermally driven system. When no noble metal catalysts were present, the DST caustic simulant inoculated with various organic complexants did not show pertechnetate reduction over a 4-day period at 65 °C. Inoculating the batch reactors containing DST simulant, but with no noble metal catalysts and no organic compounds, with hydrogen gas also did not show pertechnetate reduction, but adding any or all three noble metal catalysts (again with no organic complexants) reduced 84% to 99% of the pertechnetate to the insoluble black precipitate after 1 day at 65 °C. The need for noble metal catalysts to be present

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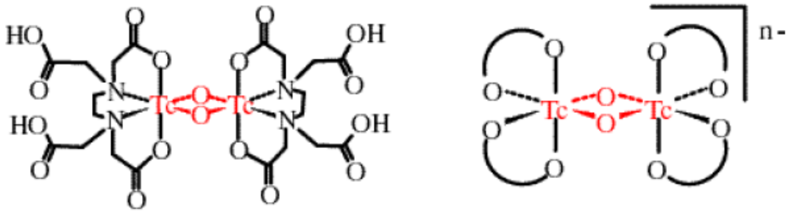
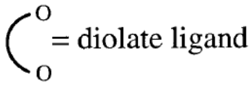
[1] Compounds with $Tc_2(\mu\text{-}O)_2$ cores are shown in the figure (modified from Shuh et al. 2001)



Left: $(H_2EDTA)_2Tc_2(\mu\text{-}O)_2$. Right: proposed structure of the product of radiolysis of TcO_4^- in alkaline solutions containing diols. The diolate ligand is represented by the curved line.



along with organics to effect TcO_4^- reduction was peculiar to this study. Shuh et al. (2001) and Lukens et al. (2002), as mentioned in the previous paragraphs, effected the TcO_4^- reduction via ^{60}Co radiolysis with no noble metal catalysts present.

Schroeder and Ashley (2005) suggest that some of the soluble ^{99}Tc in AN-107 supernate is cationic because it is removed during sample cleanup procedures to remove ^{90}Sr and other cations prior to attempting to separate Tc species using size exclusion chromatography through a Sephadex® G-10 column (registered trademark of GE Healthcare). G10 separates molecules with molecular weights <700 daltons based on the molecules' sizes. Preliminary experiments determined that $^{99}TcO_4^-$, whether dissolved in water or in a solution of 1.50 M $NaNO_3$/1.00 M NaOH, is not retained by G-10 columns. That is, pertechnetate elutes[1] from the G-10 column but AN-107 supernate (after removal of ^{90}Sr and other cationic beta emitters) had 25% of its Tc retained by the G-10 column. Because size exclusion chromatography media are not supposed to interact strongly with the solutes being separated, this strong interaction (retention within the column) confounds simple interpretation. However, based on other studies that showed AN-107 supernate contains ~60% n-Tc and 40% TcO_4^-, Schroeder and Ashley assumed that if the pertechnetate (~40% of the total ^{99}Tc in AN-107 supernate) is not held up in the G-10 column, then approximately 42% (25/60) of the n-Tc is retained in the G-10 column. Thus, G-10 beads have a strong interaction with some of the n-Tc species but not pertechnetate. Although not stated in Schroeder and Ashley 2005, one might conclude their results suggest the presence of at least two n-Tc species, one (~58%) that elutes and one that is retained (~42%) within the column. Schroeder and Ashley (2005) conclude that size exclusion chromatography (using G-10) offers the possibility of separating some of the n-Tc species from each other and from pertechnetate, and that using a different eluent (water was used in this study) such as ethylene glycol that could compete with the dextran alcohol functionalities in the G-10 beads might improve the elution of all the ^{99}Tc from the G-10 column. In a second study, the fate of ^{99}Tc in the AN-107 supernate was followed as it was pumped through two cation exchange columns (Dowex AG50WX8) and then a G-10 size exclusion column. This study determined that 10% to 20% of the total ^{99}Tc in the AN-107 supernate was retained in the cation exchange column, 53% of the ^{99}Tc eluted in two bands from the G-10 column, and 30% of the ^{99}Tc remained stuck in the G-10 column. Combining results from the various column studies, the authors hypothesized that one of the ^{99}Tc species in the AN-107 supernate might be fac-$Tc(CO)_3(H_2O)_3^+$, which would be retained on the cation exchange resin.

A companion document (Ashley et al. 2004) reports more details on the size exclusion chromatography studies. Ashley et al. (2004) studied the performance of two separation media, Sephadex G-10 and BioGel P-2[2] (Bio-Rad Inc., Hercules, California) and found that the ^{99}Tc species, including pertechnetate, in AN-107 supernate do interact with both size exclusion separation media, complicating the interpretation of the various effluent curves. Further, simple water solutions spiked with either $^{95m}TcO_4^-$ or ^{99}Tc-(gluconate) complex interacted with the BioGel P-2 packed column such that the elution volumes of both were much greater than they should be. However, the two Tc species could be resolved

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[1] Unfortunately, the authors do not provide details on when (how many elution volumes of effluent are collected before and after) the $^{99}TcO_4^-$ elutes from the G-10 column. The elution volume (V_e), void volume (V_0), and interstitial volume (V_i) of the G-10 packed column are necessary details to understand whether a solute species elutes simply as a function of size or whether there are unwanted chemical interactions between the solute and the column beads.

[2] P-2 is a polyacrylamide gel copolymer of acrylamide and N, N'-methylene-bis-acrylamide with a fractionation range 100 to 1800 daltons.

(i.e., they eluted at different elution volumes). These two studies relied on radiocounting (NaI for gross gamma and liquid scintillation counting [LSC] for ^{99}Tc) of the various effluents from the Dowex cation exchange columns and size exclusion columns. The LSC method is not exclusively separating ^{99}Tc from all other potential beta emitters in the eluents from AN-107 supernate or for that matter $^{95m}TcO_4^-$ added to some of the mixed simple solutions. Thus, the radiocounting method also complicates definitive identification of the Tc species in the size exclusion effluents. Future studies using size exclusion chromatography should use ICP-MS to measure total ^{99}Tc and NMR to differentiate pertechnetate from other n-Tc forms of Tc.

These suggested changes in ^{99}Tc measurement techniques should significantly improve the value of using size exclusion chromatography as a tool to identify ^{99}Tc species in DST supernates as long as the various eluents contain high enough ^{99}Tc total concentrations. It would appear that current ^{99}Tc detection limits for ICP-MS and NMR should be acceptable because they are comparable to or better than LSC.

4.3.4 Spectroscopic Studies Used to Determine Tc Speciation

A third method used to explore Tc speciation uses spectroscopic measurements on both carefully synthesized pure Tc compounds and actual DST supernates using several instruments and techniques, including UV-Vis, NMR, and synchrotron-based X-ray absorption spectroscopy (XAS), both XANES and EXAFS. A number of studies have been reported where ^{99}Tc-organic complexes have been synthesized and characterized in great detail in attempts to identify the ^{99}Tc speciation of the soluble n-Tc species found in DST supernates. Schroeder et al. (2002, 2004), and Ashely et al. (2004) attempted to synthesize and characterize Tc complexes with the following ligands: EDTA, ethylenediamine-N,N'-diacetic acid (EDDA), IDA, nitrilotriacetic acid (NTA), diethylenetriaminepentaacetic acid (DTPA), N-(2-hydroxyethyl)ethylenediaminetriacetic acid (HEDTA), oxalate, glycolate, citrate, gluconate, ethhylenediamine (en), diethylenetriamine (detn), and triethylenetetraamine (tetn). These preparations proved quite difficult, as the products were often not clean and required extensive chromatographic separation. XAS data that the authors were able to obtain indicated some similarity to structures in the literature, but often their synthesized products were more complex. The synthesized complexes also decomposed or oxidized when placed in caustic. The one exception to the above observations was a soluble Tc-gluconate complex that does form in base, but also will change over time to some other species.

Rapko et al. (2013b) were able to synthesize $Tc(CO)_3(H_2O)_3^+$ and $[Tc(CO)_3 \text{ gluconate}]^{2-}$ separately in alkaline solution and measured their NMR, FTIR, and UV spectra. Other key results of this work included the following:

- Many of the current synthesis procedures are not suitable for making high-milligram to gram quantities of these compounds needed for detailed studies.
- $[Tc(CO)_3(H_2O)_3]^+$ undergoes extensive hydrolysis at pH values above 6.8; thus, in tank-relevant conditions in the absence of organic complexing agents, the hydrolysis species $[Tc(CO)_3(OH)_n(H_2O)_m]^{1-n}$ will be formed.
- These tricarbonyl hydrolysis species complicate and interfere with complexation with inorganic and organic ligands in the alkaline tank environment.

Direct spectroscopic synchrotron XAS has been used in attempts to identify the ^{99}Tc species in DST supernates. The first attempts are documented by Blanchard et al. (1996, 1997), wherein XANES spectra were obtained for five DST supernates taken from three tanks. XANES is a technique that probes electronic transitions from an atomic core level to final states in the energy region of 50 - 100 eV above the selected atomic core level ionization potential. TcO_4^- molecules exhibit a distinct hump in their XANES spectra below the Tc core level ionization potential, whereas lower-valence-state Tc molecules do not exhibit such a distinct hump. XANES studies of five DST supernates showed that in four of the samples most of the ^{99}Tc present did not show the characteristic pre-edge hump for pertechnetate. The four DST supernates that did not show a dominance of pertechnetate using XANES analysis were from tanks AN-107 (as received), SY-101 (after contacting the Reillex HPQ resin that selectively removes pertechnetate), SY-103 (after removal of cations [^{90}Sr]), and SY-103 (after contacting the Reillex HPQ resin that selectively removes pertechnetate).

Based on a comparison of the XANES spectra for the DST supernates to two Tc standards (Tc(0) metal and Tc(VII) pertechnetate solution) that had known oxidation states, Blanchard et al. (1997) felt that the ^{99}Tc in four of the five DST supernates was dominated by soluble Tc(IV) species, likely some Tc(IV)-organic species. Blanchard et al. (1997) properly warn that an XAS spectrum is the sum of the XAS spectra from all the different species of a given element that are present in a sample. If more than one species is present, care must be taken to account for this in analyzing the data, and having several known standards with known oxidation states is important in order to compare with the spectrum for an unknown. As mentioned previously, Blanchard et al. (1997) only had spectra for two known standards: Tc metal, Tc(0), and pertechnetate, Tc(VII), so the interpretation was tentative and would have been improved with having a Tc(IV) standard.

The Tc XANES for the “as received” AN-107 sample and the SY-103 sample after removal of ^{90}Sr are reproduced in Figure 4.8. The spectra are very similar. The appearance of small shoulders for these two samples at the TcO_4^- pre-edge peak indicates the presence of TcO_4^-. The XANES spectra for DST supernate samples that were first contacted with Reillex HPQ resin are also nearly identical to each other and lack any sign of the pertechnetate-characteristic hump. Of course, the two supernates that were contacted first with the Reillex HPQ resin would be expected **not** to contain significant concentrations of pertechnetate. The similarity in XANES spectra indicates that the Tc species left in solution after removal of the TcO_4^- is the same for the two different tank supernates. Blanchard et al. (1997) also opine that if, following removal of the TcO_4^- using Reillex HPQ resin, pertechnetate is reformed very slowly (i.e., reestablishment of equilibrium is kinetically limited), a linear combination of the Reillex HPQ resin-contacted SY-103 XANES spectra and the TcO_4^- standard XANES spectra should fit the SY-103 XANES spectra and would indicate the difference in the amount of TcO_4^- between the two samples. Figure 4.9 shows this fit. The fit suggests that 22% TcO_4^- is present in the SY103 supernate that had ^{90}Sr removed prior to analyses. A similar fit for as-received AN-107 indicates that there is 38% TcO_4^- present in AN-107 and 64% n-Tc species. Blanchard et al. (1997) felt that the n-Tc species had ^{99}Tc present in the Tc(IV) oxidation state.

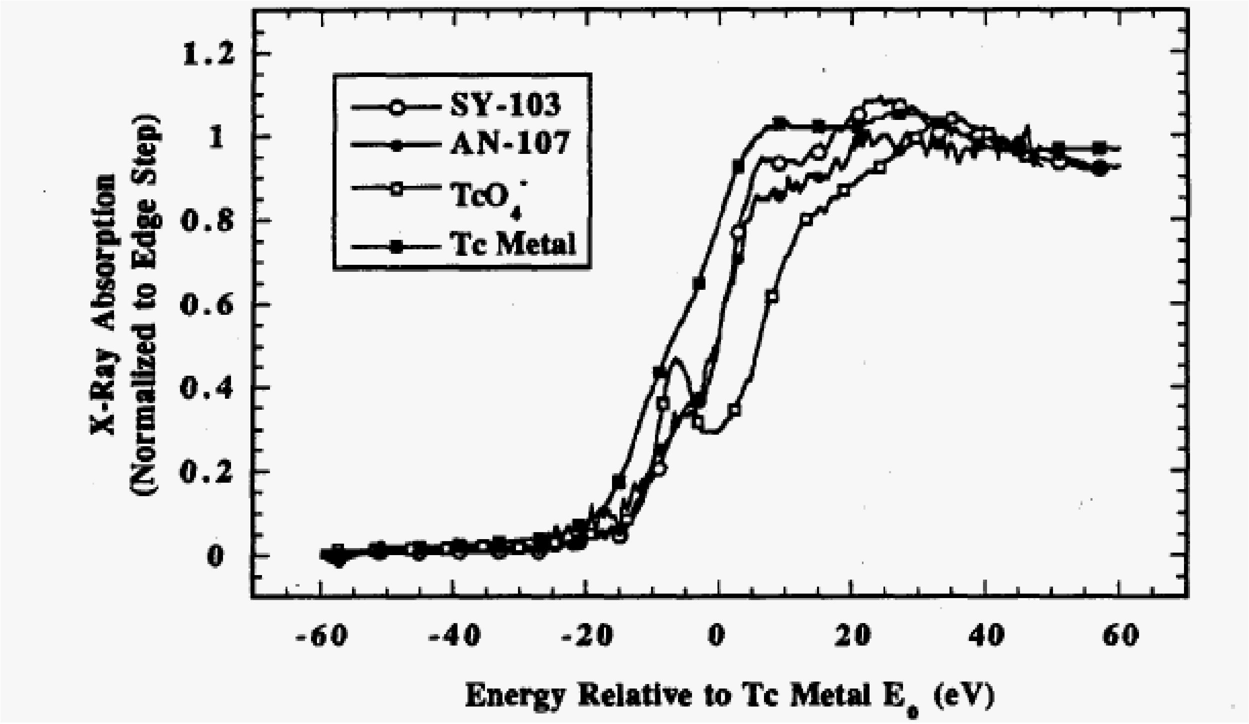

Figure 4.8. Comparison of XANES Spectra for AN-107 and SY-103 Supernates with Two Tc Standards

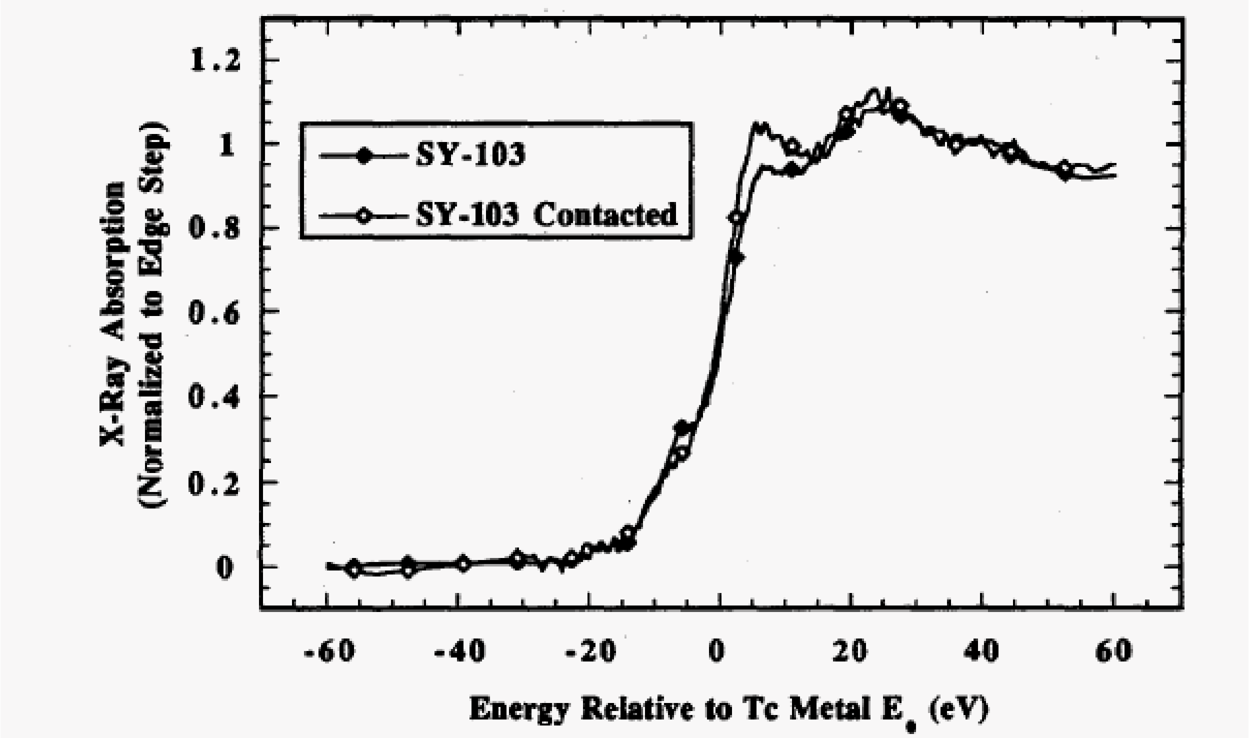

Figure 4.9. Comparison of XANES Spectra for SY-103 Supernates (contacted refers to removal of pertechnetate prior to analysis)

The AW-101 supernate had a XANES spectrum with a significant pre-edge hump, similar to the pertechnetate standard XANES spectra, suggesting a predominance of pertechnetate in the supernate but also a small amount of n-Tc (see Figure 4.10). Another XANES spectra comparison between SY-103 supernate that had been stored for an additional 4 months before analyses showed a distinct in-growth of pertechnetate in comparison to the XANES spectra taken earlier. This shows that once a DST supernate sample is removed from the high-intensity radiation environment (along with potentially higher temperatures) inside the tank, some of the n-Tc is re-oxidized to pertechnetate. This presents another challenge to identifying the speciation of the n-Tc within the DSTs, mainly because the time between obtaining samples and completion of all processing and detailed characterization and analysis is often several months.

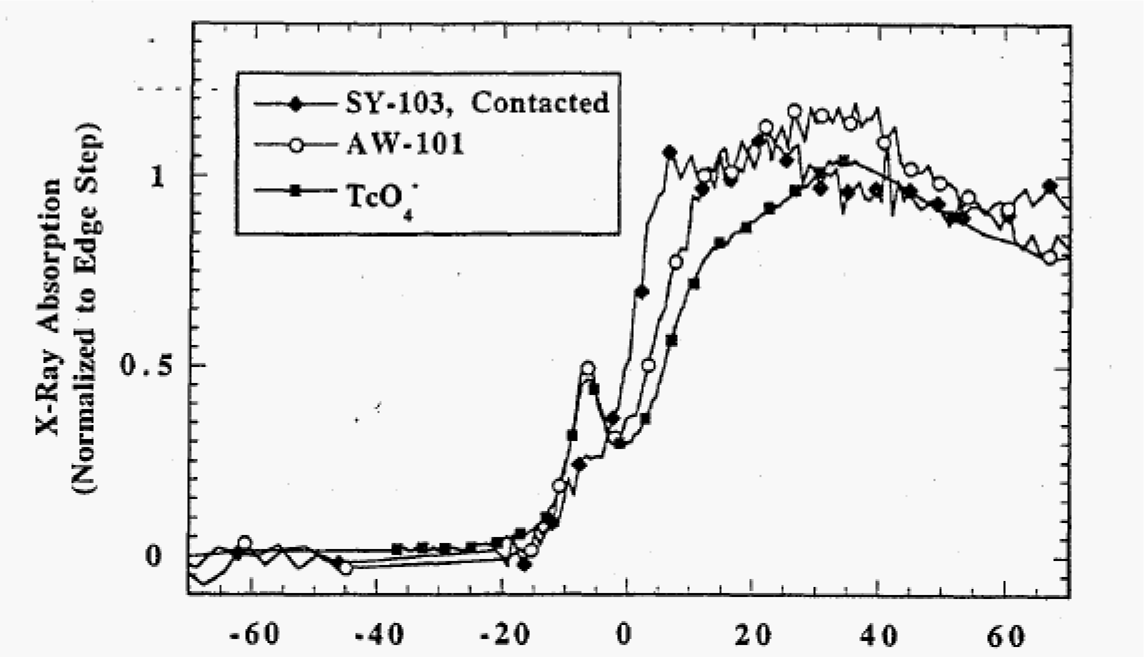

Figure 4.10. Comparison of XANES Spectra for AW-101 and SY-103 Supernates (contacted refers to removal of pertechnetate prior to analysis) with the Pertechnetate Standard

Later, after additional known Tc standards had their XANES spectra measured, other researchers took Blanchard's original DST supernate XANES spectra and compared them to the more-robust standards spectra. By using detailed curve-fitting algorithms these other researchers concluded that the n-Tc species shown in the Blanchard et al. (1996, 1997) reports (which concluded that the n-Tc species had Tc(IV) oxidation state) are better matched to known standards that have an oxidation state of Tc(I). These revised XANES interpretations are presented next.

Shuh et al. (2003) and Lukens et al. (2004, 2006) compared the original XANES spectra for SY-101 and SY-103 supernates reported by Blanchard et al. (1996, 1997) to XANES spectra of Tc(IV)-gluconate complexes that they created. Although the XANES spectra of Tc(IV) gluconate and the SY tank supernates' spectra are superficially similar, the energies of their Tc absorption edges differ by 1.6 eV, an energy difference beyond the uncertainties in the raw data. More importantly, no combination of the XANES spectra for the known standards for Tc(IV) gluconate and TcO_4^- will fit the SY DST supernates' spectra. The authors conclude that not only are the SY DST supernates not Tc(IV) gluconate, they cannot be any kind of Tc(IV) alkoxide complex. The energies of the Tc-K edges of all known Tc(IV) alkoxide complexes and other Tc(IV) complexes with oxygen neighbors, including the solid $TcO_2•2H_2O$, fall within a narrow range around 5.5 eV below the energy of the TcO_4^- absorption edge (Lukens et al. 2002). The Tc-K edge in the SY DST supernates occurs at 7.1 eV below the TcO_4^- absorption edge—a difference of 1.6 eV. Further, these researchers have shown that Tc(IV) gluconate is readily oxidized back to TcO_4^- when exposed to air. Thus, based on both findings, Shuh and Lukens do not believe that the n-Tc species in the SY tank supernates is Tc(IV) alkoxide complexes. Shuh, Lukens, and co-workers next reviewed radiopharmaceutical literature and found that [fac-$Tc(CO)_3^+$][1] complexes can be prepared from TcO_4^- in alkaline solution containing low CO concentrations. DST waste tanks contain CO in their headspace gas at concentrations of 0.25 to 0.5 mol% CO (Johnson 1996). Thus fac-$Tc(CO)_3^+$ could form in the Hanford waste tanks. These researchers then calculated theoretical XANES spectra for a variety of lower-valent Tc complexes regardless of whether the ligands were present in Hanford DST waste. Surprisingly, the best candidate theoretical spectrum that matched the DST supernate spectrum was for the $[Tc(CO)_3(H_2O)_3]^+$ --- Tc(I)-carbonyl complex shown in Figure 4.11. This complex and derivatives in which the coordinated water molecules are replaced by other ligands have received considerable attention as ^{99m}Tc radiopharmaceuticals. As mentioned, fac-$[Tc(CO)_3(H_2O)_3]^+$ can form in alkaline aqueous solution under conditions that are very similar to Hanford DST supernate waste that contains low CO concentrations.

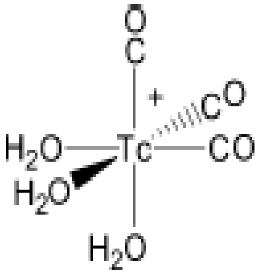

Figure 4.11. Structure for the Tc(I) Tri-Carbonyl Fully Hydrated Cation (from Lukens et al. 2004)

In other lab experiments, Shuh, Lukens, and co-workers synthesized both fac-$Tc(CO)_3$ and Tc(IV)-gluconate and performed detailed NMR and EXAFS characterization studies to obtain good NMR and EXAFS information on these pure compounds. When Tc(IV) gluconate was mixed with CO for 5 minutes and then heated to 85 °C in a closed NMR tube with air in the headspace, after 1 hour the sample contained a large amount of TcO_4^- as determined by ^{99}Tc NMR. After 18 hours at 85 °C, the solution was colorless and contained mainly TcO_4^- as determined by ^{99}Tc NMR. However, when gluconate was mixed with fac-$Tc(CO)_3^+$ it did form a complex with fac-$Tc(CO)_3^+$, which was indicated by the presence of a new peak in the ^{99}Tc-NMR spectrum at -1240 ppm. This mixed solution was 0.1-M gluconate in 1-M NaOH and an unspecified amount of fac-$Tc(CO)_3^+$. In another study, fac-$[Tc(CO)_3(H_2O)_3]^+$ was mixed with an SY-101 simulant[2] and both fac-$Tc(CO)_3(H_2O)(OH)^0$ and fac-$Tc(CO)_3(gluconate)^{2-}$ were observed. However, after one week, the only observable Tc species were fac-$[Tc(CO)_3(gluconate)]^{2-}$ and TcO_4^- based on detailed NMR measurements.

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[1] fac = When three identical ligands occupy one face of an octahedron, the isomer is said to be facial, or **fac**.

[2] SY-101 simulant: 3.78 M NaOH, 1.09 M $NaNO_2$, 0.42 M $Al(NO_3)_2$, 0.058 M NaF, 0.037 M Na_2CO_3, 0.034 M KF, 0.031 M iminodiacetic acid, 0.02 M Na_2HPO_4, 0.013 M Na_2-EDTA, 0.012 M potassium gluconate, 0.005 M sodium citrate, 0.005 M Na_2SO_4, 0.004 M $Ca(NO_3)_2$, 0.0005 M $Zn(NO_3)_3$, 0.0002 M Na_3NTA, 0.0002 M $Ni(NO_3)_2$, and 0.0002 M $Fe(NO_3)_3$.

Their experiments demonstrate that fac-$Tc(CO)_3$ species are stable in alkaline solutions approximating the composition of DST supernate. For comparison, alkaline solutions of Tc(IV) alkoxides are more air-sensitive, and will oxidize to TcO_4^- in less than a week if exposed to air. The results described above show that fac-$Tc(CO)_3$ complexes are stable under conditions found in Hanford DSTs, but do not establish whether they are actually the n-Tc species in the DST supernates.

As shown in Figure 4.12, the XANES spectra of the fac-$Tc(CO)_3$ complexes are very similar, if not identical, to those of the SY-101 and SY-103 n-Tc species reported by Blanchard et al. (1996, 1997). The Tc K-edge energies of the XANES spectra of fac-$Tc(CO)_3$ complexes occur at 7.5 eV below that of TcO_4^-, in "excellent" agreement with the observed edge shift of 7.1 eV for the n-Tc species in SY DST supernates. Most convincing is the fact that the spectrum of the n-Tc species in tank SY-103 can be fit using only the spectrum of fac-$Tc(CO)_3(gluconate)^{2-}$ and the spectrum of the n-Tc species in tank SY-101 can be fit using the spectrum of fac-$Tc(CO)_3(gluconate)^{2-}$ containing 7% TcO_4^-, presumably due to oxidation.

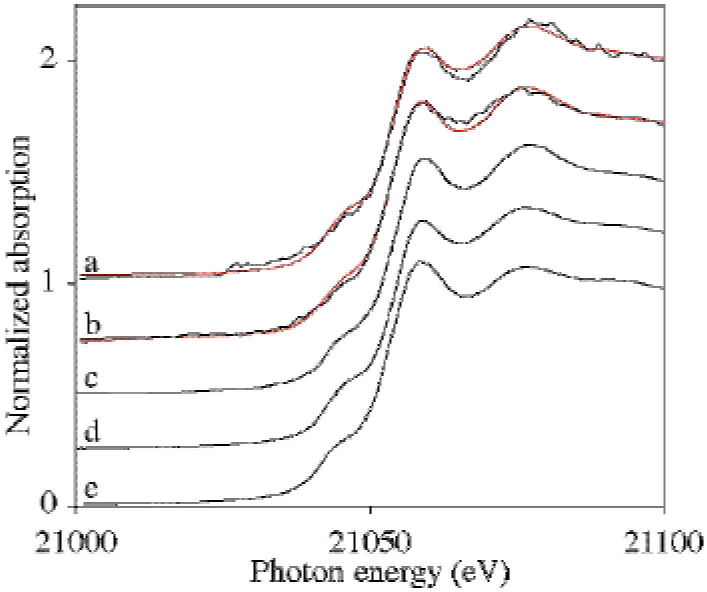

Figure 4.12. Tc K-edge XANES Spectra of a) n-Tc Species in Tank SY-103 (black) and $Tc(CO)_3(gluconate)^{2-}$ (red), b) n-Tc species in Tank SY-101 (black) and 93% $Tc(CO)_3(gluconate)^{2-}$ with 7% TcO_4^- (red), c) $Tc(CO)_3(gluconate)^{2-}$, d) $Tc(CO)_3(OH)(H_2O)_2$, e) $Tc(CO)_3(H_2O)_3^+$. The spectra of the n-Tc species in tanks SY-101 and SY-103 are from Blanchard et al. (1996, 1997).

Analogous Tc(I) dicarbonyl nitrosyl complexes, including $Tc(CO)_2(NO)(H_2O)_3^{2+}$, are known (Rattat et al. 2001). The $Tc(CO)_2(NO)$ complexes are somewhat more stable than fac-$Tc(CO)_3$ complexes and could be generated in Hanford tanks as a result of the radiolytic decomposition of nitrate and nitrite (Cook et al. 2001; Meisel et al. 2001). The XANES spectra of $Tc(CO)_2(NO)$ [not available] and fac-$Tc(CO)_3$ complexes would be similar. Given the probable similarity of the XANES spectra of the Tc(I) carbonyl complexes, assignment of the n-Tc species observed in Hanford DST supernates to a particular species is not conclusive, although $Tc(CO)_3(gluconate)^{2-}$ provides an excellent fit to the observed XANES spectrum of the n-Tc species in the SY-101 and SY-103 supernates.

The fac-Tc$(CO)_3$ complexes are not thermodynamically stable with respect to oxidation to TcO_4^-; **however, they are kinetically inert** due to their low-spin d^6 electronic structure. **As a result, they will react slowly with potential oxidizing agents, such as oxygen**. The kinetic inertness of these complexes also affects oxidation by strong oxidizers. Since fac-Tc$(CO)_3$ complexes will react relatively slowly with strong oxidizers (although presumably much faster than they react with oxygen), the strong oxidizers will preferentially react with other Hanford tank compounds, such as nitrite and/or organic carbon molecules, that are present in much higher concentrations and are much more reactive than the presumed fac-Tc$(CO)_3$ complexes.

In summary, based on the available information, Shuh, Lukens, and co-workers conclude that the problematic, n-Tc species in the Hanford DST supernates can be tentatively identified as Tc(I) carbonyl complexes derived from either $Tc(CO)_3^+$ or $Tc(CO)_2(NO)^{2+}$. These positively charged complexes may further combine with organic, inorganic, or hydroxyl anions to attain charge neutrality.

4.3.5 Correlation of n-Tc Percentages with Other DST Supernate Constituents

Rapko et al. (2013a) attempted to establish chemically logical correlations between available DST analytes and the estimates of supernate-soluble n-Tc . They used the DST supernate data from the TWINS database and tried to correlate variables with the percentage of n-Tc shown in Table 4.9 in this report. The goal was to see if strong correlations could be found that would allow extrapolation and estimates of the n-Tc percentages in supernates from other DSTs that have not been characterized for n-Tc but have been characterized for other constituents. From the work of Rapko et al. (2013a), the only acceptable chemically based correlation with data from the TWINS database was that the fraction of n-Tc present in characterized DSTs correlated with either of two closely related variables: 1) total dose experienced in the tank and 2) ^{137}Cs concentration in the tank supernatants. However, the observed inverse correlation is counterintuitive. One would expect that DST supernates with high doses/high ^{137}Cs concentrations to be excellent environments for radiolysis-induced reduction of components with concomitant formation of n-Tc . After attempting to find chemically relevant correlations, Rapko et al. (2013a) realized that each variable for which correlations were sought had a broad range of variability in measured concentrations, and thus large uncertainty bars. The impact of such large uncertainty was often compounded by the relatively steep dependence of the percent n-Tc on the variable in question. This led to situations that were described mathematically as a prediction of between 0% and 100% n-Tc due to the uncertainty associated with the variable itself. The uncertainties in ^{137}Cs concentrations were relatively small and thus a correlation was found; unfortunately, the correlation was negative, which is counterintuitive. There was no correlation between the n-Tc percentage and total soluble ^{99}Tc in the DST supernates for which n-Tc estimates were available.

5.0 Tc Recycle and Recovery in Off-gas Systems

The second objective of this literature review was to discuss the anticipated fate of ^{99}Tc during glass melter operations, and its recycle and recovery from various off-gas units downstream of the melters. Finally we describe briefly ^{99}Tc removal options from LAW feed and off-gas treatment equipment condensates and liquid effluents as an alternative to continuous recycle to the LAW melters. Sections 5.1 and 5.2 present a brief summary of reports containing such studies and key citations that provide more details.

5.1 Tc in Off-gas Train Recycle Back into Glass

Based on the current WTP process flowsheets, almost all of the ^{99}Tc in wastes retrieved from the Hanford tanks will remain in solution after various processes in waste feed delivery and pretreatment. This means that almost all of the ^{99}Tc will be found in the low-activity portion of the liquid waste stream that is sent from the PT Facility to the LAW Facility. Tc in LAW liquid waste forms species that are semi-volatile at the high temperature in the LAW melter, causing most of it to partition to the off-gas systems and to have low retention in the LAW glass. Depending on the composition of the LAW liquid waste stream, volatility studies on LAW simulants doped with ^{99m}Tc in the pertechnetate form show widely different percentages to be volatile (<10% to ~80%; according to Yanochko et al. 2012; Robbins and May 2013). However, the wide range includes experiments on LAW simulants, glass feed materials, and melter operating conditions outside of the range of parameters currently being considered. Under conditions that are realistically expected in the WTP LAW vitrification system, the best estimates of ^{99m}Tc volatility found in Matlack et al. 2010, 2011, and 2012 suggest a much narrower range clustered near 35% single-pass retention. Tc volatility from the melter is affected by other constituents in the melter feed, some of which would also build up during recycling of treated off-gases that are recycled back to the LAW melter. The testing at the Vitreous State Laboratory at The Catholic University of America (VSL) described in the cited Matlack reports contains more details for those interested. Our focus is on Tc capture in the off-gas and its recycle back to the LAW Facility, which is discussed next.

The current River Protection Project WTP baseline approach to mitigate the low ^{99}Tc single-pass retention in glass is to capture the volatile fraction in off-gas treatment systems and recycle it back to the PT Facility, where the off-gas captured ^{99}Tc is mixed with fresh LAW feed and concentrated by evaporation in the TLP-SEP-00001 evaporator prior to being sent to the LAW Facility for incorporation into glass (see the red box in Figure 5.1). A description of both the primary and secondary off-gas systems for the LAW Facility is found in Jenkins et al. (2013b), a document commonly called the BARD (Flowsheet Bases, Assumptions, and Requirements Document). The first system in the sequential LAW Facility off-gas treatment system is a film cooler that cools the exiting gases and helps prevent deposits from forming in the off-gas piping.[1] Downstream of the film cooler is the submerged bed scrubber (SBS). The SBS is designed for cooling and condensation of melter vapor emissions, removal of large particulates and aerosols, and scrubbing of water-soluble gas components. After removing larger particulates and aerosols in the SBS, the cooled off-gas is routed to the wet electrostatic precipitator (WESP) for high-efficiency removal of sub-micron particulates and aerosols. Particulates captured by the WESP are rinsed and combined with the SBS condensate in a condensate collection vessel. These fluids are transferred to the PT Facility, mixed with fresh LAW feed, and evaporated in TLP-SEP-00001. This resulting concentrate is transferred to the LAW Facility. The concentration of recycled components thereby increases in the LAW melter feed until the components are eventually incorporated in glass or are discharged through either the evaporator condensate solution or exit the WESP.

After the off-gas exits the WESP, it is heated to lower the relative humidity, preventing condensation in the HEPA filters. The HEPA filters remove additional particulate contamination. Activated carbon

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[1] Gases typically enter the film cooler at around 400 °C. Air is also injected into the film cooler for pressure control and to help minimize deposition of hard buildups in the transfer line. An off-gas flow velocity of greater than 50 ft/sec is normally maintained between the melter and the SBS. This velocity helps to minimize solids deposition within the off-gas transition line. The film cooler and transition line inlet are periodically sprayed with water during operation. This spray removes water-soluble species that then drain into the melter or SBS sump. Water spray into the film cooler and the off-gas transition line is one method to remove plated material inside the metal equipment.

adsorbers impregnated with sulfur and guard beds with lime (both located after the HEPA filters) are used to capture mercury, and acid gas and halides (including iodine), respectively. Mercury and acid gas removal is necessary to minimize poisoning of catalysts downstream of the carbon bed. Oxidation of volatile organic carbon species (VOC) and carbon monoxide and reduction of NOx are the next steps in the off-gas abatement system performed in the thermal catalytic oxidizer (TCO, also known as selective catalytic oxidation [SCO] unit). Through this catalyst the organics are generally oxidized to carbon dioxide, hydrogen chloride (if chlorinated compounds are present), and water vapor. After the TCO, ammonia is injected into the selective catalytic reducing unit (SCR) to aid in NOx reduction of the off-gas. In the SCR, ammonia reacts with the NOx and reduces it to nitrogen gas and water vapor. Finally, downstream of the SCR is a caustic scrubber that further treats the off-gas by removing the remaining acid gases such as HCl. A 5 M NaOH solution is added periodically to the recirculation line in the caustic scrubber to maintain a pH around 9. Liquids, which contain very little Tc, are collected in the caustic scrubber collection vessel and then pumped to the PT Facility, where they are combined with other fluids (primarily evaporator condensates) and sent to ETF (Yanochko and Corcoran 2012). The clean off-gas exiting the caustic scrubber is then discharged to the exhaust stack that vents to the atmosphere. In the event that the PT Facility is not operational when the LAW Facility is melting glass, the liquids from the off-gas treatment systems will be routed to the AW-102 DST for staging for the 242-A evaporator.

The SBS-WESP combined liquid waste stream is a dilute salt solution maintained near neutral pH by caustic additions, and will likely contain some insoluble glass-forming solids from melter feed carryover. One estimate of the chemical composition of this combined liquid waste stream (based on HTWOS[1] calculations) is documented in Robbins and May 2013 and Taylor-Pashow et al. (2014). These references suggest it will be a relatively high-volume waste stream (1.55 million gallons per year)[2] with concentrations of ^{99}Tc (4.14 E-05 Ci/L [2.44 mg/L]), chloride (2.68 E-02 mole/L), fluoride (7.64 E-02 mole/L), nitrites (2.32 E-04 mole/L), nitrates (8.92 E-02 mole/L), sulfate (2.43 E-02 mole/L), and ammonium (8.33 E-02 mole/L), and low concentrations of ^{137}Cs (5.21 E-06 Ci/L). After being combined with new LAW feed and concentrated by evaporation through repeated recycles, the SBS-WESP liquid wastes' ^{99}Tc concentration will build up until the Tc is either captured in the LAW glass or exits the recycle loop through the evaporator condensate or through the WESP. It is also possible for some Tc to be held up in process equipment and transfer lines.

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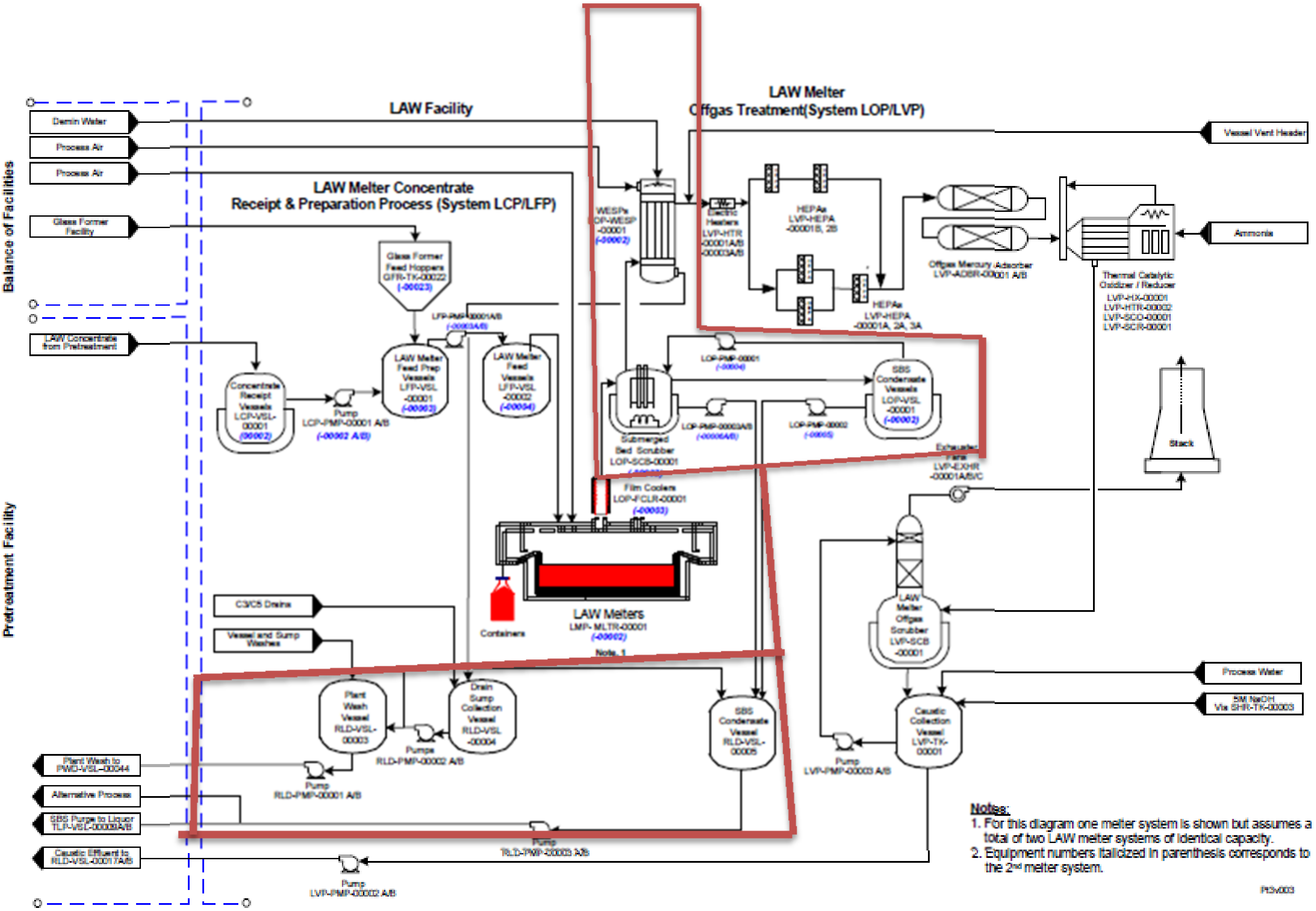
[1] A description of HTWOS and references to more details are provided in Appendix C.

[2] An alternate measure is volume per volume of LAW feed treated. For this metric, the estimates range from 1.5 to 2.0 (John Vienna, personal communication, July 2014).



Notes:
1. For this diagram one melter system is shown but assumes a total of two LAW melter systems of identical capacity.
2. Equipment numbers italicized in parenthesis corresponds to the 2nd melter system.

Figure 5.1. Baseline WTP LAW Unit Operations with Off-gas Capture Waste Streams that are Recycled Shown in Red Box (modified from Jenkins et al. 2013b)

The first studies that focused on improving the retention of Tc in LAW glass were reported in 2010 and 2011 in two reports by the VSL (Matlack et al. 2010, 2011).[1] Higher single-pass Tc retention in glass results in less Tc in off-gas system liquids and the attainment of steady state more quickly during tests where off-gas recycling is performed. Mass balance closure, although worthy of attention in these two test campaigns, was not the focus. Understanding the intrinsic volatility of Tc under the processing and melter idling conditions was the focus. Below are the most relevant (for this review) summaries from the two test series.

Tests were conducted at the crucible scale and on the DM10 vitrification system to investigate and develop methods for improving the retention of Tc in Hanford LAW glass during the vitrification process. Several strategies were evaluated for enhancing the retention of Tc and other volatiles in the glass product, including manipulation of the glass-forming additives, varying the type and concentration of reductants used in the melter feed, and varying the composition and rate of gases used to bubble the melt pool. The test results provide information on the retention of Tc and other volatiles over a range of LAW melter feed compositions as well as mechanisms for Tc volatilization during the vitrification process. In addition, in preparation for future larger-scale tests, Re was evaluated as a surrogate for Tc. Thirty-four DM10 melter tests were conducted over 400 hours of testing to investigate the retention of Tc in glass produced from simulated LAW streams. DM10 tests were performed to determine the effect of waste alkali concentration, reductant type, reductant concentration, additive type, gas composition used to bubble the melt pool, bubbling rate, and glass pool temperature (1100 °C and 1175 °C) on Tc retention in the product glass. Most of the tests were performed at nominal WTP processing conditions: 1150 °C and a bubbling rate sufficient to achieve a constant glass production rate of $\approx$2,250 kg/m^2/day. All feeds were spiked with a solution containing ^{99m}Tc in the pertechnetate form. In each test, a mass balance for Tc and other feed constituents of concern was measured across the glass pool, discharge glasses, and melter exhaust. Seven of the tests consisted of idling periods immediately following select feeding tests to differentiate between the potential sources of Tc loss.

Test results suggest the following:

- As expected, Tc retention improved as the conditions were made more reducing. However, practical limits are imposed by the increasing tendency for molten metal and sulfide formation, which can decrease melter life, and the decreased retention of sulfur in the glass melt. None of the many reductants evaluated performed significantly better than sugar.
- While many of the glass composition changes that were investigated had little effect or detrimental effects as compared to the present baseline glass compositions, positive effects of using zirconium and tin additives were identified.
- The use of glass-forming chemicals in reduced forms appears to be a promising approach with little, if any, downside. In particular, the simple change of the iron source from hematite to magnetite improved the retention of Tc. In contrast, several variables that were investigated either did not significantly improve Tc retention or have substantial associated disadvantages.
- None of the more reducing bubbling gases (than air) significantly improved Tc retention during feeding (though there was some improvement during idling). Thus, while this would have been a relatively simple change to implement, based on the present results, it does not appear to be effective.

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[1] Matlack et al. (2010) [VSL-10R1920-1] has also been published as May et al. (2010) [RPP-RPT-45887]; see complete citations in Section 8.0.

- Similarly, reducing the bubbling gas flow rate substantially did not significantly improve Tc retention. It seems likely that yet further reduction would ultimately provide some improvement but only at the cost of significantly reduced glass production rates, which would be economically unattractive.
- Processing at a lower glass pool temperature resulted in modestly increased Tc retention. Tc retention increases with decreasing glass pool temperature between 0.1 and 0.36 absolute percent per °C based on the tests conducted. The disadvantage of this approach is that the glass production rate decreases significantly with decreasing temperature. It is possible that this could provide one element of a practical strategy for the baseline LAW vitrification system, where the melter capacity is likely greater than what the balance of the facility can support. However, with the present emphasis on optimization and enhancement of the LAW Facility over and above its baseline specifications, such an approach would appear to be undesirable.

The results from the idling tests suggest the following principal findings:

- The rate constant for Tc loss during idling decreased with decreasing temperature by about 1% (absolute) per °C.
- The rate constant for Tc loss from the glass pool during idling decreased by about 50% for a 75% decrease in air bubbling rate. Re showed the same behavior with very similar absolute values for the rate constants.
- The rate constant for Tc loss during idling decreased by about 20% to 30% when air was replaced by a more reducing bubbling gas. In the tests where Re was used as a surrogate for Tc, the same behavior was observed with very similar absolute values for the Re loss rate constant.
- The rate constant for Tc loss during idling decreased by about 20% when the high-sodium glass melt was replaced by the lower-sodium glass melt.
- The rate constant for Tc loss during idling decreased by about 16% when starting from a strongly reduced glass pool. Re showed the same behavior with very similar absolute values for the rate constants.

The DM10 melter system used in this study did not have a sophisticated off-gas recycle system since the testing was focused on “single-pass” Tc retention. The DM10 melter and exhaust system is shown in Figure 5.2. A mass balance for Tc in this “single-pass” study was performed by measuring ^{99m}Tc (and Re) in the melter glass pool and in the discharged glasses (see green arrows), and in the melter exhaust (for both particulates and off-gas; see purple arrows). The mass balance for Tc in the tests was >80% for 22 of the 25 tests and >90% for 14 of the tests. The total recovery for Re was higher than Tc, with recoveries between 90% and 114% for the 11 tests that contained Re in the glass feed. The authors suggest that, given the short distance between the melter outlet and the exhaust sampling ports and the relatively small amount of deposits observed in the transition piping after the tests, deposits of Tc on the exhaust piping do not significantly affect the mass balance. The authors suggest that the Tc mass balance shortfall is caused by the exhaust samplings being performed over short periods during each melter test and subsequently being extrapolated to cover the entire testing time, while the discharged glass and feed Tc mass measurements encompass the entire testing period. Further, ^{99m}Tc emissions are largely particulate that was captured on the 0.3-micron filter in the off-gas sampling train; the small percentage captured in the impingers in the off-gas sampling train were believed to have been very fine particulate. Re was detected exclusively as particulate.

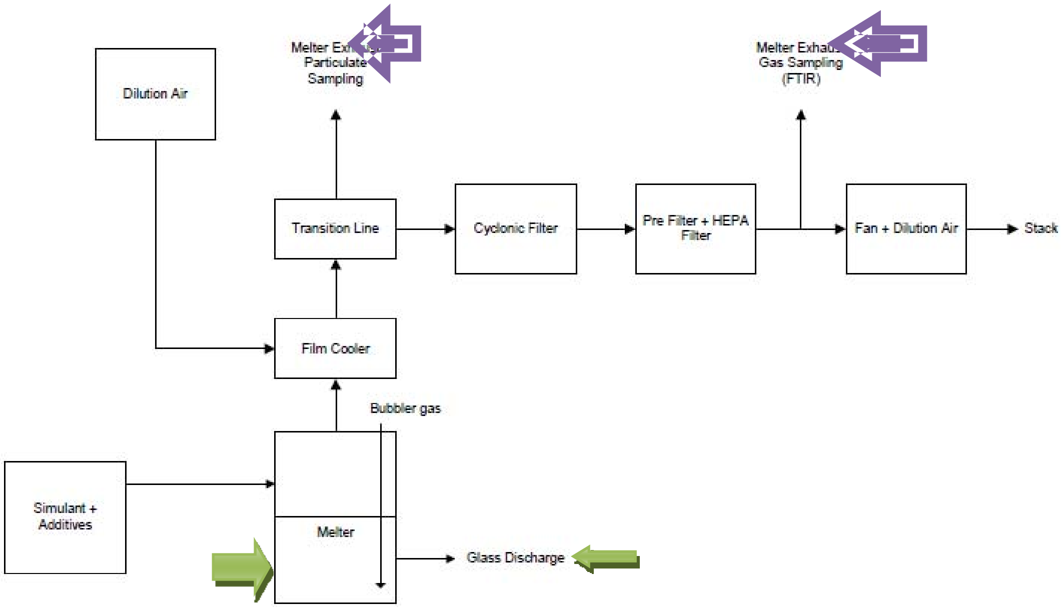

Figure 5.2. DM10 Melter and Exhaust System (taken from Matlack et al. 2010)

In a follow-on study (described as Phase 2), Matlack et al. (2011) present the results from melter testing at several scales (DM10, DM100-WV, and DM1200 melter systems).[1] These three melter systems span a factor of ~60 in scale-up, with the largest corresponding to about one-eighth scale for the WTP LAW melter. The DM1200 test employed an off-gas treatment system that is prototypical of the WTP LAW system and a one-eighth scale melter producing glass at about 2.7 metric tons per day. The key objective of Phase 2 studies was to continue the evaluation of single-pass retention of Tc and other species for a range of simulated LAW streams. Several strategies were evaluated for enhancing the retention of Tc (and other volatiles) in the glass product over a wide range of LAW waste compositions, building upon the findings from Phase 1. Methods were developed and demonstrated that result in single-pass retentions of Tc and iodine of greater than 50%. Further improvements are expected with off-gas effluent recycle.

Fifty-one DM10 melter tests totaling about 700 hours of testing, two DM100-WV melter tests totaling about 100 hours of testing, and one DM1200 test producing six metric tons of glass were

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[1] Details on VSL Melters Compared to WTP (from Matlack et al. (2011))

Parameter	DM10	DM100-WV	DM1200	LAW WTP
Melt surface area (m^2)	0.021	0.108	1.18	10.0
Plenum volume (m^3)	0.164	0.164	1.25	15.7
Glass melt pool mass (kg)	8	117	2000	Not stated

conducted. All tests were performed at 1150 °C and with a bubbling rate sufficient to achieve a constant glass production rate of about 2,250 kg/m^2/day. All DM10 and DM100-WV feeds were spiked with ^{99m}Tc in the pertechnetate form; the feeds used in these tests also contained measureable amounts of nonradioactive iodine and Re. In each DM10 and DM100-WV melter test, a mass balance for Tc and other feed constituents of concern was measured in the glass pool, discharge glasses, and melter exhaust. DM10 tests were performed to determine the effect of waste and glass composition, reductant type and concentration, and additive type on Tc retention in the product glass as well as to determine the effectiveness of iron oxalate over a range of waste compositions for enhancing Tc retention in glass. The two most promising enhancement strategies were scaled up to the DM100-WV melter and one of those tests was further scaled up to the DM1200 melter. Re was used as a surrogate for Tc during the DM1200 test and its fate was monitored throughout the off-gas system such that a mass balance could be calculated across the melter and primary off-gas system from measurements on discharge glasses, melter exhaust, primary off-gas system components exhaust, and off-gas system sump solutions.

The principal findings from these tests were:

- The amount of Tc and Re (and iodine) retained in the glass product varies widely across the seven LAW waste simulants investigated (representative of wastes from tanks 241-AP-101, -AN-105, -AN-107, -AN-104, -AN-102, -AZ-101, and -AZ-102).
- The primary factor underlying the observed variation of Tc, Re, and iodine retentions across waste types appears to be the nitrate content, with a lesser effect from the nitrite content. Retentions decrease as these species increase in the melter feed. As further corroboration of this effect, the retentions of Tc, Re, and iodine in the glass product decrease approximately linearly with increasing nitrogen oxide emissions.
- Of the various methods investigated in the present work and Phase 1, the most effective method for enhancement of Tc, Re, and iodine retention without excessive reduction of the glass melt is the use of iron (II) oxalate as an additive. Tc retentions of up to 61% were demonstrated using this method.
- The primary reaction of the iron (II) oxalate addition to the melter feed is oxidation of the divalent iron by nitrate, destroying nitrate and thereby reducing its concentration in the melter feed. As noted above, the decreased nitrate content in the melter feed results in increased retention, presumably by decreasing the tendency to form the more-volatile higher oxidation states of the species of concern. Iron (II) oxalate would therefore not be expected to provide an effective enhancement for wastes with very low concentrations of nitrates, as was observed experimentally in this work.
- Although the waste nitrate concentration was the most significant factor influencing Tc, Re, and iodine retention, other factors also influenced retention and additives other than iron (II) oxalate-enhanced retention, albeit to a lesser degree.
- Overall, Re was shown to be reasonable surrogate for Tc, although on average Re retention in the glass product was slightly higher (~10 relative %) than Tc in tests without iron (II) oxalate; tests with iron (II) oxalate showed a distinct shift in the correlation between Tc and Re retentions.
- Tests to examine scale-up from the DM10 to the DM100-WV (~5×) to DM1200(~60×) for melter feed with iron (II) oxalate as an additive showed remarkable consistency in the retentions of Tc, Re, and iodine across the first two melter scale-ups and for Re (only tracer present in the DM1200 test) in all three scale-ups.

- Testing on the DM1200 and prototypical off-gas system showed an excellent mass balance, with the amount of Re in the glass product and primary off-gas system sumps constituting greater than 99% of the feed Re. The efficient capture of volatilized Re in the DM1200 primary off-gas system suggests that recycle of the primary off-gas system effluents should be very effective in increasing the incorporation of Tc into the LAW glass product and minimizing the fraction reporting to secondary wastes.

The DM1200 off-gas treatment system, shown schematically in Figure 5.3, consists of an SBS, a WESP, a HEPA filter, a TCO, an SCR, a packed-bed caustic scrubber (PBS), and high-efficiency mist eliminators (HEME). Note, however, that HEME 1 is bypassed in the DM1200 LAW configuration and that HEME 2 is not part of the WTP off-gas train but is used to minimize entrained particle carryover into the VSL ventilation system. Components from the SBS to the HEPA remove essentially all of the particulates from the gas stream, with an estimated removal efficiency of greater than 99.9999% for particles greater than 0.3 μm in size. In the WTP facility, this provision segregates the radioactive components from the non-radioactive components in the system for maintenance and handling purposes. Water sprays in the DM1200 off-gas train are located in the WESP, PBS, and HEME 2 to wash down deposits and dissolved species into their respective collection sumps from which they can be sampled. Sampling points in the DM1200 vitrification system are shown in Figure 5.3 as the circles with S#.

With minor exceptions, the DM1200 off-gas system processing sequence follows the design for the full-scale WTP HLW and LAW melter systems, except for a cooling unit for the off-gas stream discharged from the SCR unit (which is present in the WTP off-gas train, but absent in the DM1200 system). Additionally, the WTP LAW off-gas system includes a wash in the transition line between the film cooler and the SBS. A comparison of the DM1200 and the WTP LAW off-gas treatment systems can be found by study of Figure 5.1 versus Figure 5.3.

^{99m}Tc and Re were spiked into the feed in all of the DM10 and DM100-WV tests but only Re was included in the feed for the DM1200 test. Melter off-gas emissions were sampled once per DM10 test, twice per DM100-WV test, and seven times during the DM1200 test. Also during the DM1200 tests, the SBS exhaust was sampled six times and the WESP exhaust was sampled three times.

Of interest to this review of the fate of Tc/Re in the melter off-gas in the three VSL melter systems, the following observation are important. In the DM10 and DM100-WV single-pass tests almost all the ^{99m}Tc activity measured was present on the 0.3-micron filters in the off-gas sampling system and the filter rinses, although in some samples a small amount was measured in the acidic impinger solution in the sampling train. The authors state that Tc that was collected in the impinger solutions must have been very fine particulates since the Tc had to pass through the filter. Re was detected exclusively as particulates captured on the exhaust gas sampling filters. The location of the sampling train is indicated by the purple arrow on the left in Figure 5.2.

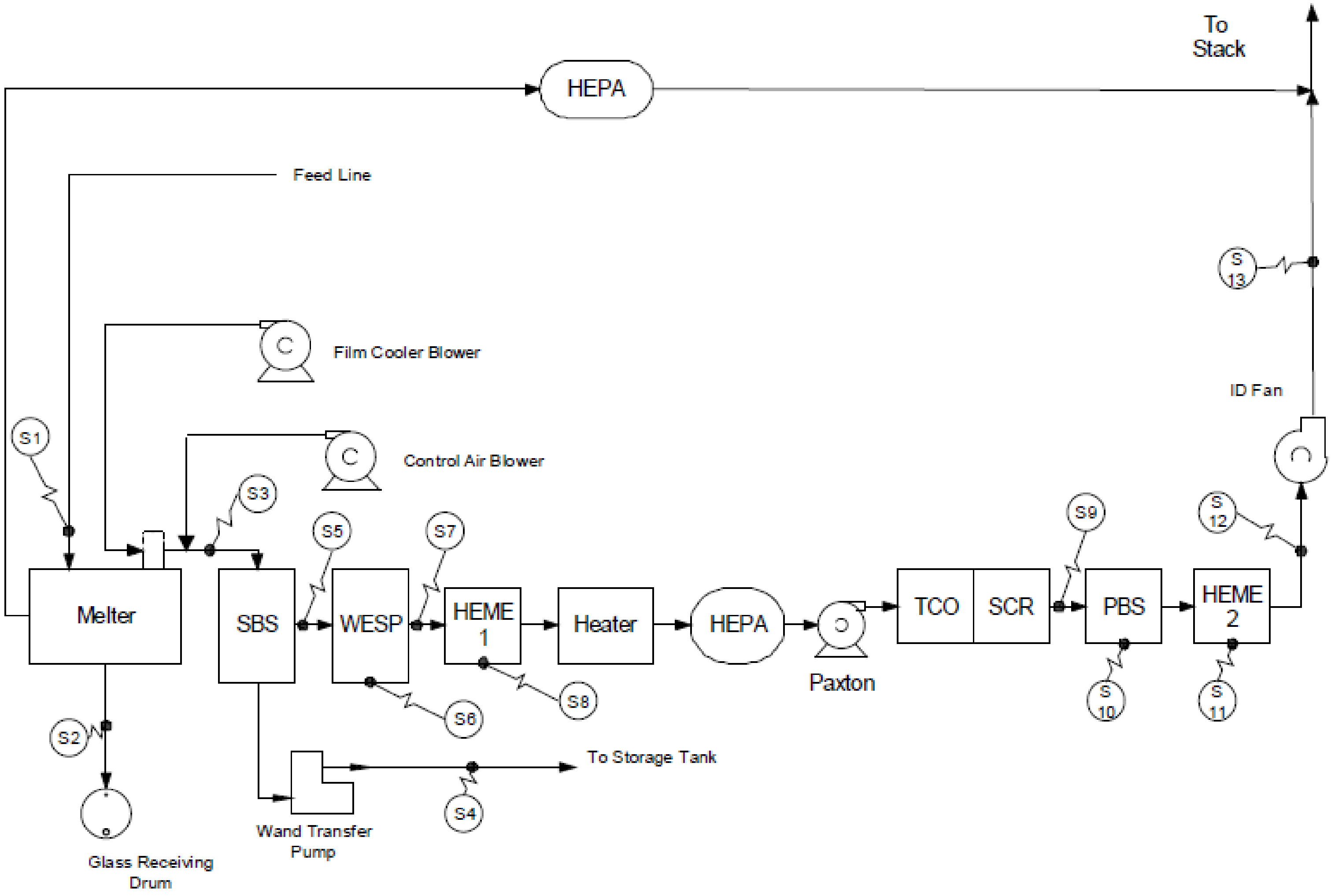

Figure 5.3. DM1200 Melter and Off-gas Treatment System (taken from Matlack et al. 2011)

The total recovery[1] of Tc and Re for the ~90 DM10 melter tests performed in both Phase 1 and Phase 2 studies (Matlack et al. 2010, 2011) averaged 97% and 100%, respectively, with some variation from test to test. In Phase 2, the ^{99m}Tc recovery in 40 of the 50 tests was within 15% of the "ideal" 100%.

For the two DM100-WV tests, the recovery of ^{99m}Tc was 97% and 88% and for Re was 118% and 117%, respectively. For both of these "mid-scale" melter tests, the total recovery for Re was generally higher than the recovery of ^{99m}Tc by about 10%. The authors state that the higher than 100% Re recovery is partly caused by the smaller relative concentration of Re in the DM100-WV tests when compared to tests performed using the smaller-scale DM10 melter. Lower Re concentrations in all the samples collected to calculate mass balance lead to increased analytical uncertainties, which means that the precision of the Re mass balance closure was correspondingly degraded.

Of greater importance to this literature review on off-gas recycle of Tc back to the LAW melter is the DM1200 test. Even though the DM1200 test described in Matlack et al. (2011) did not recycle fluids from the various off-gas equipment back to the melter, it did collect data on the percentages of Re that ended up in collection vessels related to the SBS, WESP, and PBS. These measurements along with the percentages of Re found in the melter exhaust and discharged glass allow a total Re mass balance across the entire DM-1200 melter and off-gas system to be calculated for the entire ~57-hour test. Over 12 metric tons of feed were processed through the DM1200 melter, resulting in the discharge of almost 6 metric tons of glass. As mentioned, during the DM1200 test, the SBS exhaust was sampled six times and the WESP exhaust was sampled three times. Solids carryover from the DM1200 melter ranged from 0.13 to 0.67 percent of feed solids. The SBS averaged 74% removal of the particulates emitted from the DM1200 melter and from 98% to 99.8% of the remaining particles were removed by the WESP, depending whether or not a deluge[2] of the WESP was included in the sampling interval.

The total recovery of Re for this DM1200 melter run was 100.8%, with 62% present in the glass, 28.1% in SBS solutions, 10.7% in WESP solutions, and less than 0.1% in the secondary off-gas system (solution collected and combined from the PBS and HEME2; see Figure 5.3). Matlack et al. (2011) conclude: "Testing on the DM1200 and prototypical off-gas system showed an excellent mass balance with the amount of rhenium in the glass product and primary off-gas system sumps constituting greater than 99% of the feed rhenium. The efficient capture of volatilized rhenium in the DM1200 primary off-gas system suggests that recycle of the primary off-gas system effluents should be very effective in increasing the incorporation of Tc into the LAW glass product and minimizing the fraction reporting to secondary wastes."

Matlack et al. (2010, 2011) remark that because there are no existing data on Tc or Re recycle to the melter after capture in a system prototypical of the WTP primary off-gas system, the collection of such data represents a significant need.

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[1] Recovery is defined as the sum measured in the discharge glass and inventory of the melter at the end of the test plus the amount measured in a one-hour melter exhaust sample extrapolated to the entire test duration divided by the amount of Tc fed over the course of the test.

[2] WTP plans to employ periodic water washes ("deluges") of the WESP internals to mobilize captured particles and maintain WESP collection efficiency. Electrical power to the WESP electrodes is shut off during the deluge operation, which results in essentially zero collection efficiency during that time. Each deluge event is estimated to be in the range of about ten to thirty minutes once every 24 hours.

Matlack et al. (2012)[1] produced the first-ever data on the fate of Tc during recycle of fluids from key WTP unit operations (SBS, WESP, vacuum evaporator) into the LAW glass melter. They measured the distribution and fate of Tc throughout the vitrification system and recycle loop, and the extent of partitioning to secondary waste streams. Seven different LAW simulants that mimicked waste compositions (from Hanford tanks 241-AP-101, -AN-102, -AN-104, -AN-l05, -AN-107, -AZ-101, and -AZ-102) were processed in nine, nominally 72-hour, continuous tests where glass was produced with feed that contained recycled off-gas solutions added to LAW simulant feed. All feeds were spiked with ^{99m}Tc (half-life 6.02 hours with a conveniently measured gamma emission) in the pertechnetate form. The nominal operating conditions set glass pool temperature in the DM10 melter at 1150°C, added sugar reductant at a stoichiometric sugar ratio of 0.5 (equivalent to 0.75 moles of organic carbon per mole of nitrate + nitrite), and bubbled air through the glass pool at a rate to achieve a constant target glass production rate of 2250 kg/m^2 per day. In each test, the mass of ^{99m}Tc was obtained across the glass pool, in the discharge glasses, in the SBS, WESP, and vacuum evaporators, and in the WESP exhaust and evaporator overheads. ^{99m}Tc can escape this “closed circuit” recycle loop in both the test set-up and the baseline WTP operating strategy via two routes: 1) in the off-gas stream exiting the WESP and 2) in the liquid condensate from the vacuum evaporator. When WTP is operational, the WESP exhaust effluent and vacuum evaporator condensates may be disposed ultimately in non-glass waste forms. A schematic of the DM10 melter and continuous recycle testing system is shown in Figure 5.4 and a photograph showing the size of the key components is shown in Figure 5.5.

Key findings from this work, which used the DM10 melter system, include the following (quoted from Matlack et al. (2012) where noted with “…” any words added for clarification are italicized):

- “With recycle, Tc retention in the glass product is increased by factors of at least 2 to 3 over the corresponding single-pass values for almost all glasses” made with the seven LAW simulant liquid waste compositions. For all but two LAW compositions, Tc retention in glass ranges from 68% to 84% of the total. In comparison, in 72 previously conducted single-pass DM10 tests (without recycle) using the same seven LAW simulants the average Tc retention was 35.2%.
- “The increase in Tc retention in *LAW* glass was limited by holdup of material in the system, particularly in the WESP *internals*, the film cooler, and transition line (*between the film cooler and SBS*). Mobilization of the *held up* material to make it available for recycle would likely further increase the retention in glass.”
- “The fraction of feed Tc exiting the recycle loop through the evaporator overheads was less than 0.03% during normal operations” and “the fraction of feed Tc exiting the recycle loop through the WESP exhaust ranged from 0.01% to 0.5% during normal operations”. However,” the fraction of feed Tc exiting the recycle loop through the WESP exhaust was critically dependent on the performance of the WESP and increased to above 10% (i.e., by a factor of about 500 or more) when the WESP was not functioning.”
- “In view of the complexity of the *recycle* test system and the numerous samples and analyses that were required to complete the mass balance, the measured average Tc mass balance closure of about 90% is considered very good." For the previous once-through LAW tests, Tc mass balance closures were 97%. However, the 10% recycle testing shortfall in Tc mass balance is significant with respect

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[1] This report has also been published as Abramowitz et al. (2012); RPP-54130, Rev. 0, see complete citation in Section 8.

to the ability to measure retentions in glass nearing 100%. "Additional studies are needed to evaluate potential reasons for the shortfall in the technetium mass balance."

- "Measurement of the fraction of technetium exiting the recycle loop provides a more direct and precise means of estimating the fraction of technetium reporting to liquid secondary waste and ultimately, to non-glass waste forms."

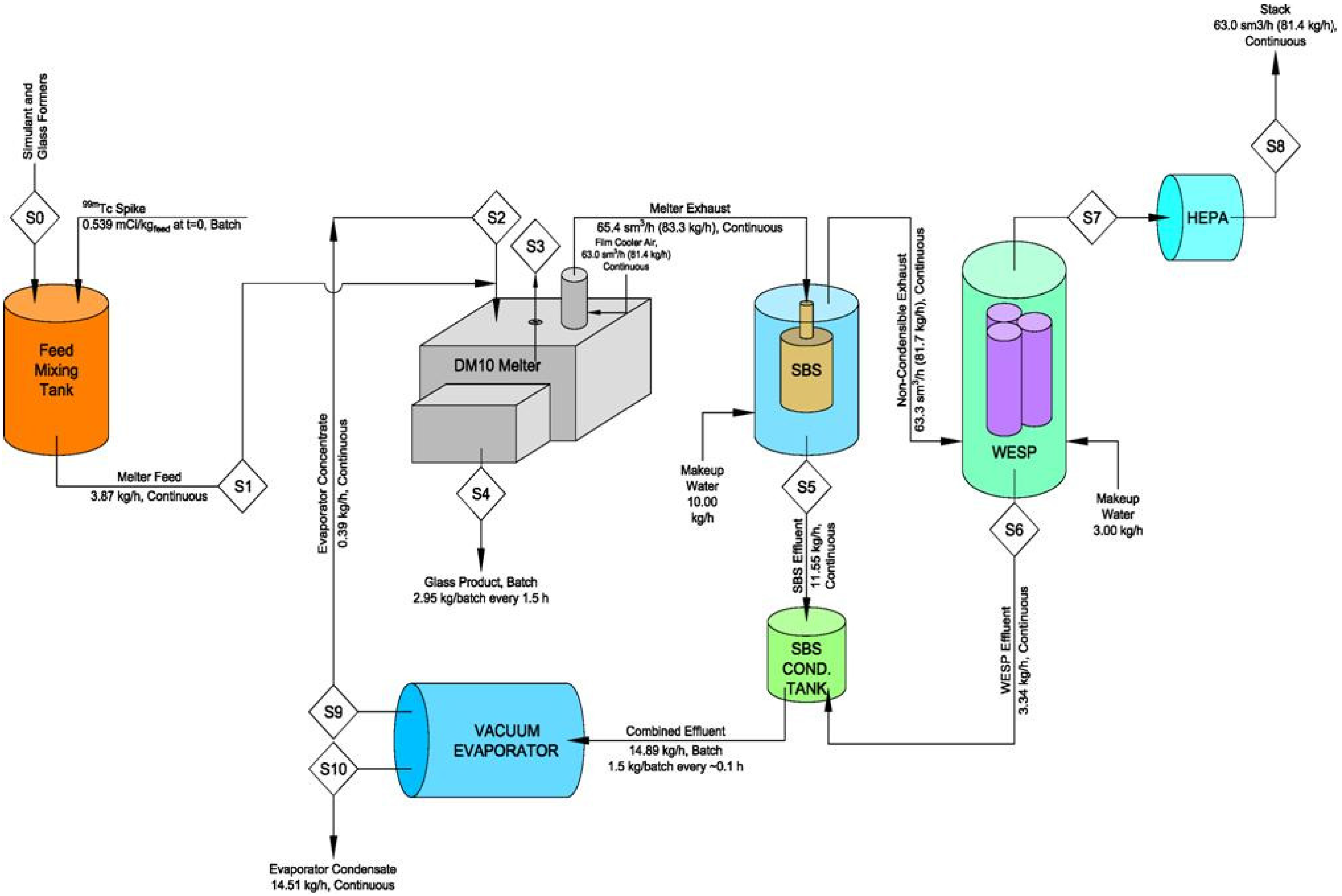

Figure 5.4. Schematic of the DM10 Melter and Continuous Recycle Testing Platform (from Matlack et al. 2012; their Figure 1.1c). Diamonds with S# represent sampling points for ^{99m}Tc and other key constituents.

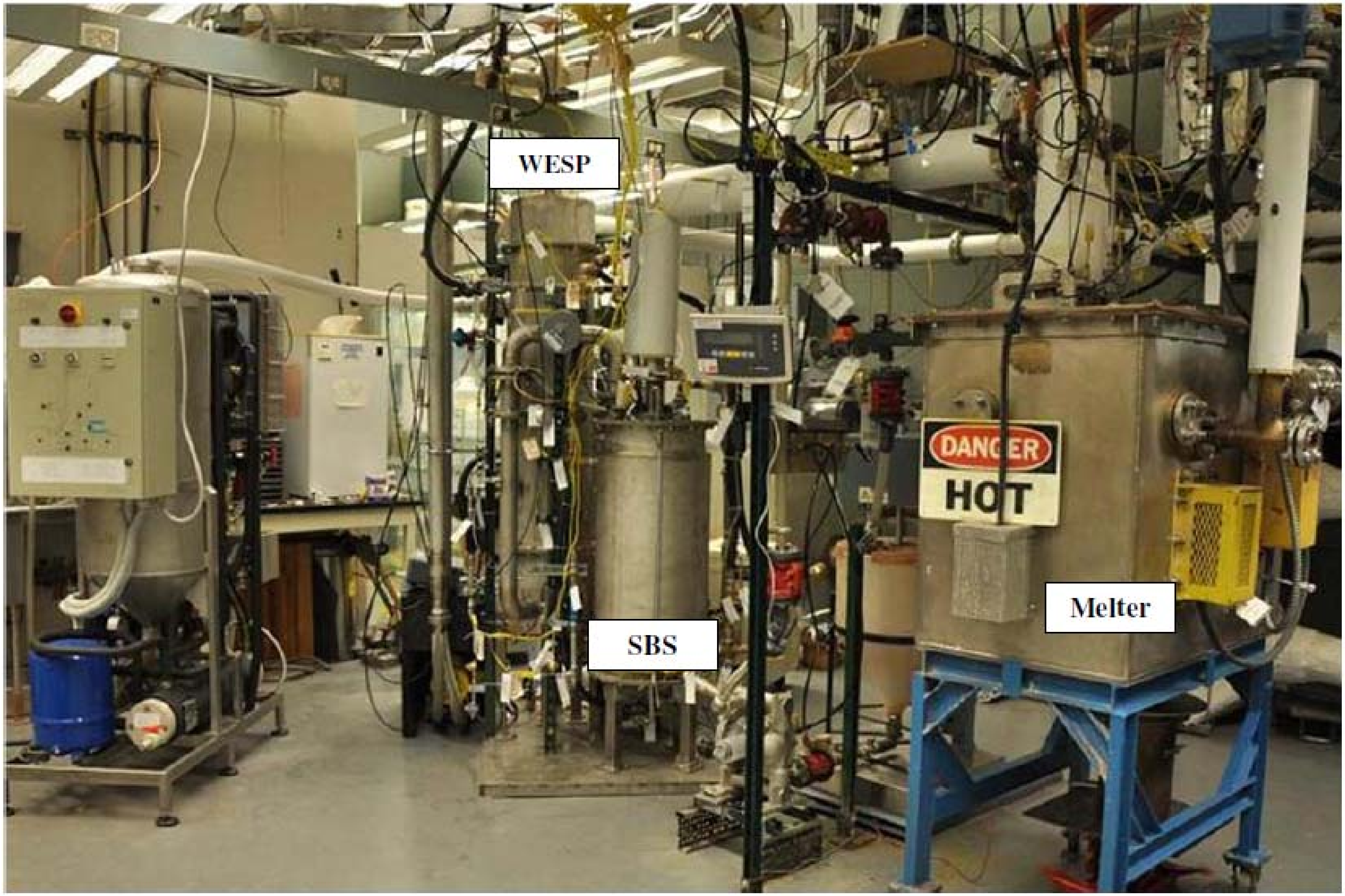

Figure 5.5. Photograph of the DM10 Melter and Continuous Off-gas Recycle Test Facility (from Ramsey 2012a; who obtained the photograph from VSL)

The significant ^{99m}Tc holdup in the WESP within the DM10 recycle train will be addressed in the WTP using daily water washes ("deluges") of the WESP internals to mobilize this material and maintain WESP collection efficiency. During the deluges, estimated to be 5 minutes once every 24 hours (or as needed, Jenkins et al. 2013, pg 3.3-8), the power is turned off such that the electrostatic precipitator plates are not efficiently capturing particles. Matlack et al. (2012) estimate that the WESP "*downtime*" will increase the percentage of Tc escaping through the WESP exhaust vent to about 15% during that downtime. The authors calculate[1] that the time-averaged fraction of Tc exiting WTP's WESP may range from about 0.1% to about 0.8%, in comparison to the 0.01% to 0.5% range measured in the DM10 recycle testing.

Within Matlack et al. (2012) it is stated "A baseline technetium mass balance has been generated[2] as part of System Plan 6[3] for the WTP facility, which employs a flowsheet that includes WTP LAW and HLW vitrification, as well as vitrification for supplemental treatment of LAW. That flowsheet includes recycle back to the WTP pretreatment facility of liquid effluents from off-gas treatment in both the WTP LAW and HLW facilities; in contrast, the supplemental LAW vitrification facility is modeled with recycle internal to that facility. Per the assumptions used in that mass balance, of the total technetium inventory in the tank farms, 99.3% is retrieved for treatment, 91.6% reports to the LAW glass product (from both WTP and supplemental LAW), 5.6% reports to the HLW glass, 0.18% reports to solidified liquid effluent, and 0.18% reports to other solid wastes."

A second estimate of the distribution of ^{99}Tc after complete vitrification is found in Jenkins et al. (2013a) in a document titled *2013 WTP Tank Utilization Assessment (TUA) Part 1: Potential Impact of Advanced Glass Models on the WTP*. Based on a scenario where the supplemental LAW (that LAW not capable of being vitrified through the first LAW facility) is also vitrified in a second, stand-alone facility with melters capable of vitrifying LAW feed at 60 metric tons per day of glass, once operating at full capacity, with recycling of off-gas Tc directly back to the second LAW facility's melters. The fate of ^{99}Tc is shown in Table 3-18 of Jenkins et al. (2013a). Their predictions are shown, along with the other predictions discussed, in Table 5.1.

Robbins and May (2013) show a third estimate, also presented in Table 5.1, for how the total ^{99}Tc presently in the 177 Hanford tanks distributes in the WTP glasses after completion of the entire vitrification campaign. The estimate is derived from the use of the HTWOS computer code, specific run MMR-13-008_Case-1-rev. 1.xlsx[4]. For this scenario/prediction, all the tank waste is processed and separated into HLW and LAW fractions. Then all the treated tank waste is vitrified; the HLW portion in the WTP HLW Facility and the LAW in two LAW vitrification facilities. That is, all LAW is vitrified as opposed to another option where a supplemental waste form is chosen (and a different solidification facility built) to process a portion of the LAW. This estimate for the final distribution of ^{99}Tc does not discuss any Tc becoming a secondary waste stream, which is not realistic no matter how small the

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[1] This calculation assumed that the WESP periodic deluge, in which the WESP power was turned off, was for 30 minutes each 24-hour period instead of the current estimate of 5 minutes every 24 hours.

[2] HTWOS run "4MinTimestep (6Melters)-mmr-ll-031" performed by RA Kirbride (WRPS) on 3/7/2011.

[3] Certa PJ, PA Empey, and MN Wells. 2011. *River Protection Project System Plan.* ORP-11242, Rev. 6, Office of River Protection, Richland, Washington.

[4] Details on this specific HTWOS computer run are found in SVF-2732, 2013, "MMR-13-008 data Case 1 rev 1.xlsx," Rev. 0, Washington River Protection Solutions, LLC, Richland, Washington.

percentage of Tc not reporting to LAW glass becomes. The objective of Robbins and May (2013) was not to provide details on the distribution of ^{99}Tc between glass and secondary waste. Rather, the objective was to describe alternative processes to treat ^{99}Tc captured in the primary off-gas system as an alternative to recycling it to the LAW melters, thus they were not focused on determining the small percentage of ^{99}Tc that will end up as a secondary waste (the small portion that escapes the primary off-gas system).

Table 5.1. Estimated Distribution of ^{99}Tc Currently in Hanford Tanks after Vitrification (from three predictions)

Robbins and May (2013)			Jenkins et al. (2013a)		Kirkbride cited in Matlack et al. (2012)
Estimated ^{99}Tc Distribution	Curies	% of Total ^{99}Tc	Curies	% of Total ^{99}Tc	% of Total ^{99}Tc
First LAW Vit Facility off-gas fluids—recycled until incorporated in LAW glass	7000	26.40%	9150	35.0%	No split between LAW Vit Facilities provided
Captured in first cycle into First Vit Plant LAW melter(s)	4190	15.80%			
Second LAW Vit Facility off-gas fluids—recycled until incorporated in LAW glass	8410	31.80%	15,100	57.8%	
Captured in first cycle into Second Vit Plant LAW melter(s)	5040	20.40%			
Total in LAW glass	24,640	94.50%	24,250	92.8%	91.6%
HLW plant glass product	1460	5.50%	1464	5.6%	5.6%
Secondary Waste (^{99}Tc captured and sent to LERF/ETF)	NA(b)	NA	52.6	0.2%	0.36%(a)
^{99}Tc retrained in equipment	NA	NA	3.05	0.01%	NA
^{99}Tc released to atmosphere	NA	NA	5	0.02%	NA
^{99}Tc residual left in tanks	NA	NA	NA	NA	0.7%
^{99}Tc not accounted for	NA	NA	370	1.4%	2.44%
Total ^{99}Tc	26,100	100%	26,145	100%	100%

(a) described as secondary "solid" wastes
(b) not addressed in this estimate

The total ^{99}Tc shown in the last row of Table 5.1 for the two estimates that give ^{99}Tc masses agree favorably with the two most recent BBI total ^{99}Tc inventory estimates shown in the bottom row of both Table 4.4 and Table 4.5. If the off-gas capture systems are as efficient as projected, the mass of ^{99}Tc that will not be processed into LAW glass (i.e., would likely become a secondary waste) is quite small (<1% of the total Hanford tank ^{99}Tc inventory).

The findings of Matlack et al. (2012, 2013) suggest that repeated recycling of the condensates and washes from the off-gas treatment unit operations (SBS, WESP, and vacuum evaporator) could incorporate >99% of the ^{99}Tc present in LAW liquid waste into LAW glass if the Tc observed to plate out on the transition pipes and internal metal surfaces of the SBS and especially the WESP can be readily flushed.

Holdup of Tc on metal surfaces in transition pipes between the LAW melter and the various off-gas capture equipment such as WESP internals remains a concern to us (and Ramsey (2012a,b); see discussion below) that would benefit from more study. Matlack et al. (2010) shows photographs of solids plated out on transition piping between the film cooler and exhaust gas sampling ports in the DM10 system after testing (~400 hours of glass production). Two of the photographs are shown in Figure 5.6. Matlack et al. (2010) caution that this particulate buildup does not necessarily represent conditions in the WTP LAW Facility, or for that matter other VSL glass vitrification facilities. For instance, in the VSL DM1200 test facility, the film cooler and transition line inlet are periodically sprayed with water during operation. This spray removes water-soluble species that then drain into the melter or Submerged Bed Scrubber (SBS) sump. Similar water flushing will be performed in the WTP vitrification facilities.

Ramsey (2012a,b) provides comments from a review panel on the DM10 off-gas recycle demonstration performed by Abramowitz et al. (2012) and Matlack et al. (2012). The comments were prepared by a review panel from several national laboratories, Hanford operations contractors, and local DOE/ORP staff solely based on information presented at the workshop and in Matlack et al. (2010, 2011); Matlack et al. (2012) was not available at that time. In addition, the most recent report, Matlack et al. (2013)—which provides information on improvements to the WESP operations, lab tests on ^{99m}Tc sorption on stainless steel, and a thorough analysis on measuring bias for direct gamma measurements of ^{99m}Tc in solid samples (glass and particulates captured in off-gas filters) versus ^{99m}Tc gamma measurements in liquids such as LAW waste simulant feed and water from SBS and WESP sumps—was not available to the reviewers.

Key issues that the reviewers felt needed more study or clarification include determining the steady-state Tc plate-out conditions using longer-duration testing and determining whether the full-scale WTP can adequately remove the plated-out material and recycle it to the LAW melters (see Figure 5.6 for examples of the solids plated onto metal surfaces in DM10 melter transition piping; but also consider Matlack et al. (2010)'s caution that this material may not exist in WTP piping and off-gas capture equipment because they are periodically flushed with water). Further, in a new report just released to the public, Matlack et al. (2013), the reliability of the WESP was addressed. Matlack et al. (2013) showed successful flushing of ^{99m}Tc from internal components into the recycled fluid stream over acceptably short flushing times.

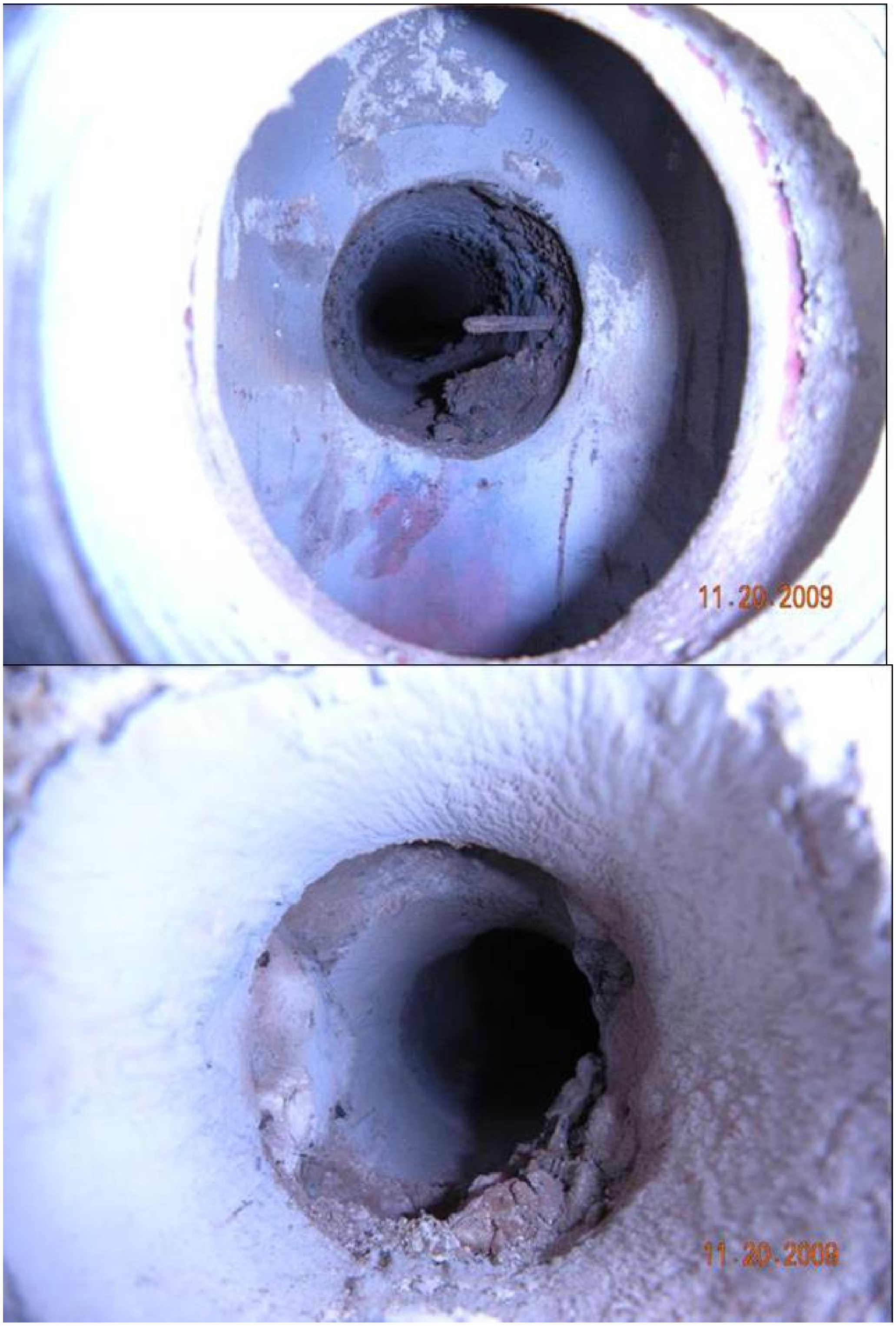

Figure 5.6. Photos of Buildup on Inside of Transition Piping in DM10 Melter Tests (from Matlack et al. 2010)

The following are other summary statements and questions from Ramsey (2012a,b) that were posed to the WTP, WRPS, and VSL staff based on reviewing available reports and several presentations. Some of the key issues that the reviewers felt needed more study or clarification include:

- Process holdup and Tc behavior in the specific WTP off-gas capture operations appear to strongly influence Tc capture and recycle prospects. During WESP malfunction, which the reviewers felt was "a strong possibility for the WESP not functioning for periods of time while WTP processes LAW," the fraction of Tc ending up as a secondary waste might be on the order of 13%. In addition, as noted in Matlack et al. (2012), the expected periodic WESP deluge flushing will generate a regular "blow-over" wherein Tc is not retained in the WESP. When the WESP is not operational, the Tc will likely be caught on the heated HEPA filters, which are next in line after the LAW WESP. Currently, the HEPA filters are planned to be placed into drums and sent to IDF. The reviewers recommend that a HEPA leaching step, with the leach water recycled to the PT Facility for return to the melter, be added to the flowsheet if it is critical to reduce the amount of Tc going into secondary wastes.
- **Tc retention in glass** for the DM10 Tc recycle demonstration **was limited to ~85% by process holdup in the system, particularly** in the **WESP internals** and the film cooler transition line, as well as the limited duration of the tests. In the light of how much of the Tc is held up on internal metal surfaces, **WTP needs to understand much better how effective the recovery of this material will be.** Another concern was whether the WTP equipment reliability, particularly the WESP, has been realistically modeled. The reviewers suggested that WESP performance needs to be explored further, particularly as it pertains to WTP for both abnormal failures and also planned maintenance outages.[1]
- The buildup of Tc on the off-gas metal internals cannot go on forever during the operation of WTP. This buildup will have solids that may contribute an appreciable amount of mass. Tc may be plating out on items that could end up as solid secondary waste that are destined to be buried at Hanford. If a larger portion of Tc will not be incorporated into the LAW glass, such "held up" Tc could potentially increase the mass of solid or liquid secondary wastes.
- Some of the reviewers recommended **longer-duration testing** to better understand Tc behavior and holdup within the off-gas system at **steady state**.
- Should additional ^{99m}Tc recycle tests be performed in the DM10 off-gas recycle system, there is a **need for data collection at more sampling points**. For example, counting data for the VSL-system HEPA filters are needed to determine whether the HEPA filters were effective in capturing any ^{99m}Tc that had passed through the WESP.

5.2 Alternative Processes for Stabilizing ^{99}Tc Captured in the WTP Primary Off-gas System Collection Vessels

Several reports have been published that describe alternative methods of treating the SBS-WESP fluids (which contain most of the ^{99}Tc after LAW liquid waste is fed through the LAW melter in each cycle). Essentially, the alternative methods would either send the SBS-WESP fluids off-site either with or without further treatment to remove the ^{99}Tc or would remove the ^{99}Tc from the fluids and then send the treated fluids to the Effluent Treatment Facility (ETF) on the Hanford Site. The ^{99}Tc removed from

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[1] Matlack et al. (2013), which was published after the review, does provide new information on the WESP operation and recommends steps that can improve WESP performance. Thus these concerns have been addressed.

the fluids would be further treated (generally adsorbed, precipitated, directly encapsulated into a low-temperature waste form such as Cast Stone, or vitrified either through the LAW, HLW, or a separate melter). The ^{99}Tc removed from the SBS-WESP fluids after "solidification" could be sent off-site or disposed on-site. Details on the various alternative methods of removing the ^{99}Tc from the SBS-WESP fluids and treating and disposing the final ^{99}Tc waste are discussed in Yanochko and Corcoran (2012), Yanochko et al. (2012), Robbins and May (2013), McCabe et al. (2013), Adamson et al. (2014), and Taylor-Pashow et al. (2014). Each report discusses the advantages of not recycling the ^{99}Tc-bearing SBS-WESP fluids to the PT Facility for recycle to the LAW melter. Many of these reports also offer cost and schedule estimates for the proposed alternatives. Nash et al. (2013) discuss removing ^{99}Tc directly from the LAW liquid waste before sending the treated LAW waste to the LAW melter or to another supplemental waste form. Removing the ^{99}Tc lessens some of the long-term performance issues for burial of non-glass waste forms in the IDF. Of course, the ^{99}Tc that is removed from the liquid LAW waste must be disposed in some solidified form at some disposal facility.

Regarding the speciation of ^{99}Tc in the off-gas and the resultant fluids, several of the reports suggest that the LAW melter is expected to convert all the ^{99}Tc that volatilizes to the pertechnetate ion when the hot gases contact the water in the SBS and the periodic WESP deluge flushing events. However, the dissociation of the volatilized Tc species upon contact with water in the SBS and WESP flush water has not been demonstrated to date. That said, the assumption seems reasonable given the likely oxidizing environment in the SBS and WESP treatment equipment. Matlack et al. (2012) did not address any ^{99}Tc speciation issues in their recycle studies because only the total ^{99m}Tc activity could be measured. Thus, at some point in the WTP studies, we recommend that some direct measurements of ^{99}Tc speciation be performed on the water within the collection vessels that store the liquids and suspended particles from the SBS and WESP prior to their being recycled to the PT Facility for recycle to the LAW melters.

6.0 Tc Inventory in Other Hanford "Pools"

Although the main focus of this review is to document the current inventory and distribution of ^{99}Tc in the Hanford storage tanks and to discuss the ^{99}Tc fate during retrieval, processing, and final disposal, there are other places on the Hanford Site where ^{99}Tc inventory exists. To get a complete picture of what has happened to the total ^{99}Tc created during nuclear fuel irradiation and its subsequent fate, a simple mass balance box model was created. A box was created for each type or place that ^{99}Tc has been disposed or currently resides, and attempts were made to assign a quantity to each box or ^{99}Tc "pool." The outcome of this mass balance exercise follows. This approach follows the spirit of EPA guidance for conducting remedial investigations (EPA 1988, p. 2-7). The guidance calls for development of a "conceptual site model" that includes known and suspected sources of contamination, types of contaminants and affected media, known and potential routes of migration, and known or potential human and environmental receptors. The stated purpose in the EPA (1988) guidance for developing and maintaining a conceptual site model is "to evaluate potential risks to human health and the environment" and to "assist in the identification of potential remedial technologies."

Figure 6.1 shows a schematic of the various "pools" or boxes in which ^{99}Tc distributes after production in the reactors used to produce plutonium. It also includes boxes for ^{99}Tc brought into the Hanford Reservation from off-site sources. Table 6.1 lists the curies of ^{99}Tc that are estimated to reside in each box or "pool" for those boxes where information is available. Many of the boxes have no values because data/estimates were not readily available. The table does capture the fate for the bulk of the ^{99}Tc generated at Hanford on a high-level view (amount produced, amount currently is storage tanks, amount released to the Hanford Site vadose zone, and amount shipped off-site with uranium). As shown in Table 6.1, the high-level ^{99}Tc mass balance is quite good; 32,600 Ci produced and ~32,600 Ci accounted for as long as we limit the estimate of ^{99}Tc inadvertently sent off-site with uranium for re-purification to a value of ~5,400 Ci. Estimates of the amount of ^{99}Tc sent off-site, based on the HDW estimates of the percentage of ^{99}Tc remaining associated with uranium in the REDOX and PUREX reprocessing flowsheet of ~21.5%, yield a value of 7,000 curies.

The partitioning of the "pool" of ^{99}Tc released to the vadose zone into the current boxes for "present in vadose zone," "present in aquifer sediments and groundwater," and ^{99}Tc that has "reached the Columbia River" can't be assigned estimates. These three "pools" are important for making site remediation decisions and site risk assessments but are not the main scope of the Tc Management Program that is funding this report. Appendix D provides some discussion and lists reports that discuss the complexities of making estimates for these three boxes (3A, 3B, and 3C in Table 6.1). An earlier estimate of where ^{99}Tc inventory resides or will reside in the future at the Hanford Site is found in Chapter 28 as Figure 28.1 of DOE/ORP 2010 and is reproduced herein as Figure 6.2, showing their estimated ^{99}Tc inventories on a log (Y-axis) bar chart.

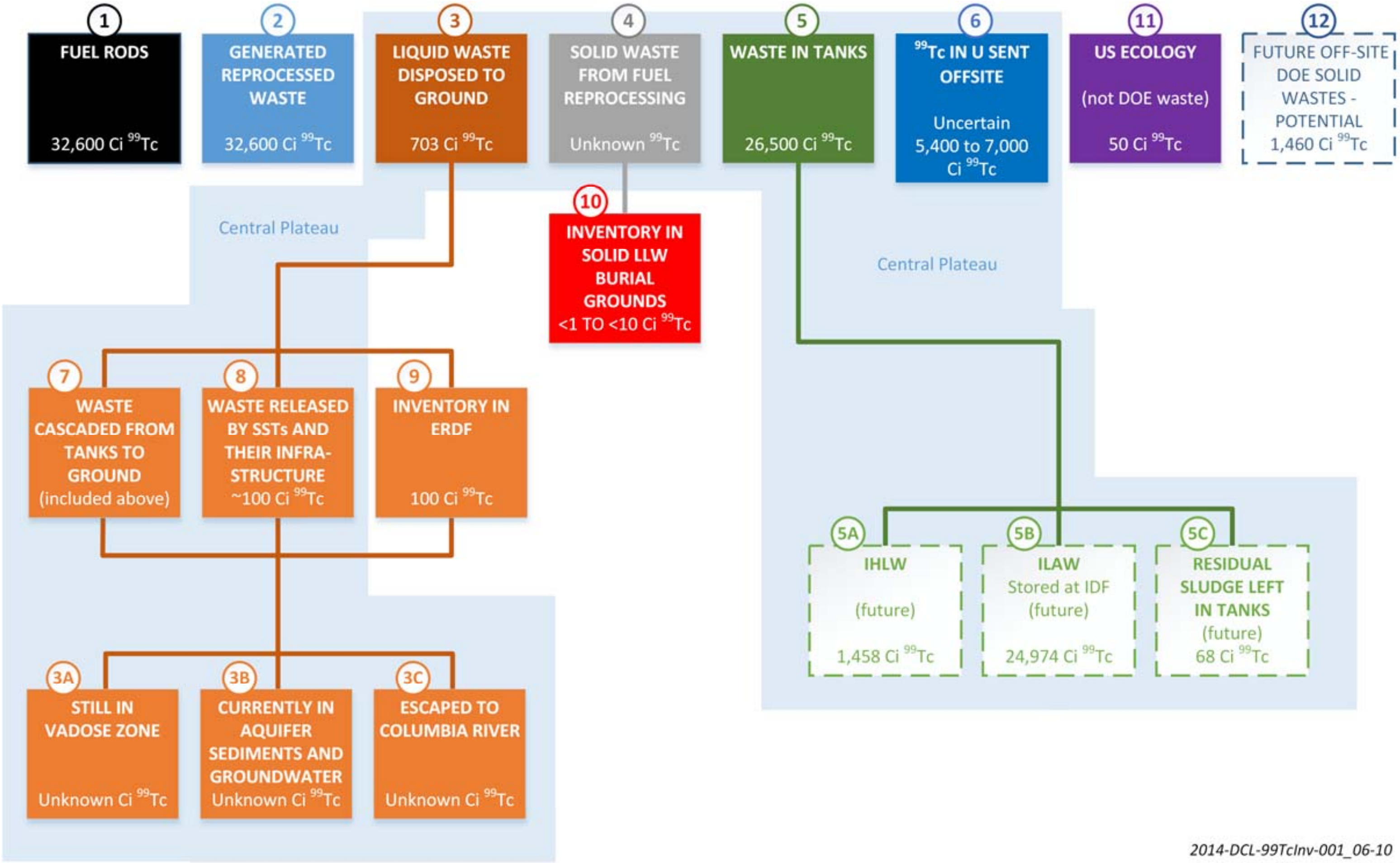

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Figure 6.1. Schematic of the Distribution and Fate of ^{99}Tc Between the Various "Pools"

Table 6.1. ^{99}Tc Distribution in Various "Pools" Used to Calculate Mass Balance or Hanford Site ^{99}Tc Conceptual Model

Box # (from Figure 6.1)	Pool	Description	^{99}Tc (Ci)	Notes
1	Fuel	Fission product in irradiated U/Th fuel rods	32,600	
2	Reprocessed Wastes Generated	Fuel dissolution	32,600	Three different processes used.
3	Liquid Wastes Disposed to Ground	Low-activity and lower-salt-content wastes	703	Soil Inventory Model (SIM) tracked & estimated.
4	Solid Wastes Generated during Fuel Reprocessing	Misc. solid materials likely not very radioactive	unknown	^{99}Tc likely very low because it remains soluble.
5	Wastes in Tanks	Current inventory in all 177 storage tanks	26,500	BBI model tracks this estimate.
6	^{99}Tc in U sent Off-site		21.5% × 32,600 = 7000 A more realistic value appears to be 5400	HDW version 5 model estimates 20%-23% went off-site with reprocessed U.
7	Waste Cascaded from Tanks to Ground		Included in Box #3	SIM tracked and estimated; combined with any direct waste co-disposal by disposal facility.
8	Waste Released by SSTs and Their Infrastructure		Included in Box #3 but when parsed out SST release ~100	SIM tracked but Tank Farm Contractor and State Ecology have updated using a Formal Joint Working Group.
9	Inventory in ERDF		100; so already included in Box #3	Most likely originates as contaminated soil from near Columbia River and spent resins from ^{99}Tc removal from groundwater (GW). Thus most of this inventory would be included in Box #3.
10	Inventory in Solid LLW Burial Grounds		<1 to <10	Estimated to be very low based on Solid Waste Burial Ground PA and documented records in Hanford reports from early 1990s.
11	US Ecology		50 (not DOE waste)	Commercial wastes come from off-site and are not Hanford-generated.

Box # (from Figure 6.1)	Pool	Description	^{99}Tc (Ci)	Notes
Box #3 can be further divided into three sub-boxes				
3A	Still in Vadose Zone		unknown	Database too sparse to estimate.
3B	Currently in Aquifer Sediments and GW		unknown	Database too sparse to estimate, areal extent of GW plumes well established but vertical concentration distribution in water too sparse to calculate volume of water contaminated and thus mass of ^{99}Tc in GW plumes.
3C	Escaped to Columbia River		unknown	Impossible to calculate. ^{99}Tc measurements began in 1993; most mobile contaminants such as ^{106}Ru reached water table many years earlier but most decayed ($t_{1/2}$ = 1 yr) before reaching Columbia River. ^{99}Tc did not decay so some likely reached Columbia River.
Mass Balance (Box #1) compared to Sum of Boxes (3, 4, 5, 6, 10) = 32,600 vs 32,600 to 34,200; dependent on Box #6				
Future WTP Activities parses Box #5 into the following three sub-boxes				
5A	IHLW	HLW glass	1460 (1)	Will be shipped off-site.
5B	ILAW	LAW glass or supplemental waste forms	24,250 to 24640(1)	Currently will stay on Hanford Site; many other possible scenarios dependent on ^{99}Tc removal vs. treated off-gas fluids recycle to LAW melter.
5C	Residual Sludge left in Tanks		68 (Appendix B of DOE/ORP (2010))	Is tracked in HTWOS and updated with real volume and concentration data when tank is closed.
Other Potential Future Activities (shipping solid wastes from small DOE site to Hanford for final burial)				
12	Potential Future Solid Wastes	Off-site DOE wastes shipped to Hanford for disposal	1460	Per TC&WM EIS Appendix D, Table D-87; uncertain fate; WA State has legal court cases underway to prohibit.

(1) See Table 5.1. The Table 5.1 ^{99}Tc inventory values are for the scenario where all tank waste is vitrified into glass in the HLW Facility and two LAW facilities. The Table 5.1 ^{99}Tc inventory estimates sum to between 26,100 and 26,150 Ci, compared to the current BBI value of 26,500 Ci. In Figure 6.1, we manipulated Boxes 5A, 5B, and 5C to sum to 26,500 Ci.

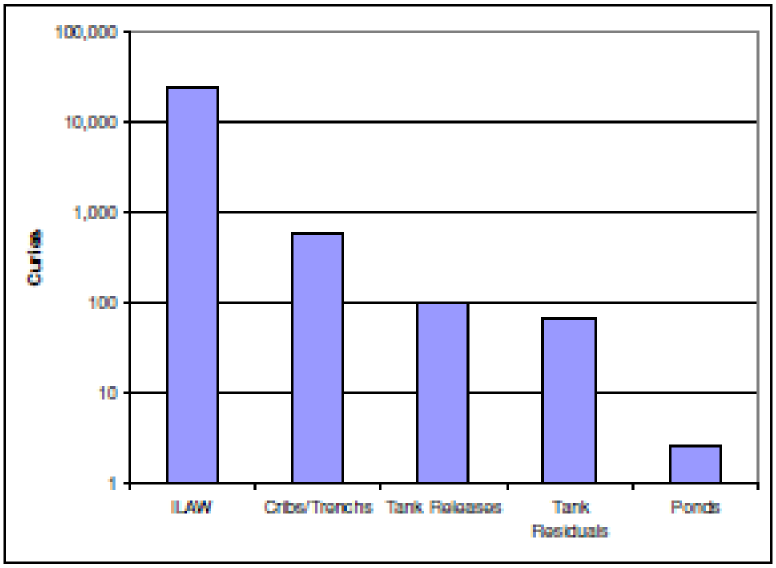

Figure 6.2. DOE/ORP 2010 Estimate of the Distribution of ^{99}Tc Between the Various "Pools"

The main focus of the Tc Management Program is to improve the understanding of the fate of the ^{99}Tc currently stored in the 177 storage tanks at Hanford. Based on Box #5 and discussions in Section 4.2, this accounts for 26,500 Ci. One future scenario for the disposition of the ^{99}Tc in the tanks is discussed in Section 5.0 and specifically Table 5.1. In this scenario, all the waste is split into two fractions (HLW and LAW) in the WTP PT Facility and each sent to WTP melters that produce glass. Within Box #5B, one can further split the ^{99}Tc inventory if other scenarios for LAW are chosen as briefly described in Section 5.0. One current discussion revolves around the fact that much of the ^{99}Tc present in LAW feed volatilizes upon entering the LAW melters and is captured in various off-gas treatment operations such that most of the ^{99}Tc is found in fluids generated in these off-gas units. Approaches for addressing the treated off-gas fluids are actively being discussed and many of the other tasks in the Tc Management Program are generating data to aid in the final decisions on how to address the treated off-gas fluids.

In summary, at a high level there is a good mass balance for how the ^{99}Tc produced at the Hanford Site is currently distributed. About 75% of the ^{99}Tc produced at Hanford currently resides in the 177 storage tanks, ~21% was shipped off-site with reprocessed uranium that was recycled/purified for other uses, and ~2% was released to the Hanford sediments. The fate of the 75% of the ^{99}Tc currently in the storage tanks is actively being discussed and several viable alternatives have been identified.

7.0 Summary and Conclusions

A thorough understanding of the inventory for mobile contaminants is key to any performance or risk assessment for Hanford Site facilities because potential groundwater and river contamination levels are proportional to the amount of contaminants disposed at the Hanford Site, especially those capable of migrating from discharge and disposal sites. At the Hanford Site, some ^{99}Tc has been purposefully or accidently released in the wastes generated during irradiated fuel reprocessing. ^{99}Tc has been found to be quite mobile and some has reached the groundwater. Because the majority of the total ^{99}Tc produced at Hanford (~32,600 Ci) is currently stored in Hanford's 177 tanks (~26,500 Ci), there is a critical need for knowledge of the fate of this ^{99}Tc as it is removed from the tanks and processed into a final solid waste forms. Based on current plans, the retrieved tank waste will be processed and solidified into glasses, with a small amount of secondary wastes from the WTP being solidified as some other low-temperature waste forms. Current flowsheets for the WTP process show most of the ^{99}Tc will be immobilized as LAW glass that will be disposed at the Hanford Site IDF; only a small fraction will be shipped off-site to a geologic repository in the form of IHLW glass. Past performance assessment studies, which focused on groundwater protection, have shown that ^{99}Tc would be the primary dose contributor to the IDF performance.

Based on this review of various technical reports and environmental impacts statements, the range in total curie inventory of ^{99}Tc produced at the Hanford Site as a consequence of irradiating fuel to produce plutonium is 31,000 to 34,000 Ci. This study's best estimate is that ~32,600 Ci of ^{99}Tc was formed, which equates to 1922 kg of ^{99}Tc. Some of this inventory was shipped off-site with uranium removed from the dissolved irradiated fuel for enrichment and reuse in making fresh fuel rods or for other purposes. As mentioned, some of the ^{99}Tc has been purposefully or accidently released to the Hanford vadose zone in wastes generated during irradiated fuel reprocessing, but most of the ^{99}Tc remains in the Hanford storage tanks. The BBI is the current official estimate of the current contents (46 radionuclides and 25 chemicals) in SSTs and DSTs. The BBI data are stored in the TWINS database. A query of the TWINS database in late January 2014 and subsequent analysis of the data leads to the following observations. The best estimate of the total ^{99}Tc inventory in the storage tanks is 26,500 Ci. There is more total ^{99}Tc in the 28 DSTs (1.51×10^4 Ci) than in the 149 SSTs (1.14 × 104 Ci). The SSTs in 200-W contain 8.11×10^3 Ci and those in 200-E contain 3.33×10^3 Ci of ^{99}Tc.

^{99}Tc present in three of the 200-W SST farms (TX, S, and SX) accounts for 54% of the ^{99}Tc in all SSTs (and 23.4% of the total ^{99}Tc tank inventory) and in 200-E the BY SST farm contains ~14% of the total ^{99}Tc in all SSTs (or 6% of the total ^{99}Tc tank inventory). The DSTs contain 57% of the total ^{99}Tc inventory in Hanford tanks, with the bulk (1.33×10^4 Ci) within four DST farms (AP, AN, AW, and AZ) in the 200-E Area.

A key issue addressed in this literature review was to estimate the amount of ^{99}Tc present as n-Tc species because these species have been shown to be difficult to separate from treated liquid tank wastes, should ^{99}Tc removal prior to vitrification become the preferred path forward. Further, there are no data on how n-Tc species will interact in the melters or whether they will partition into the glass similar to the pertechnetate form, which has been studied. First literature on the n-Tc issue, including several companion documents authored by Rapko and colleagues, was reviewed to understand how the n-Tc species were discovered.

Numerous reports describe the use of organic-based resins that preferentially sequester pertechnetate anions out of Hanford liquid waste streams. Most of the studies used pertechnetate-specific resins called SuperLig 639 or Reillex HPQ, both highly selective to sequestering only the pertechnetate form of ^{99}Tc. While processing DST supernates through columns packed with SuperLig 639, it was observed for some of the supernates that there was an immediate breakthrough of some of the ^{99}Tc present.

A second methodology that identified the presence of n-Tc species was batch sorption tests. For the batch tests, known amounts of SuperLig 639 or Reillex HPQ are contacted with actual Hanford DST supernatant liquids, usually at 100:1 (liquid volume to resin) for 4-day contacts. The batch slurry is then separated by centrifugation and/or filtration and the concentration of ^{99}Tc in the effluent is compared to the concentration in the influent using the traditional K_d construct. The Tc K_ds values for DST supernatants that were suspected to contain n-Tc species were significantly lower than the K_d values for DST supernatants that contained only the pertechnetate species. A third method used to explore Tc speciation used spectroscopic measurements on both carefully synthesized pure Tc compounds and actual DST supernates using several instruments and techniques, including UV-Vis, NMR, and synchrotron-based XAS, both XANES and EXAFS.

The conclusion from reviewing of all these studies was that the identities of the n-Tc species have not been definitively determined. However, based on all the various investigations and detailed measurements, n-Tc species in the Hanford DST supernates are tentatively identified as Tc(I) carbonyl complexes derived from either $Tc(CO)_3^+$ or $[Tc(CO)_2(NO)]^{2+}$ precursor molecules that may lead to final species such as $[Tc(CO)_3(\text{gluconate})]^{2-}$ as a result of the radiolytic decomposition of organics and nitrite/nitrate in the DSTs, which contain carbon monoxide in their head spaces as well as dissolved in the supernates. The XANES spectrum for “pure” $[Tc(CO)_3(\text{gluconate})]^{2-}$ provides an excellent fit to the observed XANES spectrum of the n-Tc species in the SY-101 and SY-103 supernates.

A summary of the n-Tc percentages in the supernates from DSTs that have had samples analyzed is shown in Table 7.1, which is based on an evaluation of Table 4.9. Table 7.1 shows the type of supernate waste in each DST, the ^{99}Tc inventory in each tank’s supernate, the percentage of n-Tc species in each tank’s supernate, and the ^{99}Tc curies of n-Tc . For a few of the DSTs, based on the similarity of the supernate waste type for some of the DSTs that have not been characterized for n-Tc , an estimate of whether they might contain “high” (assumed to be ≥70%) or “low” (assumed to be ≤10%) n-Tc is given. Then the n-Tc ^{99}Tc content is calculated, as shown in black type in the last column. For the DSTs that did have n-Tc estimated by techniques described in Section 4.3, the n-Tc ^{99}Tc content is shown in colored type. The bottom rows of Table 7.1 show statistics on the total n-Tc ^{99}Tc inventory that has been characterized (this quantity is labeled either as “Confirmed” [963 Ci], the sum of “confirmed” n-Tc values, or “Estimable” [1160 Ci], estimated from similarity of waste types). There are 10,900 Ci of ^{99}Tc in the supernate of the 28 DSTs, and an estimated 5370 of those curies cannot be categorized for n-Tc content given that the supernate waste type is designated as NA (not assignable to one of the HDW conceptual model waste types). The uncategorizable DST ^{99}Tc inventory represents 49% of the total ^{99}Tc in DST supernates, or 36% of the total ^{99}Tc in all phases of the DSTs, or 20.3% of the total Hanford tank BBI estimate for ^{99}Tc.

Table 7.1. Estimates of N-Tc ^{99}Tc Inventory in DST Supernates

DST Tank	Waste Type	Supernate ^{99}Tc (Ci)	n-^{99}Tc (%)	n-^{99}Tc (Ci)
241-AN-101	NA (Liquid)	7.59E+01	high	5.31E+01
241-AN-102	NA (Liquid)	4.60E+02	60	**2.76E+02**
241-AN-103	A2-SltSlr (Liquid)	2.81E+02	4	**1.12E+01**
241-AN-104	A2-SltSlr (Liquid)	4.62E+02	low	4.62E+01
241-AN-105	A2-SltSlr (Liquid)	5.88E+02	low	5.88E+01
241-AN-106	NA (Liquid)	1.31E+01	high	9.17E+00
241-AN-107	A2-SltSlr (Liquid)	3.03E+02	70	**2.12E+02**
241-AP-101	NA (Liquid)	7.24E+02	0	0
241-AP-102	NA (Liquid)	6.90E+02	unknown	unknown
241-AP-103	NA (Liquid)	9.21E+02	unknown	unknown
241-AP-104	NA (Liquid)	6.13E+01	70	**4.29E+01**
241-AP-105	NA (Liquid)	8.60E+02	unknown	unknown
241-AP-106	NA (Liquid)	3.68E+02	unknown	unknown
241-AP-107	NA (Liquid)	1.77E+02	unknown	unknown
241-AP-108	NA (Liquid)	6.65E+02	unknown	unknown
241-AW-101	A2-SltSlr (Liquid)	4.68E+02	4	1.87E+01
241-AW-102	NA (Liquid)	3.21E+02	unknown	unknown
241-AW-103	NA (Liquid)	3.42E+02	unknown	unknown
241-AW-104	NA (Liquid)	5.67E+02	unknown	unknown
241-AW-105	NA (Liquid)	5.35E+00	unknown	unknown
241-AW-106	**NA Lower (Liquid)**	5.56E+01	unknown	unknown
241-AW-106	**NA Upper (Liquid)**	2.01E+02	unknown	unknown
241-AY-101	**NA Lower (Liquid)**	2.08E+01	unknown	unknown
241-AY-101	**NA Upper (Liquid)**	1.84E+01	unknown	unknown
241-AY-102	NA (Liquid)	1.58E+02	unknown	unknown
241-AZ-101	NA (Liquid)	1.18E+03	0	0
241-AZ-102	NA (Liquid)	3.14E+02	4	**1.26E+01**
241-SY-101	NA (Liquid)	4.04E+01	65	**2.63E+01**
241-SY-102	NA (Liquid)	3.70E+01	high	2.59E+01
241-SY-103	S2-SltSlr (Liquid)	5.19E+02	70	**3.63E+02**
	Total ^{99}Tc in Supernate (Ci)	1.09E+04	Total “Confirmed” Non-pertech (Ci)	**9.63E+02**
	Total ^{99}Tc Ci in DST supernates with no info	5.37E+03	Total “Estimable” Non-pertech (Ci)	1.16E+03

Notes: high is assumed to be ≥70% n-^{99}Tc and low is assumed to be ≤10% n-^{99}Tc
AW-106 and AY-102 have two distinct layers of supernate within them that have different properties.
Waste types:
NA = not assignable.
A2-salt slurry comes from the second 242-Evaporator campaign using AW-102 feed tank (1981-1988).
S2-salt slurry comes from the second 242-S Evaporator campaign using SY-102 feed tank (1977-1980).

Based on the current WTP process flowsheets, almost all of the ^{99}Tc in retrieved tank wastes after processing in the PT Facility will be found in the low-activity portion of the liquid waste sent to the LAW melter. In the LAW melter, Tc is semi-volatile, causing most of it (estimates average around two-thirds) to partition to the off-gas systems. Off-gas treatment equipment downstream of the melters include a film cooler, SBS, WESP, heater, HEPA filters, a carbon bed to remove mercury, TCO and SCR units, and a caustic scrubber. Most of these off-gas units are used to capture suspended particulates, to condense steam, to scrub acid gases, and to convert volatile nitrous oxides into nitrogen. The current WTP flowsheets plan on continually recycling the condensates and liquid effluents from the SBS and WESP to a receiver tank in the PT Facility that sends the liquid to be mixed with fresh LAW and then this waste is further evaporated within the PT Facility. The combined evaporator concentrate is then sent to the LAW Facility or the supplemental LAW (SLAW) treatment facility (to be built in the future) where it is vitrified. Based on mass balance principles, at some point the ^{99}Tc in the recycle LAW feed will reach a steady-state concentration wherein the mass of ^{99}Tc being retained in the LAW glass will be the same as the combined mass of ^{99}Tc in the incoming recycled off-gas waste stream and the fresh LAW feed minus the small amount lost to the WESP off-gas or the evaporator condensate.

Matlack et al. (2012 and 2013) performed LAW treated off-gas fluids recycle testing of a prototype off-gas system using the DM10 melter at the VSL. They used seven different LAW simulants, each spiked with known amounts of ^{99m}Tc, a short-lived gamma-emitting isotope of Tc, in the pertechnetate form. They performed a mass balance for the ^{99m}Tc throughout the DM-10 melter and off-gas treatment system configured in a continuous recycle loop modeled after the WTP process. Key findings from this work include the following (sentences in quotes are directly from the cited documents):

- "With recycle, retention of Tc and Re in the glass product is increased by factors of at least 2 to 3 over the corresponding single-pass values for almost all glasses" made. For "all but two *LAW* compositions", Tc retention in glass *ranged* from 68% to 84%" of the total Tc in the feed.
- "The increase in Tc retention in LAW glass was limited by holdup of material, including Tc in the system, particularly in the WESP internals, the film cooler, and transition line. Mobilization of this material in order to make it available for recycle would likely further increase the retention in glass."
- In the DM-10 off-gas recycle system and the WTP baseline process there were two places where volatile species could exit the recycle system: 1) as overheads from the evaporator that captured SBS condensates and WESP flush water, and 2) as an exhaust downstream of the WESP. The % of ^{99m}Tc in the melter feed that escaped through these two recycle exits "was less than 0.03% during normal operations and much lower for many tests" for the overheads from the evaporator and "from 0.01 to 0.5%" from the WESP "during normal operations" of the WESP. "The fraction of feed technetium exiting the recycle loop through the WESP exhaust was critically dependent on the performance of the WESP and increased to above 10% (i.e., by a factor of about 500 or more) when the WESP was not functioning." Matlack et al. (2012) estimate that the WESP "*downtime*" in the WTP will increase the percentage of Tc escaping through the WESP exhaust vent from about 0.1% to about 0.8%, in comparison to the 0.01% to 0.5% measured in the VSL DM10 recycle testing.
- Based on Matlack et al. (2013) there is a strong indication that Tc hung up on the interior metal surfaces of the off-gas recycle equipment and transition lines can be flushed during the periodic cleaning cycles and that the WESP down time can be minimized, therefore the small amount (0.04 to 0.5%) of Tc that was directly measured exiting the overhead of the evaporator and the WESP exhaust suggests that LAW off-gas recycle will continuously return up to 99% of the Tc to the melter until Tc becomes incorporated in the glass.
- Measurement of the fraction of Tc exiting the recycle loop provides a more direct and precise means of estimating the fraction of Tc reporting to liquid secondary waste. One potential issue remains in the Tc off-gas capture and continual recycle back through the PT Facility to the LAW and SLAW melters. When the WESP is not operational, the Tc will likely be caught on the heated HEPA filter,

which is next in line after the LAW WESP. Currently, the HEPAs are planned to be placed into drums and sent to IDF. We recommend that a HEPA filter leaching step, with the leach water recycled to the PT for return to the melter, be added to the flowsheet if it is critical to reduce the amount of Tc going into secondary wastes.

Regarding the speciation of ^{99}Tc in the off-gas condensates and water flushes, several reports suggest that the ^{99}Tc volatilizes in the melter and when the hot gases contact the water in the SBS and WESP, the volatilized technetium becomes pertechnetate. However, the speciation of ^{99}Tc in treated off-gas fluids has not been explored to date. That said, the assumption seems reasonable given the likely oxidizing environment in the SBS and WESP solutions. Matlack et al. (2012) did not address any ^{99}Tc speciation issues because only the total ^{99m}Tc activity could be measured.

Several reports have been published that describe alternative methods of treating the SBS-WESP condensates/flush water (which contain most of the ^{99}Tc after LAW liquid waste is fed through the LAW melter in each cycle). Essentially, the alternative methods would either send the SBS condensates and WESP flush waters off-site either with or without further treatment to remove the ^{99}Tc or would remove the ^{99}Tc from the treated off-gas fluids and then send the treated liquid wastes to the ETF on the Hanford Site. The ^{99}Tc removed from these secondary wastes would be further treated (generally adsorbed, precipitated, directly encapsulated into a low-temperature waste form such as Cast Stone, or vitrified either through the LAW, HLW, or a separate melter). The ^{99}Tc removed from the SBS-WESP liquid wastes after "solidification" could be sent off-site or disposed on-site. Several reports on alternative strategies for addressing the ^{99}Tc in off-gas liquid wastes are briefly reviewed in this document.

The main focus of the Tc Management Program is to improve the understanding of the fate of the ^{99}Tc currently stored in the 177 storage tanks at Hanford. Based on Box #5 at the top of Table 6.1 and discussions in Section 4.2, this accounts for 26,500 Ci. At a high level there is a good mass balance for how the ^{99}Tc produced at the Hanford Site is currently distributed. About 80% of the ^{99}Tc produced at Hanford currently resides in the 177 storage tanks, ~20% was shipped off-site with reprocessed uranium that was recycled/purified for other uses, and ~2% was released to the Hanford sediments. The fate of the 80% of the ^{99}Tc currently in the storage tanks is actively being discussed and several viable alternatives have been identified. However, the partitioning of the "pool" of ^{99}Tc released to the vadose zone into "pools" for "present in vadose zone," "present in aquifer sediments and groundwater," and ^{99}Tc that has "reached the Columbia River" cannot be assigned estimates. The reasons that no technically defensible values of ^{99}Tc inventory can be assigned for these "pools" include lack of sufficient data on ^{99}Tc in the vadose zone, the complexity of the vadose zone hydrogeology (caused in part from the Ice Age catastrophic flood deposits), and lack of knowledge on the vertical distribution of ^{99}Tc in the aquifer plumes (most monitoring wells sample only the top 5 to 10 meters, and the aquifer is much thicker; where vertical distributions of groundwater contaminants have been measured there is large variability), and finally ^{99}Tc in groundwater has been monitored only since 1993 whereas most of the mobile contaminants were released in the mid 1940s through late 1970s. These three "pools" are important for making site remediation decisions and site risk assessments but are not the main scope of the Tc Management Program that is funding this report.

Final observations and recommendations for determining the amount and forms of n-Tc in Hanford tank wastes from this report and the companion document (Rapko 2014) follow. Any proposed characterization of n-Tc in Hanford tank waste supernate must consider that no means of isolating or concentrating n-Tc has been discovered (aside from evaporation—which also concentrates all other species present except water), although efforts in this area have been made (Schroeder and Ashley 2005). Therefore, any characterization must be made on the actual supernates, which contain around 10^{-5} M Tc at their maximum concentration.

Unfortunately, there is no clear method to characterize n-Tc species in Hanford tank supernate or the resultant liquids from dissolving tank saltcake and sludges. Of the commonly used methods for inorganic compounds, UV-vis and vibrational spectroscopy are perhaps the most routine, but to date these methods have not provided any useful information or any characteristic signal that can be associated with the presence of n-Tc. To date, the only two methods that have given information as to the oxidation state and structural features of n-Tc are XAS of one form or another and ^{99}Tc NMR. Unfortunately, XAS analysis is both time- and labor-intensive. Still, to the extent possible, continued analysis of new DST supernates for n-Tc by XANES and/or EXAFS is recommended. Size exclusion chromatography may also have some merit in separating ^{99}Tc species from each other prior to analysis (in hopes of simplifying the sample matrix) by the recommended NMR and XAS methods.

8.0 References

Abramowitz H, M Brandys, R Cecil, N D'Angelo, KS Matlack, IS Muller, IL Pegg, and RA Callow. 2012. *Technetium Retention in WTP LAW Glass with Recycle Flow-Sheet: DM10 Melter Testing.* RPP-54130, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.

Adamson DJ, CA Nash, DJ McCabe, CL Crawford, and WR Wilmarth. 2014. *Laboratory Evaporation of Hanford Waste Treatment Plant Low Activity Waste Off-Gas Condensate Simulant.* SRNL-STI-2013-0071, Rev. 0, Savannah River National Laboratory, Aiken, SC.

Ashely KR, N Schroeder, JA Olivares, and B Scott. 2004. *Identification of Non-Pertechnetate Species in Hanford Tank Waste, Their Synthesis, Characterization, and Fundamental Chemistry. Final Report September 15, 2001 through September 14, 2004.* DE-FG07-01ER63281, Texas A&M University-Commerce, Commerce, TX.

Bernard JG, E Bauer, MP Richards, JB Arterburn, and RM Chamberlin. 2001. "Catalytic Reduction of Pertechnetate (99TcO4-) in Simulated Alkaline Nuclear Wastes." *Radiochimica Acta* 89(1):59-61. doi:10.1524/ract.2001.89.1.059.

Blanchard Jr. DL, DE Kurath, GR Golcar, and SD Conradson. 1996. *Technetium Removal Column Flow Testing with Alkaline, High Salt, Radioactive Tank Waste.* PNNL-11398, Pacific Northwest National Laboratory, Richland, WA.

Blanchard Jr. DL, GN Brown, SD Conradson, SK Fadeff, GR Golcar, NJ Hess, GS Klinger, and DE Kurath. 1997. *Technetium in Alkaline, High-Salt, Radioactive Tank Waste Supernate: Preliminary Characterization and Removal.* PNNL-11386, Pacific Northwest National Laboratory, Richland, WA.

Blanchard Jr. DL, DE Kurath, and JR Bontha. 2000a. *Small Column Testing of Superlig 639® for Removal of 99Tc from Hanford Tank Waste Envelope A (Tank 241-AW-101).* PNWD-3004, Battelle-Pacific Northwest Division, Richland, WA.

Blanchard Jr. DL, DE Kurath, and BM Rapko. 2000b. *Small Column Testing of Superlig® 639 for Removal of ^{99}Tc from Hanford Tank Waste Envelope C (Tank 241-AN-107).* PNWD-3028, Battelle-Pacific Northwest Division, Richland, WA.

Burgeson IE, DL Blanchard Jr., and JR Deschane. 2002. *Small Column Testing of Superlig® 639 for Removing ^{99}Tc from Hanford Tank Waste Envelope A (Tank 241-AP-101).* PNWD-3222, Battelle-Pacific Northwest Division, Richland, WA.

Burgeson IE, DL Blanchard Jr., and JR Deschane. 2004a. *Small Column Testing of Superlig® 639 for Removing 99Tc from Hanford Tank Waste 241-AN-102 Supernate (Envelope C) Mixed with Tank 241-C-104 Solids (Envelope D) Wash and Permeate Solutions.* PNWD-3252, Rev. 1, Battelle-Pacific Northwest Division, Richland, WA.

Burgeson IE, DL Blanchard Jr., and JR Deschane. 2004b. *Small Column Testing of Superlig® 639 for Removing 99Tc from Hanford Tank Waste Envelope B (Tank 241-AZ-101).* PNWD-3281, Battelle-Pacific Northwest Division, Richland, WA.

Burgeson IE, JR Deschane, and DL Blanchard. 2005. "Removal of Technetium from Hanford Tank Waste Supernates." *Separation Science and Technology* 40(1-3):201-23.

Certa PJ, PA Empey, and MN Wells. 2011. *River Protection Project System Plan.* ORP-11242 Rev. 6, Office of River Protection, Richland, WA.

Cook AR, N Dimitrijevic, BW Dreyfus, D Meisel, LA Curtiss, and DM Camaioni. 2001. "Reducing Radicals in Nitrate Solutions." The NO_3^{2-} System Revisited." *J. Phys. Chem. A.* 105: 3658-3666.

Croff AG. 1980. *ORIGEN2 – A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*. ORNL-5621, Oak Ridge National Laboratory, Oak Ridge, TN.

DOE. 1996. *Tank Waste Remediation System, Hanford Site, Richland Washington Final Environmental Impact Statement (TWRS EIS).* DOE/EIS-0189. U.S. Department of Energy, Richland, WA.

DOE. 1997. *Final Waste Management Programmatic Environmental Impact Statement for Managing, Treatment, Storage, and Disposal of Radioactive and Hazardous Waste.* DOE/EIS-0200-F. U.S. Department of Energy, Office of Environmental Management, Washington, D.C.

DOE. 2012. *Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site, Richland, Washington (TC & WM EIS)* DOE/EIS-0391. Office of River Protection, U.S. Department of Energy, Richland, WA. http://www.hanford.gov/page.cfm/FinalTCWMEIS.

DOE/ORP. 2010. *RCRA Facility Investigation Report for Hanford Single-Shell Tank Waste Management—Tier 1 & 2*. DOE/ORP-2008-01 Rev. 1 Reissue, Office of River Protection, U.S. Department of Energy, Richland, WA.

Duncan JB, SE Kelly, RA Robbins, RD Adams, MA Thorson, and CC Haass. 2011. *Technetium Sorption Media Review*, RPP-RPT-50122, Washington River Protection Solutions, Richland, WA.

Egorov OB, MJ O’Hara, and JW Grate. 2004. “Microwave-Assisted Sample Treatment in a Fully Automated Flow-Based Instrument: Oxidation of Reduced Technetium Species in the Analysis of Total Technetium-99 in Caustic Aged Nuclear Waste Samples.” *Analytical Chemistry* 76:3869–3877.

Egorov OB, MJ O’Hara, and JW Grate. 2012. “Automated Radioanalytical System Incorporating Microwave-Assisted Sample Preparation, Chemical Separation, and Online Radiometric Detection for the Monitoring of Total ^{99}Tc in Nuclear Waste Processing Streams.” *Analytical Chemistry* 84:3090-3098.

EPA. 1988. *Guidance for Conducting Remedial Investigations and Feasibility Studies Under CERCLA, Interim Final.* OSWER Directive 9355.3.3-01, U.S. Environmental Protection Agency, Washington, D.C.

Geeting JGH, RT Hallen, LK Jagoda, AP Poloski, RD Scheele, and DR Weier. 2003. *Filtration, Washing, and Caustic Leaching of Hanford Tank AZ-101 Sludge.* PNWD-3206, Rev. 1, WTP-RPT-043, Rev. 1, Battelle-Pacific Northwest Division, Richland, WA.

Gumprecht RO. 1968. *Mathematical Basis of Computer Code RIBD.* DUN-4136, Douglas United Nuclear, Inc., Richland, WA.

Hassan NM, DJ McCabe, and WD King. 2000a. *Small-Scale Ion Exchange Removal of Cesium and Technetium from Hanford Tank 241-AN-103.* BNF-003-98-0146, Rev. 1, Westinghouse Savannah River Company, Aiken, SC.

Hassan NM, DJ McCabe, WD King, and ML Crowder. 2000b. *Small-Scale Ion Exchange Removal of Cesium and Technetium from Hanford Tank 241-AN-102.* BNF-003-98-0219, Westinghouse Savannah River Company, Aiken, SC.

Hassan NM, WD King, DJ McCabe, and ML Crowder. 2001a. *Small-Scale Ion Exchange Removal of Cesium and Technetium Form Envelope B Hanford Tank 241-AZ-102.* WSRC-TR-2000-00419, Rev. 0, Westinghouse Savannah River Company, Aiken, SC.

Hassan NM, WD King, DJ McCabe, LL Hamm, and ME Johnson. 2001b. *Superlig® 639 Equilibrium Sorption Data for Technetium from Hanford Tank Waste Supernates.* WSRC-MS-200l-00573, Westinghouse Savannah River Company, Aiken, SC.

Hassan NM, WD King, DJ McCabe, LL Hamm, and ME Johnson . 2002. “Superlig Equilibrium Sorption Data for Technetium from Hanford Tank Waste Supernates.” *Sol. Extr. Ion. Exch.* 20:211–226.

Hassan NM, K Adu-Wusu, CA Nash, and JC Marra. 2003. *Multiple Ion Exchange Column Tests for Technetium Removal from Hanford Tank Waste Supernate (U).* WSRC-MS-2003-00789, Westinghouse Savannah River Company, Aiken, SC.

Higley BA and DE Place. 2005. *Hanford Defined Waste Model – Revision 5.0.* RPP-19822, Rev. 0-A, CH2M Hill Hanford Group, Inc., Richland, WA.

Jenkins KD, Y Deng, and R Gimpel. 2013a. *2013 Tank Utilization Assessment (TUA) Part 1: Potential Impact of Advanced Glass Models on the WTP.* 24590-WTP-RPT-PE- 13 -003, Rev. 0, River Protection Project Waste Treatment Plant, Richland, WA.

Jenkins KD, Y Deng, RC Chen, MR Gros, R Gimpel and C Peredos. 2013b. *Flowsheet Bases, Assumptions, and Requirements.* 24590-WTP-RPT-PT-02-005, Rev. 7, River Protection Project, Waste Treatment Plant, Richland, WA.

Johnson GD. 1996. *Flammable Gas Program Report*. WHC-SP-1193, Westinghouse Hanford Company, Richland, WA.

Jungfliesh FM and BC Simpson. 1993. *Preliminary Estimate of Waste Inventories in Hanford Tanks through 1980*. SD-WM-TI-057, Rev. 0a, Westinghouse Hanford Company, Richland, WA.

Kincaid CT, PW Eslinger, RL Aaberg, TB Miley, IC Nelson, DL Strenge and JC Evans, Jr. 2006. *Inventory Data Package for Hanford Assessments.* PNNL-15829, Rev. 0, Pacific Northwest National Laboratory, Richland, WA.

King WD, NM Hassan, and DJ McCabe. 2000. *Intermediate-Scale Ion Exchange Removal of Cesium and Technetium from Hanford Tank 241-AN-102*. Report No. WSRC-TR-2000-00420, Westinghouse Savannah River Company, Aiken, SC.

King WD, NM Hassan, DJ McCabe, LL Hamm, and ME Johnson. 2001. *Technetium Removal from Hanford and Savannah River Site Actual Tank Waste Supernates with Superlig® 639 Resin.* WSRC-MS-2001-00760, Westinghouse Savannah River Co., Aiken, SC.

King WD, NM Hassan, DJ McCabe, LL Hamm, and ME Johnson. 2003. "Technetium Removal from Hanford and Savannah River Actual Tank Waste Supernates with Superlig® 639 Resin." *Sep. Sci. Tech.* 38(12,13): 3093–3114.

Kirkbride RA, GK Allen, PJ Certa, JA Lechlet, and S. Orcutt. 2005. *Hanford Tank Waste Operations Simulator Model Data Package for the Development Run for the Refined Target Case*. RPP-RPT-23412, Rev. 1, CH2M Hill Hanford Group, Inc., Richland, WA.

Krupka KM, WJ Deutsch, MJ Lindberg, KJ Cantrell, NJ Hess, HT Schaef, and BW Arey. 2004. *Hanford Tanks 241-AY-102 and 241-BX-101: Sludge Composition and Contaminant Release Data*. PNNL-14614, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Kupfer MJ, AL Boldt, BA Higley, KM Hodgson, LW Shelton, BC Simpson, RA Watrous, MD LeClair, GL Borsheim, RT Winward, RM Orme, NG Colton, SL Lambert, DE Place, and WW Schulz. 1997. *Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes*. HNF-SD-WM-TI-740, Rev. 0, Lockheed Martin Hanford Corporation, Richland, WA.

Kupfer MJ, AL Boldt, KM Hodgson, LW Shelton, BC Simpson, RA Watrous, MD LeClair, GL Borsheim, RT Winward, BA Higley, RM Orme, NG Colton, SL Lambert, DE Place, and WW Schulz. 1999. *Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes*. HNF-SD-WM-TI-740, Rev. 0C, Lockheed Martin Hanford Corporation, Richland, WA.

Kurath DE, DL Blanchard Jr., and JR Bontha. 2000. *Ion Exchange Distribution Coefficients for 137Cs and 99Tc Removal from Hanford Tank Supernatants AW-101 (Envelope A) and AN-107 (Envelope C).* PNWD-2467, Battelle-Pacific Northwest Division, Richland, WA.

Lindberg MJ and WJ Deutsch. 2003. *Tank 241-AY-102 Data Report*. PNNL-14344, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Lukens WW, JJ Bucher, NM Edelstein, and DK Shuh. 2001. "Radiolysis of TcO_4^-in Alkaline, Nitrate Solutions: Reduction by NO_3^{2-}." *J. Phys. Chem. A* 105:9611-9615.

Lukens WW, JJ Buchner, NM Edelstein, and DS Shuh. 2002. "Products of Pertechnetate Radiolysis in Highly Alkaline Solution: Structure of TcO_2●H_2O." *Environ. Sci. Technol.* 36:1124–1129.

Lukens WW, DK Shuh, NC Schroeder, and KR Ashley. 2004. "Identification of the Non-Pertechnetate Species in Hanford Waste Tanks, Tc(I)-Carbonyl Complexes." *Environmental Science & Technology* 38:229–233.

Lukens WW, DK Shuh, NC Schroeder, and KR Ashley. 2006. "Behavior of Technetium in Alkaline Solution: Identification of Non-Pertechnetate Species in High-Level Nuclear Waste Tanks at the Hanford Reservation." *ACS Symposium* 943:302–317.

Mann FM, RJ Puigh, SH Finfrock, EJ Freeman, R Khaleel, DH Bacon, MP Bergeron, BP McGrail, SK Wurstner, K Burgard, WR Root, and P LaMont. 2001. *Hanford Immobilized Low-Activity Tank Waste Performance Assessment: 2001 Version.* DOE/ORP-2000-24, Rev. 0, Office of River Protection, U.S. Department of Energy, Richland, WA.

Mann FM, RJ Puigh, R Khaleel, S Finfrock, BP McGrail, DH Bacon, and RJ Serne. 2003. *Risk Assessment Supporting the Decision on the Initial Selection of Supplemental ILAW Technologies.* RPP-17675, Rev. 0, CH2M Hill Hanford Group, Inc., Richland, WA.

Matlack KS, IS Muller, I Joseph, and IL Pegg. 2010. *Final Report Improving Technetium Retention in Hanford LAW Glass - Phase 1.* VSL-10R1920-1, Rev. 0, The Catholic University of America, Vitreous State Laboratory, Washington, DC.

Matlack KS, IS Muller, RA Callow, N D'Angelo, T Bardakci, I Joseph, and IL Pegg. 2011. *Final Report Improving Technetium Retention in Hanford LAW Glass - Phase 2.* VSL-11R2260-1, Rev. 0, The Catholic University of America, Vitreous State Laboratory, Washington, DC.

Matlack KS, H Abramowitz, M Brandys, IS Muller, RA Callow, N D'Angelo, R Cecil, I Joseph, and IL Pegg. 2012. *Technetium Retention in WTP LAW Glass with Recycle Flow-Sheet: DM10 Melter Testing.* VSL-12R2640-1, Rev. 0, The Catholic University of America, Vitreous State Laboratory, Washington, DC.

Matlack KS, H Abramowitz, IS Muller, K Gilbo, M Penafiel, and IL Pegg. 2013. *Wet Electrostatic Precipitator Performance and Technetium-Rhenium Behavior in LAW Recycle Flow-Sheet.* VSL-13R2800-1, Rev. 0, The Catholic University of America, Vitreous State Laboratory, Washington, DC.

McCabe DJ, NM Hassan, WD King, JL Steimke, MA Norato, LL Hamm, LN Oji, and ME Johnson. 2000. *Comprehensive Scale Testing of the Ion Exchange Removal of Cesium and Technetium from Hanford Tank Wastes.* WSRC-MS-2000-00499, CH2M Hill Hanford Group, Richland, WA.

McCabe DJ, WR Wilmarth, and CA Nash. 2013. *Waste Treatment Technology Process Development Plan for Hanford Waste Treatment Plant Low Activity Waste Recycle*. SRNL-STI-2013-00351, Rev. 0, Savannah River National Laboratory, Aiken, SC.

Meisel D, DM Camaioni, and TM Orlando. 2001. "Radiation and Chemistry in Nuclear Waste: The NOx System and Organic Aging. Nuclear Site Remediation: First Accomplishments of the Environmental Management Science Program." *American Chemical Society Symposium*. 778: 342-361 Washington DC, edited by PG Eller and WR Heinemann.

Morgan LM, WW Schulz, MR Adams, and KW Owens. 1988. *Summary of Single-Shell Tank Waste Characterization: 1985 to 1987*. WHC-EP-0075, Westinghouse Hanford Company, Richland, WA.

Nash CA, MM Morse, DJ McCabe, LL Hamm, and FG Smith. 2013. *Ion Exchange Column Tests Supporting Technetium Removal Resin Maturation*. SRNL-STI-2013-00573, Rev. 1, Savannah River National Laboratory, Aiken, SC.

Nguyen DM. 2010. *Guidelines for Updating Best-Basis Inventory*. RPP-7625, Rev. 10, Washington River Protection Solutions LLC, Richland, WA.

Place DE. 2006. *Best-Basis Inventory Process Requirements*. RPP-7625, Rev. 6a, CH2M Hill Hanford Group, Inc., Richland, WA.

Puigh RJ and MI Wood. 2005. *Inventory Data Package for the 2005 Integrated Disposal Facility Performance Assessment*. RPP-20692, Rev. 1, CH2M Hill Company, Richland, WA.

Ramsey WG. 2012a. *Tc/LAW Melter Recycle Demonstration Project Update: February 2012.* RPP-RPT-51964, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.

Ramsey WG. 2012b. *Tc/LAW Melter Recycle Demonstration Project Summary: September 2012*. RPP-RPT-53649, Rev. 1, Washington River Protection Solutions LLC, Richland, WA.

Rapko BM. 2014. *Protocol for Identifying the Presence of and Understanding the Nature of Soluble, Non-pertechnetate Technetium in Hanford Tank Supernatants*. PNNL-23180; EMSP-RPT-020, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Rapko BM, SA Bryan, JL Bryant, S Chatterjee, MK Edwards, JY Houchin, T Janik, TG Levitskaia, JM Peterson, RA Peterson, SI Sinkov, FN Smith, and R Wittman. 2013a. *Development of a Chemistry-Based, Predictive Method for Determining the Amount of Non-Pertechnetate Technetium in the Hanford Tanks: FY2012 Progress Report*. PNNL-22173, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Rapko BM, SA Bryan, S Chatterjee, MK Edwards, TG Levitskaia, JM Peterson, RA Peterson, and SI Sinkov. 2013b. *Investigations into the Nature of Alkaline Soluble, Nor-Pertechnetate Technetium.* PNNL-22957, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Rattat D, AP Schubiger, HG Berke, H Schmalle, and R Alberto. 2001. "Dicarbonyl-Nitrosyl-Complexes of Rhenium (Re) and Technetium (Tc), a Potentially New Class of Compounds for the Direct Radiolabeling of Biomolecules." *Cancer Biotherapy and Radiopharmaceuticals* 16: 339-343.

Robbins RA and TH May. 2013. *Submerged Bed Scrubber Condensate Technetium Removal and Disposal Preconceptual Engineering Study*. RPP-RPT-55213, Rev. 0, Washington River Protection Solutions, LLC, Richland, WA.

Schroeder NC and KR Ashley. 2005. "Separation of Non-Pertechnetate Species from Hanford AN-107 Tank Waste." *Journal of Radioanalytical and Nuclear Chemistry* 263(3):567-73. doi: 10.1007/s10967-005-0625-9.

Schroeder NC, SD Radzinski, JR Ball, KR Ashley, SL Cobb, B Cutrell, JM Adams, C Johnson, and D Whitener. 1995. *Technetium Partitioning for the Hanford Tank Waste Remediation System: Anion Exchange Studies for Partitioning Technetium from Synthetic DSSF and DSS Simulants and Actual Hanford Waste (101-SY and 103-SY) Using Reilliex-HPQ Resin*. LA-UR-95-4440, Los Alamos National Laboratory, Los Alamos, NM.

Schroeder NC, DL Blanchard, Jr., and KR Ashley. 1998. *Fundamental Chemistry, Characterization, and Separation of Technetium Complexes in Hanford Waste*. EMSP-59990-98, Los Alamos National Laboratory, Los Alamos, NM.

Schroeder NC, SD Radzinski, KR Ashley, AP Truong, and GD Whitener. 2001. "Feed adjustment chemistry for Hanford 101-SY and 103-SY tank waste: Attempts to oxidize the non-pertechnetate species." *Journal of Radioanalytical and Nuclear Chemistry* 250:271–284.

Schroeder NC, KR Ashley, and DL Blanchard. 2002. *Fundamental Chemistry, Characterization, and Separation of Technetium Complexes in Hanford Waste*. DOE Project Number: 59990, Los Alamos National Laboratory, Los Alamos, NM.

Schroeder NC, JA Olivares, and KR Ashley. 2004. *Identification of Non-Pertechnetate Species in Hanford Tank Waste, Their Synthesis, Characterization, and Fundamentals Chemistry*. LA-UR-04-4277, Los Alamos National Laboratory, Los Alamos, NM.

Serne RJ, TE Jones, MJ Lindberg, HT Schaef, and KM Krupka. 2007. *Laboratory Scale Bismuth Phosphate Extraction Process Simulation to Track Fate of Fission Products*. PNNL-14120, Pacific Northwest National Laboratory, Richland, WA.

Shuh DK, NM Edelstein, CJ Burns, WW Lukens, JJ Bucher, MG Fickes, and BL Scott. 2001. *Final Report: Research Program to Investigate The Fundamental Chemistry of Technetium 10/1/97-10/1/00*. EMSP-60296, Lawrence Berkeley National Laboratory, Berkeley, CA.

Shuh DK, WW Lukens, and CJ Burns. 2003. *Final Report, Research Program to Investigate the Fundamental Chemistry of Technetium--10/2000- 9/2003*. LBNL-54240, Lawrence Berkeley National Laboratory, Berkeley, CA.

Taylor-Pashow KM, CA Nash, CL Crawford, DJ McCabe, and WR Wilmarth. 2014. *Laboratory Scoping Tests of Decontamination of Hanford Waste Treatment Plant Low Activity Waste Off-Gas Condensate Simulant*. SRNL-STI-2013-00719, Rev. 0, Savannah River National Laboratory, Aiken, SC.

Watrous. RA. 2002. *Activity of Fuel Batches Processed through Hanford Separations Plants, 1944 through 1989*. RPP-13489, Rev. 0, CH2MHILL Hanford Group, Inc., Richland, WA.

Yanochko RM and C Corcoran. 2012. *Waste Treatment and Immobilization Plant of Submerged Bed Scrubber Condensate Disposition Project.* RPP-53982-FP, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.

Yanochko RM, C Corcoran, GR Golcar, P Johnson, LM Nolan, and A Pajunen. 2012. *Submerged Bed Scrubber Condensate Disposal Preconceptual Engineering Study*. RPP-RPT-52321, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.

Appendix A

Additional Outputs from TWINS 1-27-2014

Appendix A

Additional Outputs from TWINS 1-27-2014

This appendix contains two large tables imported from Excel® that provide additional data on the ^{99}Tc inventory currently estimated to be contained in each phase within each of the 177 Hanford storage tanks. The concentration of ^{99}Tc in each phase is also listed in Table A.1. Table A.2 is a ranking from the highest inventory tank to the lowest inventory tank. Note that double-shell tanks (DSTs) hold the top 15 positions for highest inventories and 20 of the top 25 positions for tanks. Three single-shell tanks (SSTs) in the SX tank farm and TX-113 and AX-101 have the top five ^{99}Tc inventories among the SSTs.

These rankings may have importance for retrieval risk assessment should there be fluid losses that reach the vadose zone outside the tanks. Another consideration is knowledge of which tanks are considered to have leaked fluids in the past and may not be structurally sound. Known and suspected SST leakers are identified in reports referenced in Appendix D.

The complete TWINS data query from January 27, 2014 in Excel format is available from the authors if desired. It has too many columns to be easily reproduced as Word® tables.

Table A.1. ^{99}Tc Inventory, ^{99}Tc Concentration by Phase, and Phase Volumes in Each Tank

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-A-101	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	4.19E+01	TS	1.49	148	2.83E-01	µCi/mL
241-A-101	Saltcake Solid	A1-SltCk (Solid)	1.86E+02	TE	1.74	878	1.22E-01	µCi/g
241-A-101	Sludge (Liquid & Solid)	P2 (Solid)	1.30E+00	TE	1.61	11	7.36E-02	µCi/g
241-A-101	Total		2.29E+02	TS/TE				
241-A-102	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	1.22E+01	E	1.57	33	3.69E-01	µCi/mL
241-A-102	Saltcake Solid	A1-SltCk (Solid)	2.04E+01	TE	1.7	106	1.13E-01	µCi/g
241-A-102	Supernatant	A1-SltCk (Liquid)	4.43E+00	E	1.57	12	3.69E-01	µCi/mL
241-A-102	Total		3.70E+01	E/TE				
241-A-103	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	8.06E+01	E	1.51	338	2.38E-01	µCi/mL
241-A-103	Saltcake Solid	A1-SltCk (Solid)	1.74E+02	E	1.32	1071	1.23E-01	µCi/g
241-A-103	Sludge (Liquid & Solid)	AR (Solid)	4.75E-01	TE	1.34	8	4.43E-02	µCi/g
241-A-103	Supernatant	A1-SltCk (Liquid)	4.05E+00	E	1.51	17	2.38E-01	µCi/mL
241-A-103	Total		2.59E+02	E/TE				
241-A-104	Sludge Solid	AR (Solid)	1.36E+01	TE	0.95	102	1.41E-01	µCi/g
241-A-104	Sludge Solid	P1 (Solid)	4.99E-01	TE	0.95	4	1.32E-01	µCi/g
241-A-104	Total		1.41E+01	TE				
241-A-105	Sludge Solid	P2 (Solid)	2.82E+01	TE	1.54	139	1.32E-01	µCi/g
241-A-105	Total		2.82E+01	TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-A-106	Saltcake Solid	A1-SltCk (Solid)	5.45E+01	E	1.7	110	2.92E-01	µCi/g
241-A-106	Sludge (Liquid & Solid)	AR (Solid)	3.92E+01	E	1.7	79	2.92E-01	µCi/g
241-A-106	Sludge (Liquid & Solid)	SRR (Solid)	5.45E+01	E	1.7	110	2.92E-01	µCi/g
241-A-106	Total		1.48E+02	E				
241-AN-101	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	7.39E+00	TS	1.45	28	2.64E-01	µCi/mL
241-AN-101	Saltcake Solid	A1-SltCk (Solid)	1.52E+01	TE	1.58	90	1.07E-01	µCi/g
241-AN-101	Sludge (Liquid & Solid)	CWP1 (Solid)	1.92E+01	E	1.68	326	3.50E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	CWP2 (Solid)	1.35E+01	E	1.68	229	3.50E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	CWZr1 (Solid)	5.29E+00	E	1.68	90	3.50E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	HS (Solid)	1.14E-02	E	1.58	1	7.21E-03	µCi/g
241-AN-101	Sludge (Liquid & Solid)	NA (Sludge)	7.88E+00	E	1.68	134	3.50E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	NA C-104 HHR	5.63E-01	E	1.68	7	4.79E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	OWW3 (Solid)	6.06E+00	E	1.68	103	3.50E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	TFeCN (Solid)	4.13E+01	E	1.6	266	9.71E-02	µCi/g
241-AN-101	Sludge (Liquid & Solid)	TH2 (Solid)	4.70E+00	E	1.68	80	3.50E-02	µCi/g
241-AN-101	Sludge Interstitial Liquid	C-101 CWP1 (Liquid)	2.09E+00	S	1.16	52	4.02E-02	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AN-101	Sludge Interstitial Liquid	C-101 TBP (Liquid)	1.96E+00	S	1.16	34	5.75E-02	µCi/mL
241-AN-101	Sludge Solid	C-101 CWP1 (Solid)	9.06E-02	E	2.14	112	3.78E-04	µCi/g
241-AN-101	Sludge Solid	C-101 TBP (Solid)	2.96E-01	E	2.14	75	1.85E-03	µCi/g
241-AN-101	Supernatant	NA (Liquid)	7.59E+01	S	1.17	1402	5.41E-02	µCi/mL
241-AN-101	Total		2.01E+02	S/E/TS/TE				
241-AN-102	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	1.20E+02	E	1.53	584	1.35E-01	µCi/g
241-AN-102	Supernatant	NA (Liquid)	4.60E+02	S	1.44	3475	1.32E-01	µCi/mL
241-AN-102	Total		5.80E+02	S/E				
241-AN-103	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	4.00E+02	S	1.72	1638	1.42E-01	µCi/g
241-AN-103	Supernatant	A2-SltSlr (Liquid)	2.81E+02	S	1.48	1769	1.59E-01	µCi/mL
241-AN-103	Total		6.81E+02	S				
241-AN-104	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	3.32E+02	S	1.59	1566	1.33E-01	µCi/g
241-AN-104	Supernatant	A2-SltSlr (Liquid)	4.62E+02	S	1.4	2298	2.01E-01	µCi/mL
241-AN-104	Total		7.94E+02	S				
241-AN-105	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	5.34E+02	S	1.57	1925	1.77E-01	µCi/g
241-AN-105	Supernatant	A2-SltSlr (Liquid)	5.88E+02	S	1.42	2227	2.64E-01	µCi/mL
241-AN-105	Total		1.12E+03	S				
241-AN-106	Saltcake (Liquid & Solid)	NA (SltCk)	1.10E+01	TE	1.58	65	1.07E-01	µCi/g
241-AN-106	Saltcake (Liquid & Solid)	NA from C-110 (Solids)	3.62E-01	E	1.56	30	7.73E-03	µCi/g
241-AN-106	Sludge (Liquid & Solid)	1C from C-107 (Solid)	1.64E+01	E	1.72	288	3.32E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AN-106	Sludge (Liquid & Solid)	1C from C-108 (Solid)	9.56E-03	E	1.71	1	3.99E-03	µCi/g
241-AN-106	Sludge (Liquid & Solid)	1C from C-109 (Solids)	3.33E-03	E	1.71	5	3.67E-04	µCi/g
241-AN-106	Sludge (Liquid & Solid)	CWP2 from C-107 (Solid)	3.59E+00	E	1.72	63	3.31E-02	µCi/g
241-AN-106	Sludge (Liquid & Solid)	Garnet	0.00E+00	E	2.4	0	0.00E+00	µCi/g
241-AN-106	Sludge (Liquid & Solid)	NA (Sludge)	8.59E+01	E	1.71	903	5.56E-02	µCi/g
241-AN-106	Sludge (Liquid & Solid)	SRR from C-107 (Solid)	1.37E+01	E	1.72	239	3.32E-02	µCi/g
241-AN-106	Supernatant	NA (Liquid)	1.31E+01	E	1.09	1163	1.13E-02	µCi/mL
241-AN-106	Total		1.44E+02	E/TE				
241-AN-107	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	1.31E+02	E	1.48	912	9.72E-02	µCi/g
241-AN-107	Supernatant	A2-SltSlr (Liquid)	3.03E+02	S	1.42	3208	9.45E-02	µCi/mL
241-AN-107	Total		4.34E+02	S/E				
241-AP-101	Saltcake (Liquid & Solid)	NA (SltCk)	3.12E+01	E	1.61	126	1.54E-01	µCi/g
241-AP-101	Supernatant	NA (Liquid)	7.24E+02	E	1.39	4555	1.59E-01	µCi/mL
241-AP-101	Total		7.55E+02	E				
241-AP-102	Sludge (Liquid & Solid)	NA (Sludge)	1.34E+01	E	1.73	105	7.36E-02	µCi/g
241-AP-102	Supernatant	NA (Liquid)	6.90E+02	E	1.37	4212	1.64E-01	µCi/mL
241-AP-102	Total		7.03E+02	E				
241-AP-103	Saltcake (Liquid & Solid)	NA (SltCk)	3.80E+01	TS	1.5	198	1.28E-01	µCi/g
241-AP-103	Supernatant	NA (Liquid)	9.21E+02	E	1.38	4487	2.05E-01	µCi/mL
241-AP-103	Total		9.59E+02	E/TS				
241-AP-104	Saltcake (Liquid & Solid)	NA (SltCk)	9.37E+01	E	1.61	378	1.54E-01	µCi/g
241-AP-104	Supernatant	NA (Liquid)	6.13E+01	E	1.18	1643	3.73E-02	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AP-104	Total		1.55E+02	E				
241-AP-105	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	9.02E+01	TS	1.61	396	1.42E-01	µCi/g
241-AP-105	Supernatant	NA (Liquid)	8.60E+02	E	1.4	4321	1.99E-01	µCi/mL
241-AP-105	Total		9.51E+02	E/TS				
241-AP-106	Supernatant	NA (Liquid)	3.68E+02	E	1.21	4291	8.59E-02	µCi/mL
241-AP-106	Total		3.68E+02	E				
241-AP-107	Supernatant	NA (Liquid)	1.77E+02	S	1.21	1670	1.06E-01	µCi/mL
241-AP-107	Total		1.77E+02	S				
241-AP-108	Saltcake (Liquid & Solid)	NA (SltCk)	5.94E+01	E	1.57	425	8.90E-02	µCi/g
241-AP-108	Supernatant	NA (Liquid)	6.65E+02	E	1.42	4290	1.55E-01	µCi/mL
241-AP-108	Total		7.24E+02	E				
241-AW-101	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	3.50E+02	S	1.59	1403	1.57E-01	µCi/g
241-AW-101	Supernatant	A2-SltSlr (Liquid)	4.68E+02	S	1.47	2770	1.69E-01	µCi/mL
241-AW-101	Total		8.18E+02	S				
241-AW-102	Sludge (Liquid & Solid)	NA (Sludge)	8.63E-02	TE	1.6	196	2.75E-04	µCi/g
241-AW-102	Supernatant	NA (Liquid)	3.21E+02	E	1.22	3088	1.04E-01	µCi/mL
241-AW-102	Total		3.21E+02	E/TE				
241-AW-103	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	9.04E+00	TS	1.42	36	2.51E-01	µCi/mL
241-AW-103	Saltcake Solid	A1-SltCk (Solid)	4.45E+00	S	1.69	115	2.29E-02	µCi/g
241-AW-103	Sludge (Liquid & Solid)	CWZr2 (Solid)	4.10E+01	S	1.47	1060	2.63E-02	µCi/g
241-AW-103	Supernatant	NA (Liquid)	3.42E+02	E	1.24	2884	1.19E-01	µCi/mL
241-AW-103	Total		3.97E+02	S/E/TS				
241-AW-104	Saltcake Interstitial Liquid	A2-SltSlr (Liquid)	3.54E+01	TS	1.45	142	2.49E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AW-104	Saltcake Solid	A2-SltSlr (Solid)	1.37E+02	E	1.6	451	1.90E-01	µCi/g
241-AW-104	Sludge Interstitial Liquid	PL2 (Liquid)	1.98E+01	TS	1.33	115	1.72E-01	µCi/mL
241-AW-104	Sludge Solid	PL2 (Solid)	4.26E-03	TE	1.28	255	1.31E-05	µCi/g
241-AW-104	Supernatant	NA (Liquid)	5.67E+02	E	1.36	3070	1.85E-01	µCi/mL
241-AW-104	Total		7.59E+02	E/TS/TE				
241-AW-105	Sludge (Liquid & Solid)	CWZr2 (Solid)	3.99E+00	TE	1.33	841	3.57E-03	µCi/g
241-AW-105	Sludge (Liquid & Solid)	PL2 (Solid)	2.11E-03	TE	1.35	99	1.58E-05	µCi/g
241-AW-105	Supernatant	NA (Liquid)	5.35E+00	TE	1.06	592	9.04E-03	µCi/mL
241-AW-105	Total		9.35E+00	TE				
241-AW-106	Saltcake (Liquid & Solid)	A2-SltSlr (Solid)	3.36E+02	TS	1.77	1001	1.90E-01	µCi/g
241-AW-106	Supernatant	NA Lower (Liquid)	5.56E+01	S	1.4	285	1.95E-01	µCi/mL
241-AW-106	Supernatant	NA Upper (Liquid)	2.01E+02	S	1.24	3019	6.65E-02	µCi/mL
241-AW-106	Total		5.93E+02	S/TS				
241-AX-101	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	5.54E+01	TS	1.53	178	3.12E-01	µCi/mL
241-AX-101	Saltcake Solid	A1-SltCk (Solid)	2.34E+02	TE	1.73	1164	1.16E-01	µCi/g
241-AX-101	Sludge (Liquid & Solid)	SRR (Solid)	3.55E-01	TE	1.51	11	2.14E-02	µCi/g
241-AX-101	Total		2.90E+02	TS/TE				
241-AX-102	Saltcake (Liquid & Solid)	A1-SltCk (Solid)	5.42E+00	S	1.58	90	3.81E-02	µCi/g
241-AX-102	Sludge (Liquid & Solid)	B (Solid)	8.80E-01	TE	1.57	23	2.44E-02	µCi/g
241-AX-102	Total		6.30E+00	S/TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AX-103	Saltcake Interstitial Liquid	A1-SltCk (Liquid)	1.98E+01	TS	1.45	75	2.64E-01	µCi/mL
241-AX-103	Saltcake Solid	A1-SltCk (Solid)	4.69E+01	TE	1.61	298	9.78E-02	µCi/g
241-AX-103	Sludge (Liquid & Solid)	P2 (Solid)	3.55E+00	TE	1.61	30	7.36E-02	µCi/g
241-AX-103	Total		7.03E+01	TS/TE				
241-AX-104	Sludge Solid	P2 (Solid)	2.06E+01	S	1.8	28	4.08E-01	µCi/g
241-AX-104	Total		2.06E+01	S				
241-AY-101	Sludge Interstitial Liquid	NA (Liquid)	2.54E+00	S	1.29	60	4.23E-02	µCi/mL
241-AY-101	Sludge Solid	NA (Sludge)	1.37E+01	E	1.87	338	2.17E-02	µCi/g
241-AY-101	Supernatant	NA Lower (Liquid)	2.08E+01	S	1.11	1247	1.67E-02	µCi/mL
241-AY-101	Supernatant	NA Upper (Liquid)	1.84E+01	S	1.06	1977	9.30E-03	µCi/mL
241-AY-101	Total		5.55E+01	S/E				
241-AY-102	Sludge Interstitial Liquid	BL (Liquid)	1.54E+00	S	1.15	120	1.28E-02	µCi/mL
241-AY-102	Sludge Solid	BL (Solid)	3.81E+00	S	1.71	75	2.97E-02	µCi/g
241-AY-102	Sludge Solid	NA (Sludge)	1.91E+01	S	1.71	376	2.97E-02	µCi/g
241-AY-102	Supernatant	NA (Liquid)	1.58E+02	E	1.36	2538	6.23E-02	µCi/mL
241-AY-102	Total		1.82E+02	S/E				
241-AZ-101	Sludge (Liquid & Solid)	NA (Sludge)	7.73E+00	E	1.59	30	1.62E-01	µCi/g
241-AZ-101	Sludge (Liquid & Solid)	P3AZ1 (Solid)	4.30E+01	E	1.59	167	1.62E-01	µCi/g
241-AZ-101	Supernatant	NA (Liquid)	1.18E+03	S	1.27	3019	3.92E-01	µCi/mL
241-AZ-101	Total		1.23E+03	S/E				
241-AZ-102	Sludge (Liquid & Solid)	NA (Sludge)	1.75E+00	E	1.41	8	1.55E-01	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-AZ-102	Sludge (Liquid & Solid)	P3AZ2 (Solid)	6.02E+01	E	1.41	275	1.55E-01	µCi/g
241-AZ-102	Sludge (Liquid & Solid)	PL2 (Solid)	3.28E+00	E	1.41	15	1.55E-01	µCi/g
241-AZ-102	Sludge (Liquid & Solid)	SRR (Solid)	2.15E+01	E	1.41	98	1.55E-01	µCi/g
241-AZ-102	Supernatant	NA (Liquid)	3.14E+02	S	1.17	3356	9.36E-02	µCi/mL
241-AZ-102	Total		4.01E+02	S/E				
241-B-101	Saltcake Interstitial Liquid	B-SltCk (Liquid)	4.80E-01	TE	1.53	73	6.57E-03	µCi/mL
241-B-101	Saltcake Solid	B-SltCk (Solid)	2.35E-01	TE	1.48	232	6.83E-04	µCi/g
241-B-101	Sludge (Liquid & Solid)	B (Solid)	6.64E-01	TE	1.74	19	2.01E-02	µCi/g
241-B-101	Sludge (Liquid & Solid)	BL (Solid)	4.26E-01	TE	1.5	76	3.74E-03	µCi/g
241-B-101	Sludge (Liquid & Solid)	MW1 (Solid)	2.04E-02	TE	1.8	11	1.03E-03	µCi/g
241-B-101	Total		1.83E+00	TE				
241-B-102	Saltcake Interstitial Liquid	B-SltCk (Liquid)	8.20E-02	TE	1.26	25	3.28E-03	µCi/mL
241-B-102	Saltcake Solid	B-SltCk (Solid)	8.37E-02	TE	1.72	81	6.01E-04	µCi/g
241-B-102	Supernatant	B-SltCk (Liquid)	4.92E-02	TE	1.26	15	3.28E-03	µCi/mL
241-B-102	Total		2.15E-01	TE				
241-B-103	Saltcake Interstitial Liquid	B-SltCk (Liquid)	1.64E-01	TE	1.26	50	3.28E-03	µCi/mL
241-B-103	Saltcake Solid	B-SltCk (Solid)	1.62E-01	TE	1.72	157	6.01E-04	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-B-103	Sludge (Liquid & Solid)	MW1 (Solid)	7.42E-03	TE	1.8	4	1.03E-03	µCi/g
241-B-103	Total		3.34E-01	TE				
241-B-104	Saltcake Interstitial Liquid	B-SltCk (Liquid)	1.80E-01	TE	1.26	55	3.28E-03	µCi/mL
241-B-104	Saltcake Solid	B-SltCk (Solid)	1.43E-01	TE	1.39	192	5.35E-04	µCi/g
241-B-104	Sludge (Liquid & Solid)	1C (Solid)	1.86E+01	TS	1.39	473	2.82E-02	µCi/g
241-B-104	Sludge (Liquid & Solid)	2C (Solid)	1.50E-03	TE	1.39	697	1.54E-06	µCi/g
241-B-104	Total		1.89E+01	TS/TE				
241-B-105	Saltcake Interstitial Liquid	B-SltCk (Liquid)	2.39E-01	TE	1.26	73	3.28E-03	µCi/mL
241-B-105	Saltcake Solid	B-SltCk (Solid)	9.50E-01	TE	1.72	919	6.01E-04	µCi/g
241-B-105	Sludge (Liquid & Solid)	1C (Solid)	1.56E+00	TS	1.43	45	2.43E-02	µCi/g
241-B-105	Sludge (Liquid & Solid)	2C (Solid)	7.57E-05	TE	1.28	61	9.70E-07	µCi/g
241-B-105	Total		2.75E+00	TS/TE				
241-B-106	Sludge (Liquid & Solid)	1C (Solid)	5.30E+00	TS	1.42	163	2.29E-02	µCi/g
241-B-106	Sludge (Liquid & Solid)	TBP (Solid)	3.09E-01	TE	1.36	297	7.64E-04	µCi/g
241-B-106	Supernatant	B-SltCk (Liquid)	1.31E-02	TE	1.26	4	3.28E-03	µCi/mL
241-B-106	Total		5.62E+00	TS/TE				
241-B-107	Saltcake Interstitial Liquid	B-SltCk (Liquid)	3.33E-01	TE	1.34	68	4.89E-03	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-B-107	Saltcake Solid	B-SltCk (Solid)	2.47E-01	TE	1.7	216	6.73E-04	µCi/g
241-B-107	Sludge (Liquid & Solid)	1C (Solid)	1.42E+01	TS	1.63	285	3.07E-02	µCi/g
241-B-107	Sludge (Liquid & Solid)	CWP2 (Solid)	9.31E-03	TE	1.68	42	1.32E-04	µCi/g
241-B-107	Total		1.48E+01	TS/TE				
241-B-108	Saltcake Interstitial Liquid	B-SltCk (Liquid)	1.51E-01	TE	1.38	42	3.60E-03	µCi/mL
241-B-108	Saltcake Solid	B-SltCk (Solid)	2.35E-01	TE	1.72	204	6.70E-04	µCi/g
241-B-108	Sludge (Liquid & Solid)	CWP2 (Solid)	3.10E-02	TE	1.8	104	1.65E-04	µCi/g
241-B-108	Total		4.17E-01	TE				
241-B-109	Saltcake Interstitial Liquid	B-SltCk (Liquid)	1.34E-01	TE	1.26	41	3.28E-03	µCi/mL
241-B-109	Saltcake Solid	B-SltCk (Solid)	2.87E-01	TE	1.89	245	6.19E-04	µCi/g
241-B-109	Sludge (Liquid & Solid)	CWP2 (Solid)	4.98E-02	TE	1.85	189	1.43E-04	µCi/g
241-B-109	Total		4.71E-01	TE				
241-B-110	Sludge (Liquid & Solid)	2C (Solid)	2.05E+01	E	1.36	914	1.65E-02	µCi/g
241-B-110	Sludge (Liquid & Solid)	B (Solid)	2.47E-01	E	1.36	11	1.65E-02	µCi/g
241-B-110	Supernatant	CSR (Liquid)	5.76E-01	TE	1.19	4	1.44E-01	µCi/mL
241-B-110	Total		2.13E+01	E/TE				
241-B-111	Sludge (Liquid & Solid)	2C (Solid)	1.17E+02	S	1.27	809	1.14E-01	µCi/g
241-B-111	Sludge (Liquid & Solid)	B (Solid)	1.46E+01	S	1.27	101	1.14E-01	µCi/g
241-B-111	Supernatant	CSR (Liquid)	5.76E-01	TE	1.19	4	1.44E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-B-111	Total		1.32E+02	S/TE				
241-B-112	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	3.15E+00	TE	1.51	16	1.97E-01	µCi/mL
241-B-112	Saltcake Solid	BY-SltCk (Solid)	3.83E+00	TE	1.49	49	5.24E-02	µCi/g
241-B-112	Sludge (Liquid & Solid)	2C (Solid)	1.44E-04	TE	1.49	56	1.73E-06	µCi/g
241-B-112	Supernatant	BY-SltCk (Liquid)	2.17E+00	TE	1.51	11	1.97E-01	µCi/mL
241-B-112	Total		9.15E+00	TE				
241-B-201	Sludge (Liquid & Solid)	224-1 (Solid)	1.72E-06	TE	1.26	111	1.23E-08	µCi/g
241-B-201	Total		1.72E-06	TE				
241-B-202	Sludge (Liquid & Solid)	224-2 (Solid)	6.85E-01	S	1.22	108	5.20E-03	µCi/g
241-B-202	Total		6.85E-01	S				
241-B-203	Sludge (Liquid & Solid)	224-2 (Solid)	4.07E-06	TE	1.19	188	1.82E-08	µCi/g
241-B-203	Supernatant	NA (Liquid)	8.86E-08	TE	1.05	2	4.43E-08	µCi/mL
241-B-203	Total		4.16E-06	TE				
241-B-204	Sludge (Liquid & Solid)	224-2 (Solid)	3.72E-06	TE	1.19	184	1.70E-08	µCi/g
241-B-204	Supernatant	NA (Liquid)	6.58E-08	TE	1.05	3	2.19E-08	µCi/mL
241-B-204	Total		3.79E-06	TE				
241-BX-101	Sludge (Liquid & Solid)	BL (Solid)	4.25E+00	E	1.68	74	3.42E-02	µCi/g
241-BX-101	Sludge (Liquid & Solid)	CWP2 (Solid)	2.70E+00	E	1.68	47	3.42E-02	µCi/g
241-BX-101	Sludge (Liquid & Solid)	TBP (Solid)	3.39E+00	E	1.68	59	3.42E-02	µCi/g
241-BX-101	Total		1.03E+01	E				
241-BX-102	Sludge Solid	CWP2 (Solid)	1.80E-02	TE	1.68	81	1.32E-04	µCi/g
241-BX-102	Sludge Solid	DE (Solid)	0.00E+00	E	0.65	147	0.00E+00	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BX-102	Sludge Solid	TBP (Solid)	1.01E-01	TE	1.47	70	9.85E-04	µCi/g
241-BX-102	Total		1.19E-01	E/TE				
241-BX-103	Sludge (Liquid & Solid)	CWP2 (Solid)	4.05E-02	TE	1.68	214	1.13E-04	µCi/g
241-BX-103	Sludge (Liquid & Solid)	TBP (Solid)	3.10E-02	TE	1.47	21	1.00E-03	µCi/g
241-BX-103	Supernatant	CWP2 (Liquid)	1.05E-02	TE	1.07	50	2.09E-04	µCi/mL
241-BX-103	Total		8.19E-02	TE				
241-BX-104	Sludge (Liquid & Solid)	CWP2 (Solid)	6.25E+00	E	1.68	51	7.29E-02	µCi/g
241-BX-104	Sludge (Liquid & Solid)	CWR1 (Solid)	1.35E+01	E	1.68	110	7.29E-02	µCi/g
241-BX-104	Sludge (Liquid & Solid)	MW1 (Solid)	1.90E+01	E	1.68	155	7.29E-02	µCi/g
241-BX-104	Sludge (Liquid & Solid)	TBP (Solid)	6.49E+00	E	1.68	53	7.29E-02	µCi/g
241-BX-104	Supernatant	CWR1 (Liquid)	1.32E+00	E	1.28	11	1.20E-01	µCi/mL
241-BX-104	Total		4.65E+01	E				
241-BX-105	Saltcake (Liquid & Solid)	BY-SltCk (Solid)	2.24E+01	E	1.69	94	1.41E-01	µCi/g
241-BX-105	Sludge (Liquid & Solid)	CWP2 (Solid)	2.29E+01	E	1.69	96	1.41E-01	µCi/g
241-BX-105	Sludge (Liquid & Solid)	MW1 (Solid)	1.67E-02	TE	1.8	9	1.03E-03	µCi/g
241-BX-105	Sludge (Liquid & Solid)	TBP (Solid)	1.31E+01	E	1.69	55	1.41E-01	µCi/g
241-BX-105	Supernatant	CWP2 (Liquid)	2.05E+00	E	1.29	18	1.14E-01	µCi/mL
241-BX-105	Total		6.04E+01	E/TE				
241-BX-106	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	4.92E+00	TE	1.51	25	1.97E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BX-106	Saltcake Solid	BY-SltCk (Solid)	7.04E+00	TE	1.64	80	5.37E-02	µCi/g
241-BX-106	Sludge (Liquid & Solid)	CWP2 (Solid)	4.04E-03	TE	1.64	18	1.37E-04	µCi/g
241-BX-106	Sludge (Liquid & Solid)	TBP (Solid)	4.00E-02	TE	1.64	20	1.22E-03	µCi/g
241-BX-106	Total		1.20E+01	TE				
241-BX-107	Sludge (Liquid & Solid)	1C (Solid)	6.98E+01	S	1.44	1313	3.69E-02	µCi/g
241-BX-107	Total		6.98E+01	S				
241-BX-108	Sludge (Liquid & Solid)	1C (Solid)	2.38E+00	TS	1.43	38	4.38E-02	µCi/g
241-BX-108	Sludge (Liquid & Solid)	TBP (Solid)	1.96E-01	TE	1.47	81	1.65E-03	µCi/g
241-BX-108	Total		2.58E+00	TS/TE				
241-BX-109	Sludge (Liquid & Solid)	TBP (Solid)	1.09E+00	TE	1.52	730	9.83E-04	µCi/g
241-BX-109	Total		1.09E+00	TE				
241-BX-110	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.12E+01	TE	1.44	129	1.64E-01	µCi/mL
241-BX-110	Saltcake Solid	BY-SltCk (Solid)	4.30E+01	TE	1.79	433	5.55E-02	µCi/g
241-BX-110	Sludge (Liquid & Solid)	1C (Solid)	5.25E+00	TS	1.43	151	2.43E-02	µCi/g
241-BX-110	Sludge (Liquid & Solid)	NA (Sludge)	5.65E+00	TS	1.79	94	3.36E-02	µCi/g
241-BX-110	Supernatant	BY-SltCk (Liquid)	8.20E-01	TE	1.44	5	1.64E-01	µCi/mL
241-BX-110	Total		7.59E+01	TS/TE				
241-BX-111	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	9.28E+00	TE	1.45	54	1.72E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BX-111	Saltcake Solid	BY-SltCk (Solid)	5.76E+01	TE	1.45	538	7.39E-02	µCi/g
241-BX-111	Sludge (Liquid & Solid)	1C (Solid)	4.20E+00	TS	1.43	121	2.43E-02	µCi/g
241-BX-111	Total		7.11E+01	TS/TE				
241-BX-112	Sludge (Liquid & Solid)	1C (Solid)	1.57E+01	TS	1.31	617	1.94E-02	µCi/g
241-BX-112	Supernatant	1C2 (Liquid)	2.04E-03	TE	1.18	5	4.07E-04	µCi/mL
241-BX-112	Total		1.57E+01	TS/TE				
241-BY-101	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	1.02E+01	TE	1.51	52	1.97E-01	µCi/mL
241-BY-101	Saltcake Solid	BY-SltCk (Solid)	1.57E+02	TE	1.87	1208	6.97E-02	µCi/g
241-BY-101	Sludge (Liquid & Solid)	PFeCN (Solid)	1.94E-01	TE	1.68	140	8.24E-04	µCi/g
241-BY-101	Total		1.68E+02	TE				
241-BY-102	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.74E+01	TE	1.46	157	1.75E-01	µCi/mL
241-BY-102	Saltcake Solid	BY-SltCk (Solid)	9.01E+01	TE	1.59	897	6.32E-02	µCi/g
241-BY-102	Total		1.18E+02	TE				
241-BY-103	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.30E+01	TE	1.29	214	1.07E-01	µCi/mL
241-BY-103	Saltcake Solid	BY-SltCk (Solid)	1.55E+02	TE	1.72	1316	6.86E-02	µCi/g
241-BY-103	Sludge (Liquid & Solid)	CWP2 (Solid)	7.54E-03	TE	1.68	34	1.32E-04	µCi/g
241-BY-103	Total		1.78E+02	TE				
241-BY-104	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.87E+01	TE	1.51	153	1.87E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BY-104	Saltcake Solid	BY-SltCk (Solid)	1.43E+02	TE	1.75	1208	6.75E-02	µCi/g
241-BY-104	Sludge (Liquid & Solid)	PFeCN (Solid)	2.44E-01	TE	1.64	172	8.65E-04	µCi/g
241-BY-104	Total		1.72E+02	TE				
241-BY-105	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	1.35E+01	S	1.44	159	8.48E-02	µCi/mL
241-BY-105	Saltcake Solid	BY-SltCk (Solid)	3.26E+01	S	1.85	1481	1.19E-02	µCi/g
241-BY-105	Sludge (Liquid & Solid)	PFeCN (Solid)	2.30E-01	TE	1.68	151	9.08E-04	µCi/g
241-BY-105	Sludge (Liquid & Solid)	Portland Cement (Solid)	0.00E+00	E	1.9	30	0.00E+00	µCi/g
241-BY-105	Total		4.63E+01	S/E/TE				
241-BY-106	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	1.78E+01	TE	1.31	138	1.29E-01	µCi/mL
241-BY-106	Saltcake Solid	BY-SltCk (Solid)	1.54E+02	TE	1.68	1365	6.72E-02	µCi/g
241-BY-106	Sludge (Liquid & Solid)	PFeCN (Solid)	1.54E-01	TE	1.68	120	7.65E-04	µCi/g
241-BY-106	Total		1.72E+02	TE				
241-BY-107	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.46E+01	TE	1.46	135	1.82E-01	µCi/mL
241-BY-107	Saltcake Solid	BY-SltCk (Solid)	8.01E+01	TE	1.72	835	5.58E-02	µCi/g
241-BY-107	Sludge (Liquid & Solid)	PFeCN (Solid)	7.85E-02	TE	1.78	58	7.61E-04	µCi/g
241-BY-107	Total		1.05E+02	TE				
241-BY-108	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.24E+01	TE	1.33	103	2.17E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BY-108	Saltcake Solid	BY-SltCk (Solid)	5.74E+01	TE	1.5	587	6.52E-02	µCi/g
241-BY-108	Sludge (Liquid & Solid)	PFeCN (Solid)	1.94E-01	TE	1.53	151	8.41E-04	µCi/g
241-BY-108	Total		8.00E+01	TE				
241-BY-109	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.50E+01	TE	1.5	146	1.71E-01	µCi/mL
241-BY-109	Saltcake Solid	BY-SltCk (Solid)	7.39E+01	TE	1.71	851	5.08E-02	µCi/g
241-BY-109	Sludge (Liquid & Solid)	CWP2 (Solid)	2.85E-02	TE	2	89	1.60E-04	µCi/g
241-BY-109	Total		9.89E+01	TE				
241-BY-110	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.08E+01	TE	1.44	99	2.10E-01	µCi/mL
241-BY-110	Saltcake Solid	BY-SltCk (Solid)	1.10E+02	TE	1.54	1123	6.37E-02	µCi/g
241-BY-110	Sludge (Liquid & Solid)	PFeCN (Solid)	2.56E-01	TE	1.82	162	8.69E-04	µCi/g
241-BY-110	Total		1.31E+02	TE				
241-BY-111	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	2.31E+01	TE	1.42	145	1.59E-01	µCi/mL
241-BY-111	Saltcake Solid	BY-SltCk (Solid)	1.36E+02	TE	1.7	1378	5.81E-02	µCi/g
241-BY-111	Total		1.59E+02	TE				
241-BY-112	Saltcake Interstitial Liquid	BY-SltCk (Liquid)	1.45E+01	TE	1.47	79	1.84E-01	µCi/mL
241-BY-112	Saltcake Solid	BY-SltCk (Solid)	1.10E+02	TE	1.76	996	6.28E-02	µCi/g
241-BY-112	Sludge (Liquid & Solid)	MW2 (Solid)	1.34E-02	TE	1.85	8	9.04E-04	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-BY-112	Total		1.25E+02	TE				
241-C-101	Sludge (Liquid & Solid)	TBP (Solid)	4.34E-02	TE	1.78	16	1.52E-03	µCi/g
241-C-101	Supernatant	NA (Liquid)	0.00E+00	E	1	3	0.00E+00	µCi/mL
241-C-101	Total		4.34E-02	E/TE				
241-C-102	Sludge (Liquid & Solid)	CWP1 (Solid)	5.50E-02	TE	1.63	125	2.70E-04	µCi/g
241-C-102	Sludge (Liquid & Solid)	CWP2 (Solid)	1.90E-01	TE	1.74	855	1.27E-04	µCi/g
241-C-102	Sludge (Liquid & Solid)	CWZr1 (Solid)	3.37E-01	TE	1.74	38	5.10E-03	µCi/g
241-C-102	Sludge (Liquid & Solid)	MW1 (Solid)	3.52E-02	TE	1.8	19	1.03E-03	µCi/g
241-C-102	Sludge (Liquid & Solid)	TBP (Solid)	8.83E-02	TE	1.47	61	9.85E-04	µCi/g
241-C-102	Sludge (Liquid & Solid)	TH1 (Solid)	1.81E-01	TE	1.32	98	1.40E-03	µCi/g
241-C-102	Total		8.87E-01	TE				
241-C-103	Sludge (Liquid & Solid)	NA (Sludge)	4.47E-02	S	1.61	8.64	3.22E-03	µCi/g
241-C-103	Supernatant	NA (Liquid)	6.02E-05	S	0.98	0.93	6.48E-05	µCi/mL
241-C-103	Total		4.48E-02	S				
241-C-104	Sludge (Liquid & Solid)	NA (Sludge)	1.73E-01	S	1.58	4	2.67E-02	µCi/g
241-C-104	Supernatant	NA (Liquid)	0.00E+00	E	1	2	0.00E+00	µCi/mL
241-C-104	Total		1.73E-01	S/E				
241-C-105	Sludge (Liquid & Solid)	CWP1 (Solid)	7.32E+01	E	1.55	450	1.05E-01	µCi/g
241-C-105	Sludge (Liquid & Solid)	TBP (Solid)	8.14E+00	E	1.55	50	1.05E-01	µCi/g
241-C-105	Total		8.14E+01	E				
241-C-106	Sludge (Liquid & Solid)	NA (Sludge)	1.64E-01	S	1.56	10.166	1.04E-02	µCi/g
241-C-106	Supernatant	NA (Liquid)	3.44E-06	S	1.02	0.322	1.07E-05	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-C-106	Total		1.64E-01	S				
241-C-107	Sludge (Liquid & Solid)	1C (Solid)	3.99E+00	S	1.55	99	2.60E-02	µCi/g
241-C-107	Supernatant	Liquid (NA)	5.61E-03	E	1	33	1.70E-04	µCi/mL
241-C-107	Total		4.00E+00	S/E				
241-C-108	Sludge (Liquid & Solid)	NA (Solid)	3.96E-02	S	2.09	10.5	1.80E-03	µCi/g
241-C-108	Supernatant	NA (Liquid)	0.00E+00	E	1	0.7	0.00E+00	µCi/mL
241-C-108	Total		3.96E-02	S/E				
241-C-109	Sludge (Liquid & Solid)	1C (Solid)	7.50E-03	S	1.74	6.5	6.63E-04	µCi/g
241-C-109	Total		7.50E-03	S				
241-C-110	Saltcake (Liquid & Solid)	NA (SltCk)	5.77E-02	S	1.44	6.7	5.98E-03	µCi/g
241-C-110	Total		5.77E-02	S				
241-C-111	Sludge (Liquid & Solid)	1C (Solid)	1.70E+00	TS	1.43	49	2.43E-02	µCi/g
241-C-111	Sludge (Liquid & Solid)	CWP1 (Solid)	1.11E-02	TE	1.58	26	2.71E-04	µCi/g
241-C-111	Sludge (Liquid & Solid)	HS (Solid)	1.94E-01	TE	1.58	17	7.21E-03	µCi/g
241-C-111	Sludge (Liquid & Solid)	TFeCN (Solid)	2.80E-01	TE	1.58	40	4.42E-03	µCi/g
241-C-111	Total		2.19E+00	TS/TE				
241-C-112	Sludge (Liquid & Solid)	1C (Solid)	1.98E+00	TS	1.43	57	2.43E-02	µCi/g
241-C-112	Sludge (Liquid & Solid)	CWP1 (Solid)	7.61E+00	S	1.6	49	9.71E-02	µCi/g
241-C-112	Sludge (Liquid & Solid)	CWP2 (Solid)	1.71E+00	S	1.6	11	9.71E-02	µCi/g
241-C-112	Sludge (Liquid & Solid)	HS (Solid)	6.21E-01	S	1.6	4	9.71E-02	µCi/g
241-C-112	Sludge (Liquid & Solid)	TFeCN (Solid)	9.32E-01	S	1.6	6	9.71E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-C-112	Total		1.29E+01	S/TS				
241-C-201	Sludge Solid	HS (Solid)	2.63E-03	E	1.75	0.537	2.80E-03	µCi/g
241-C-201	Supernatant	NA (Liquid)	0.00E+00	E	1	0.007	0.00E+00	µCi/mL
241-C-201	Total		2.63E-03	E				
241-C-202	Sludge (Liquid & Solid)	HS (Solid)	2.50E-03	S	1.75	0.548	2.61E-03	µCi/g
241-C-202	Supernatant	NA (Liquid)	0.00E+00	E	1	0.009	0.00E+00	µCi/mL
241-C-202	Total		2.50E-03	S/E				
241-C-203	Sludge (Liquid & Solid)	HS (Solid)	2.32E-03	S	1.93	0.476	2.53E-03	µCi/g
241-C-203	Supernatant	NA (Liquid)	0.00E+00	E	1	0.048	0.00E+00	µCi/mL
241-C-203	Total		2.32E-03	S/E				
241-C-204	Sludge (Liquid & Solid)	HS (Solid)	3.18E-03	S	1.77	0.507	3.54E-03	µCi/g
241-C-204	Supernatant	NA (Liquid)	0.00E+00	E	1	0.011	0.00E+00	µCi/mL
241-C-204	Total		3.18E-03	S/E				
241-S-101	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	4.99E+00	S	1.47	16	3.12E-01	µCi/mL
241-S-101	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	3.43E+00	S	1.47	11	3.12E-01	µCi/mL
241-S-101	Saltcake Solid	S1-SltCk (Solid)	3.08E+01	TE	1.56	246	8.03E-02	µCi/g
241-S-101	Saltcake Solid	S2-SltSlr (Solid)	2.66E+01	TE	1.56	169	1.01E-01	µCi/g
241-S-101	Sludge (Liquid & Solid)	NA (Sludge)	4.02E+01	E	1.7	890	2.66E-02	µCi/g
241-S-101	Total		1.06E+02	S/E/TE				
241-S-102	Saltcake (Liquid & Solid)	NA (SltCk)	1.81E+01	E	1.69	219	4.89E-02	µCi/g
241-S-102	Sludge (Liquid & Solid)	R1 (Solid)	1.85E+00	TE	1.88	71	1.39E-02	µCi/g
241-S-102	Supernatant	NA (Liquid)	0.00E+00	E	1	9	0.00E+00	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-S-102	Total		2.00E+01	E/TE				
241-S-103	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.02E+01	TS	1.45	116	2.60E-01	µCi/mL
241-S-103	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	2.47E+01	TS	1.45	95	2.60E-01	µCi/mL
241-S-103	Saltcake Solid	S1-SltCk (Solid)	5.68E+01	TE	1.68	347	9.74E-02	µCi/g
241-S-103	Saltcake Solid	S2-SltSlr (Solid)	5.57E+01	TE	1.63	300	1.14E-01	µCi/g
241-S-103	Sludge (Liquid & Solid)	R1 (Solid)	8.30E-01	TE	1.77	34	1.38E-02	µCi/g
241-S-103	Supernatant	S2-SltSlr (Liquid)	1.04E+00	TS	1.45	4	2.60E-01	µCi/mL
241-S-103	Total		1.69E+02	TS/TE				
241-S-104	Saltcake Interstitial Liquid	R-SltCk (Liquid)	1.19E+01	TE	1.37	139	8.55E-02	µCi/mL
241-S-104	Saltcake Solid	R-SltCk (Solid)	1.86E+01	S	1.64	451	2.52E-02	µCi/g
241-S-104	Sludge (Liquid & Solid)	CWR1 (Solid)	3.76E+00	S	1.8	91	2.30E-02	µCi/g
241-S-104	Sludge (Liquid & Solid)	R1 (Solid)	1.69E+01	S	1.77	409	2.34E-02	µCi/g
241-S-104	Total		5.12E+01	S/TE				
241-S-105	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	4.14E+01	TS	1.45	157	2.64E-01	µCi/mL
241-S-105	Saltcake Solid	S1-SltCk (Solid)	3.10E+02	TE	1.68	1371	1.35E-01	µCi/g
241-S-105	Sludge (Liquid & Solid)	R1 (Solid)	1.95E-01	TE	1.77	8	1.38E-02	µCi/g
241-S-105	Total		3.52E+02	TS/TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-S-106	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	2.32E+01	TS	1.43	99	2.34E-01	µCi/mL
241-S-106	Saltcake Solid	S1-SltCk (Solid)	2.62E+02	TE	1.74	1624	9.28E-02	µCi/g
241-S-106	Total		2.85E+02	TS/TE				
241-S-107	Saltcake Solid	S1-SltCk (Solid)	7.61E+00	TE	1.57	63	7.69E-02	µCi/g
241-S-107	Saltcake Solid	S2-SltSlr (Solid)	4.54E+00	TE	1.57	30	9.65E-02	µCi/g
241-S-107	Saltcake Solid	T2-SltCk (Solid)	4.13E+00	TE	1.57	52	5.06E-02	µCi/g
241-S-107	Sludge (Liquid & Solid)	CWR1 (Solid)	2.09E+00	TS	1.8	447	2.60E-03	µCi/g
241-S-107	Sludge (Liquid & Solid)	CWR2 (Solid)	1.18E-01	TE	1.8	211	3.11E-04	µCi/g
241-S-107	Sludge (Liquid & Solid)	CWZr1 (Solid)	2.16E+00	TE	1.8	91	1.32E-02	µCi/g
241-S-107	Sludge (Liquid & Solid)	R1 (Solid)	9.94E+00	TE	1.8	462	1.20E-02	µCi/g
241-S-107	Total		3.06E+01	TS/TE				
241-S-108	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	8.18E+00	TS	1.45	31	2.64E-01	µCi/mL
241-S-108	Saltcake Solid	S1-SltCk (Solid)	3.32E+02	TE	1.68	2032	9.74E-02	µCi/g
241-S-108	Sludge (Liquid & Solid)	R1 (Solid)	4.64E-01	TE	1.77	19	1.38E-02	µCi/g
241-S-108	Total		3.41E+02	TS/TE				
241-S-109	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	1.58E+01	TS	1.49	63	2.50E-01	µCi/mL
241-S-109	Saltcake Solid	S1-SltCk (Solid)	4.01E+02	TE	1.66	1905	1.27E-01	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-S-109	Sludge (Liquid & Solid)	R1 (Solid)	1.20E+00	TE	1.77	49	1.38E-02	µCi/g
241-S-109	Total		4.18E+02	TS/TE				
241-S-110	Saltcake Interstitial Liquid	S1-Sltck (Liquid)	2.98E+01	TS	1.43	115	2.59E-01	µCi/mL
241-S-110	Saltcake Solid	S1-Sltck (Solid)	2.00E+02	TE	1.65	994	1.22E-01	µCi/g
241-S-110	Sludge (Liquid & Solid)	CWR1 (Solid)	3.58E-01	TS	1.77	76	2.66E-03	µCi/g
241-S-110	Sludge (Liquid & Solid)	R1 (Solid)	6.23E+00	TE	1.77	288	1.22E-02	µCi/g
241-S-110	Total		2.37E+02	TS/TE				
241-S-111	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.15E+01	TS	1.45	124	2.54E-01	µCi/mL
241-S-111	Saltcake Solid	S1-SltCk (Solid)	1.56E+02	TE	1.53	1028	9.90E-02	µCi/g
241-S-111	Sludge (Liquid & Solid)	CWR1 (Solid)	1.98E-01	TS	1.67	38	3.13E-03	µCi/g
241-S-111	Sludge (Liquid & Solid)	R1 (Solid)	4.98E+00	TE	1.67	207	1.44E-02	µCi/g
241-S-111	Total		1.92E+02	TS/TE				
241-S-112	Sludge (Liquid & Solid)	R1 (Solid)	1.37E-01	S	1.9	8.574	8.39E-03	µCi/g
241-S-112	Supernatant	NA (liquid)	0.00E+00	E	1.27	0.469	0.00E+00	µCi/mL
241-S-112	Total		1.37E-01	S/E				
241-SX-101	Saltcake Interstitial Liquid	R-SltCk (Liquid)	4.29E+00	TE	1.48	41	1.05E-01	µCi/mL
241-SX-101	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	6.74E+00	TS	1.48	25	2.70E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-SX-101	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	3.77E+00	TS	1.48	14	2.70E-01	µCi/mL
241-SX-101	Saltcake Solid	R-SltCk (Solid)	4.81E+01	TE	1.69	497	5.73E-02	µCi/g
241-SX-101	Saltcake Solid	S1-SltCk (Solid)	5.19E+01	TE	1.69	296	1.04E-01	µCi/g
241-SX-101	Saltcake Solid	S2-SltSlr (Solid)	3.66E+01	TE	1.69	166	1.30E-01	µCi/g
241-SX-101	Sludge (Liquid & Solid)	R1 (Solid)	1.22E+01	TE	1.69	545	1.33E-02	µCi/g
241-SX-101	Total		1.64E+02	TS/TE				
241-SX-102	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	2.85E+01	TS	1.46	105	2.71E-01	µCi/mL
241-SX-102	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	2.98E+00	TS	1.46	11	2.71E-01	µCi/mL
241-SX-102	Saltcake Solid	S1-SltCk (Solid)	1.29E+02	TE	1.72	875	8.58E-02	µCi/g
241-SX-102	Saltcake Solid	S2-SltSlr (Solid)	1.70E+01	TE	1.72	92	1.08E-01	µCi/g
241-SX-102	Sludge (Liquid & Solid)	R1 (Solid)	3.95E+00	TE	1.72	209	1.10E-02	µCi/g
241-SX-102	Total		1.82E+02	TS/TE				
241-SX-103	Saltcake Interstitial Liquid	R-SltCk (Liquid)	2.40E+00	TE	1.47	22	1.09E-01	µCi/mL
241-SX-103	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	2.64E+01	TS	1.47	94	2.81E-01	µCi/mL
241-SX-103	Saltcake Solid	R-SltCk (Solid)	6.40E+00	TE	1.72	77	4.83E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-SX-103	Saltcake Solid	S1-Sltck (Solid)	2.39E+02	TE	1.72	1439	9.65E-02	µCi/g
241-SX-103	Sludge (Liquid & Solid)	R1 (Solid)	7.71E+00	TE	1.88	294	1.39E-02	µCi/g
241-SX-103	Total		2.82E+02	TS/TE				
241-SX-104	Saltcake Interstitial Liquid	R-SltCk (Liquid)	1.90E+00	TE	1.47	17	1.12E-01	µCi/mL
241-SX-104	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.19E+01	TS	1.47	111	2.87E-01	µCi/mL
241-SX-104	Saltcake Solid	R-SltCk (Solid)	1.44E+01	TE	1.72	142	5.90E-02	µCi/g
241-SX-104	Saltcake Solid	S1-SltCk (Solid)	1.48E+02	TE	1.68	903	9.74E-02	µCi/g
241-SX-104	Sludge (Liquid & Solid)	R1 (Solid)	1.26E+01	TE	1.77	515	1.38E-02	µCi/g
241-SX-104	Total		2.09E+02	TS/TE				
241-SX-105	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.69E+01	TS	1.47	123	3.00E-01	µCi/mL
241-SX-105	Saltcake Solid	S1-SltCk (Solid)	1.53E+02	TE	1.64	1061	8.78E-02	µCi/g
241-SX-105	Sludge (Liquid & Solid)	R1 (Solid)	4.36E+00	TE	1.67	189	1.38E-02	µCi/g
241-SX-105	Sludge (Liquid & Solid)	R2 (Solid)	3.10E+00	TE	1.67	49	3.79E-02	µCi/g
241-SX-105	Total		1.97E+02	TS/TE				
241-SX-106	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	6.30E+00	TS	1.29	27	2.33E-01	µCi/mL
241-SX-106	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	2.85E+01	TS	1.29	122	2.33E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-SX-106	Saltcake Solid	S1-SltCk (Solid)	3.36E+01	TE	1.61	245	8.52E-02	µCi/g
241-SX-106	Saltcake Solid	S2-SltSlr (Solid)	1.90E+02	TE	1.61	1107	1.07E-01	µCi/g
241-SX-106	Total		2.59E+02	TS/TE				
241-SX-107	Sludge (Liquid & Solid)	R1 (Solid)	5.84E+00	TE	1.77	239	1.38E-02	µCi/g
241-SX-107	Sludge (Liquid & Solid)	R2 (Solid)	7.85E+00	TE	1.77	117	3.79E-02	µCi/g
241-SX-107	Total		1.37E+01	TE				
241-SX-108	Sludge Solid	R1 (Solid)	5.76E+00	TE	1.77	186	1.75E-02	µCi/g
241-SX-108	Sludge Solid	R2 (Solid)	8.00E+00	TE	1.77	94	4.81E-02	µCi/g
241-SX-108	Total		1.38E+01	TE				
241-SX-109	Saltcake Solid	R-SltCk (Solid)	6.72E+01	TE	1.72	662	5.90E-02	µCi/g
241-SX-109	Sludge Solid	R1 (Solid)	4.15E+00	TE	1.77	170	1.38E-02	µCi/g
241-SX-109	Sludge Solid	R2 (Solid)	5.44E+00	TE	1.77	81	3.79E-02	µCi/g
241-SX-109	Total		7.68E+01	TE				
241-SX-110	Saltcake Solid	R-SltCk (Solid)	2.84E+00	TE	1.72	28	5.90E-02	µCi/g
241-SX-110	Sludge Solid	R2 (Solid)	1.24E+01	TE	1.77	184	3.79E-02	µCi/g
241-SX-110	Total		1.52E+01	TE				
241-SX-111	Saltcake Solid	R-SltCk (Solid)	6.80E+00	TE	1.72	67	5.90E-02	µCi/g
241-SX-111	Sludge (Liquid & Solid)	R1 (Solid)	4.01E+00	TE	1.77	164	1.38E-02	µCi/g
241-SX-111	Sludge (Liquid & Solid)	R2 (Solid)	1.38E+01	TE	1.77	205	3.79E-02	µCi/g
241-SX-111	Total		2.46E+01	TE				
241-SX-112	Sludge (Liquid & Solid)	R1 (Solid)	3.52E+00	TE	1.77	144	1.38E-02	µCi/g
241-SX-112	Sludge (Liquid & Solid)	R2 (Solid)	9.33E+00	TE	1.77	139	3.79E-02	µCi/g
241-SX-112	Total		1.28E+01	TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-SX-113	Sludge Solid	DE (Solid)	1.36E+00	TE	1.43	64	1.48E-02	µCi/g
241-SX-113	Sludge Solid	R1 (Solid)	1.95E-01	TE	1.43	8	1.71E-02	µCi/g
241-SX-113	Total		1.55E+00	TE				
241-SX-114	Saltcake Interstitial Liquid	R-SltCk (Liquid)	2.60E+00	TE	1.5	26	1.00E-01	µCi/mL
241-SX-114	Saltcake Solid	R-SltCk (Solid)	8.52E+00	TE	1.72	84	5.90E-02	µCi/g
241-SX-114	Sludge (Liquid & Solid)	R1 (Solid)	7.28E+00	TE	1.77	298	1.38E-02	µCi/g
241-SX-114	Sludge (Liquid & Solid)	R2 (Solid)	1.21E+01	TE	1.77	180	3.79E-02	µCi/g
241-SX-114	Total		3.05E+01	TE				
241-SX-115	Sludge Solid	R2 (Solid)	1.25E+00	TE	1.77	16	4.41E-02	µCi/g
241-SX-115	Total		1.25E+00	TE				
241-SY-101	Saltcake (Liquid & Solid)	S2-SltSlr (Solid)	2.58E+02	S	1.66	878	1.77E-01	µCi/g
241-SY-101	Supernatant	NA (Liquid)	4.04E+01	E	1.13	3246	1.25E-02	µCi/mL
241-SY-101	Total		2.98E+02	S/E				
241-SY-102	Sludge Interstitial Liquid	NA (Liquid)	1.79E+01	E	1.28	151	1.18E-01	µCi/mL
241-SY-102	Sludge Interstitial Liquid	Z (Liquid)	1.58E+01	S	1.36	82	1.93E-01	µCi/mL
241-SY-102	Sludge Solid	NA (Sludge)	1.12E+02	E	1.65	336	2.02E-01	µCi/g
241-SY-102	Sludge Solid	Z (Solid)	6.10E+01	S	1.65	183	2.02E-01	µCi/g
241-SY-102	Supernatant	NA (Liquid)	3.70E+01	E	1.15	1382	2.67E-02	µCi/mL
241-SY-102	Total		2.44E+02	S/E				
241-SY-103	Saltcake (Liquid & Solid)	S2-SltSlr (Solid)	4.92E+02	S	1.67	1258	2.34E-01	µCi/g
241-SY-103	Supernatant	S2-SltSlr (Liquid)	5.19E+02	E	1.49	1444	3.59E-01	µCi/mL
241-SY-103	Total		1.01E+03	S/E				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-T-101	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	1.50E+01	TS	1.45	57	2.64E-01	µCi/mL
241-T-101	Saltcake Solid	T2-SltCk (Solid)	1.93E+01	TE	1.64	179	6.58E-02	µCi/g
241-T-101	Sludge (Liquid & Solid)	CWR2 (Solid)	4.58E-02	TE	1.46	140	2.24E-04	µCi/g
241-T-101	Total		3.44E+01	TS/TE				
241-T-102	Sludge (Liquid & Solid)	CWP2 (Solid)	1.49E+00	S	1.79	64	1.30E-02	µCi/g
241-T-102	Sludge (Liquid & Solid)	MW2 (Solid)	1.34E-02	TE	1.85	8	9.04E-04	µCi/g
241-T-102	Supernatant	CSR (Liquid)	5.63E+00	TE	1.14	48	1.17E-01	µCi/mL
241-T-102	Total		7.13E+00	S/TE				
241-T-103	Sludge (Liquid & Solid)	CWP2 (Solid)	1.42E-02	TE	1.68	64	1.32E-04	µCi/g
241-T-103	Sludge (Liquid & Solid)	CWR1 (Solid)	1.00E-01	TS	1.8	19	2.93E-03	µCi/g
241-T-103	Sludge (Liquid & Solid)	MW2 (Solid)	6.69E-03	TE	1.85	4	9.04E-04	µCi/g
241-T-103	Supernatant	CSR (Liquid)	2.16E+00	TE	1.19	15	1.44E-01	µCi/mL
241-T-103	Total		2.28E+00	TS/TE				
241-T-104	Sludge (Liquid & Solid)	1C (Solid)	9.74E-01	S	1.29	1199	6.30E-04	µCi/g
241-T-104	Total		9.74E-01	S/E				
241-T-105	Sludge (Liquid & Solid)	1C (Solid)	2.14E+00	S	1.32	6	2.70E-01	µCi/g
241-T-105	Sludge (Liquid & Solid)	2C (Solid)	5.55E-04	TE	1.51	273	1.35E-06	µCi/g
241-T-105	Sludge (Liquid & Solid)	CWR1 (Solid)	3.28E+01	S	1.32	92	2.70E-01	µCi/g
241-T-105	Total		3.50E+01	S/TE				
241-T-106	Sludge (Liquid & Solid)	1C (Solid)	2.66E+00	TS	1.43	38	4.90E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-T-106	Sludge (Liquid & Solid)	CWR1 (Solid)	1.75E-01	TS	1.8	34	2.86E-03	µCi/g
241-T-106	Sludge (Liquid & Solid)	CWR2 (Solid)	6.15E-03	TE	1.46	10	4.22E-04	µCi/g
241-T-106	Total		2.85E+00	TS/TE				
241-T-107	Sludge (Liquid & Solid)	1C (Solid)	4.12E+01	S	1.56	559	4.73E-02	µCi/g
241-T-107	Sludge (Liquid & Solid)	CWP2 (Solid)	2.36E+00	S	1.56	32	4.73E-02	µCi/g
241-T-107	Sludge (Liquid & Solid)	TBP (Solid)	4.72E+00	S	1.56	64	4.73E-02	µCi/g
241-T-107	Total		4.83E+01	S				
241-T-108	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	3.28E-02	TE	1.26	10	3.28E-03	µCi/mL
241-T-108	Saltcake Solid	T1-SltCk (Solid)	4.40E-02	TE	1.72	30	8.54E-04	µCi/g
241-T-108	Sludge (Liquid & Solid)	1C (Solid)	6.95E-01	TS	1.43	20	2.43E-02	µCi/g
241-T-108	Total		7.72E-01	TS/TE				
241-T-109	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	1.25E-01	TE	1.26	38	3.28E-03	µCi/mL
241-T-109	Saltcake Solid	T1-SltCk (Solid)	1.77E-01	TE	1.72	197	5.24E-04	µCi/g
241-T-109	Total		3.02E-01	TE				
241-T-110	Sludge (Liquid & Solid)	224-2 (Solid)	8.49E-07	TE	1.25	37	1.84E-08	µCi/g
241-T-110	Sludge (Liquid & Solid)	2C (Solid)	1.21E-03	TE	1.25	1360	7.09E-07	µCi/g
241-T-110	Supernatant	2C1 (Liquid)	9.02E-06	TE	1.05	3	3.01E-06	µCi/mL
241-T-110	Total		1.22E-03	TE				
241-T-111	Sludge (Liquid & Solid)	224-2 (Solid)	8.87E+00	S	1.24	904	7.92E-03	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-T-111	Sludge (Liquid & Solid)	2C (Solid)	7.73E+00	S	1.24	787	7.92E-03	µCi/g
241-T-111	Total		1.66E+01	S				
241-T-112	Sludge (Liquid & Solid)	224-2 (Solid)	2.28E-06	TE	1.28	91	1.96E-08	µCi/g
241-T-112	Sludge (Liquid & Solid)	2C (Solid)	1.31E-04	TE	1.28	135	7.56E-07	µCi/g
241-T-112	Supernatant	NA (Liquid)	2.12E+00	E	1.1	27	7.87E-02	µCi/mL
241-T-112	Total		2.12E+00	E/TE				
241-T-201	Sludge (Liquid & Solid)	224-1 (Solid)	1.57E-06	TE	1.31	107	1.12E-08	µCi/g
241-T-201	Supernatant	224-1 (Liquid)	1.81E-07	TE	1.06	8	2.27E-08	µCi/mL
241-T-201	Total		1.75E-06	TE				
241-T-202	Sludge (Liquid & Solid)	224-2 (Solid)	1.65E-06	TE	1.18	77	1.81E-08	µCi/g
241-T-202	Total		1.65E-06	TE				
241-T-203	Sludge (Liquid & Solid)	224-2 (Solid)	2.96E-06	TE	1.22	136	1.78E-08	µCi/g
241-T-203	Total		2.96E-06	TE				
241-T-204	Sludge (Liquid & Solid)	224-2 (Solid)	3.00E-06	TE	1.18	136	1.87E-08	µCi/g
241-T-204	Total		3.00E-06	TE				
241-TX-101	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	3.17E+00	TS	1.45	12	2.64E-01	µCi/mL
241-TX-101	Saltcake Solid	T2-SltCk (Solid)	4.21E+00	TE	1.64	39	6.58E-02	µCi/g
241-TX-101	Sludge (Liquid & Solid)	MW2 (Solid)	1.84E-02	TE	1.85	11	9.04E-04	µCi/g
241-TX-101	Sludge (Liquid & Solid)	R1 (Solid)	6.47E+00	TE	1.77	265	1.38E-02	µCi/g
241-TX-101	Sludge (Liquid & Solid)	Z (Solid)	0.00E+00	TE	1.76	1	0.00E+00	µCi/g
241-TX-101	Total		1.39E+01	TS/TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TX-102	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	3.19E+01	TS	1.45	121	2.64E-01	µCi/mL
241-TX-102	Saltcake Solid	T2-SltCk (Solid)	7.47E+01	TE	1.64	692	6.58E-02	µCi/g
241-TX-102	Sludge (Liquid & Solid)	MW2 (Solid)	1.34E-02	TE	1.85	8	9.04E-04	µCi/g
241-TX-102	Total		1.07E+02	TS/TE				
241-TX-103	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	9.84E-03	TE	1.26	3	3.28E-03	µCi/mL
241-TX-103	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	2.16E+01	TS	1.45	82	2.64E-01	µCi/mL
241-TX-103	Saltcake Solid	T1-SltCk (Solid)	1.19E-02	TE	1.72	9	7.69E-04	µCi/g
241-TX-103	Saltcake Solid	T2-SltCk (Solid)	4.90E+01	TE	1.64	454	6.58E-02	µCi/g
241-TX-103	Total		7.07E+01	TS/TE				
241-TX-104	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	7.43E+00	TS	1.44	30	2.48E-01	µCi/mL
241-TX-104	Saltcake Solid	T2-SltCk (Solid)	7.58E+00	TE	1.62	93	5.03E-02	µCi/g
241-TX-104	Sludge (Liquid & Solid)	R1 (Solid)	2.43E+00	TE	1.89	130	9.89E-03	µCi/g
241-TX-104	Supernatant	T2-SltCk (Liquid)	2.23E+00	TS	1.44	9	2.48E-01	µCi/mL
241-TX-104	Total		1.97E+01	TS/TE				
241-TX-105	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	2.64E+01	TS	1.45	100	2.64E-01	µCi/mL
241-TX-105	Saltcake Solid	T2-SltCk (Solid)	2.18E+02	TE	1.64	2020	6.58E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TX-105	Sludge (Liquid & Solid)	MW2 (Solid)	5.18E-02	TE	1.85	31	9.04E-04	µCi/g
241-TX-105	Sludge (Liquid & Solid)	Z (Solid)	0.00E+00	TE	1.76	11	0.00E+00	µCi/g
241-TX-105	Total		2.44E+02	TS/TE				
241-TX-106	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	4.01E+01	TS	1.45	152	2.64E-01	µCi/mL
241-TX-106	Saltcake Solid	T2-SltCk (Solid)	1.24E+02	TE	1.64	1147	6.58E-02	µCi/g
241-TX-106	Sludge (Liquid & Solid)	MW2 (Solid)	6.69E-03	TE	1.85	4	9.04E-04	µCi/g
241-TX-106	Sludge (Liquid & Solid)	R1 (Solid)	3.66E-01	TE	1.77	15	1.38E-02	µCi/g
241-TX-106	Total		1.64E+02	TS/TE				
241-TX-107	Saltcake Interstitial Liquid	R-SltCk (Liquid)	7.00E-01	TE	1.5	7	1.00E-01	µCi/mL
241-TX-107	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	5.02E+00	TS	1.45	19	2.64E-01	µCi/mL
241-TX-107	Saltcake Solid	R-SltCk (Solid)	2.44E+00	TE	1.72	24	5.90E-02	µCi/g
241-TX-107	Saltcake Solid	T2-SltCk (Solid)	8.59E+00	TE	1.94	62	7.14E-02	µCi/g
241-TX-107	Total		1.67E+01	TS/TE				
241-TX-108	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	1.06E+01	TS	1.45	40	2.64E-01	µCi/mL
241-TX-108	Saltcake Solid	T2-SltCk (Solid)	4.48E+01	TE	1.64	415	6.58E-02	µCi/g
241-TX-108	Sludge (Liquid & Solid)	MW2 (Solid)	1.34E-02	TE	1.85	8	9.04E-04	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TX-108	Sludge (Liquid & Solid)	TBP (Solid)	2.17E-02	TE	1.47	15	9.85E-04	µCi/g
241-TX-108	Total		5.54E+01	TS/TE				
241-TX-109	Sludge (Liquid & Solid)	1C (Solid)	4.65E+01	TS	1.43	1339	2.43E-02	µCi/g
241-TX-109	Sludge (Liquid & Solid)	Z (Solid)	0.00E+00	TE	1.76	19	0.00E+00	µCi/g
241-TX-109	Total		4.65E+01	TS/TE				
241-TX-110	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	1.29E+01	TS	1.45	49	2.64E-01	µCi/mL
241-TX-110	Saltcake Solid	T2-SltCk (Solid)	1.70E+02	TE	1.64	1580	6.58E-02	µCi/g
241-TX-110	Sludge (Liquid & Solid)	1C (Solid)	4.86E+00	TS	1.43	140	2.43E-02	µCi/g
241-TX-110	Total		1.88E+02	TS/TE				
241-TX-111	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	6.34E+00	TS	1.45	24	2.64E-01	µCi/mL
241-TX-111	Saltcake Solid	T2-SltCk (Solid)	1.29E+02	TE	1.64	1194	6.58E-02	µCi/g
241-TX-111	Sludge (Liquid & Solid)	1C (Solid)	5.66E+00	TS	1.43	163	2.43E-02	µCi/g
241-TX-111	Total		1.41E+02	TS/TE				
241-TX-112	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	4.92E-02	TE	1.26	15	3.28E-03	µCi/mL
241-TX-112	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	2.46E+01	TS	1.45	93	2.64E-01	µCi/mL
241-TX-112	Saltcake Solid	T1-SltCk (Solid)	9.92E-02	TE	1.72	75	7.69E-04	µCi/g
241-TX-112	Saltcake Solid	T2-SltCk (Solid)	2.39E+02	TE	1.64	2215	6.58E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TX-112	Total		2.64E+02	TS/TE				
241-TX-113	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	5.28E+00	TS	1.45	20	2.64E-01	µCi/mL
241-TX-113	Saltcake Solid	T2-SltCk (Solid)	2.79E+02	TE	1.64	2045	8.31E-02	µCi/g
241-TX-113	Sludge (Liquid & Solid)	1C (Solid)	1.22E+01	TS	1.43	351	2.43E-02	µCi/g
241-TX-113	Total		2.96E+02	TS/TE				
241-TX-114	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	1.51E-01	TE	1.26	46	3.28E-03	µCi/mL
241-TX-114	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	7.39E+00	TS	1.45	28	2.64E-01	µCi/mL
241-TX-114	Saltcake Solid	T1-SltCk (Solid)	2.29E-01	TE	1.72	173	7.69E-04	µCi/g
241-TX-114	Saltcake Solid	T2-SltCk (Solid)	1.89E+02	TE	1.64	1750	6.58E-02	µCi/g
241-TX-114	Sludge (Liquid & Solid)	1C (Solid)	5.21E-01	TS	1.43	15	2.43E-02	µCi/g
241-TX-114	Total		1.97E+02	TS/TE				
241-TX-115	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	2.72E+01	TS	1.45	103	2.64E-01	µCi/mL
241-TX-115	Saltcake Solid	T2-SltCk (Solid)	2.12E+02	TE	1.64	1960	6.58E-02	µCi/g
241-TX-115	Sludge (Liquid & Solid)	TBP (Solid)	4.34E-02	TE	1.47	30	9.85E-04	µCi/g
241-TX-115	Total		2.39E+02	TS/TE				
241-TX-116	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	5.88E-01	TE	1.39	113	5.20E-03	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TX-116	Saltcake Solid	T1-SltCk (Solid)	1.56E+00	TE	1.72	977	9.29E-04	µCi/g
241-TX-116	Saltcake Solid	T2-SltCk (Solid)	9.96E+01	TE	1.64	926	6.56E-02	µCi/g
241-TX-116	Sludge (Liquid & Solid)	DE (Solid)	5.40E+01	TS	1.6	248	1.36E-01	µCi/g
241-TX-116	Total		1.56E+02	TS/TE				
241-TX-117	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	1.57E-01	TE	1.26	48	3.28E-03	µCi/mL
241-TX-117	Saltcake Solid	T1-SltCk (Solid)	8.28E-01	TE	1.72	626	7.69E-04	µCi/g
241-TX-117	Saltcake Solid	T2-SltCk (Solid)	1.71E+02	TE	1.64	1585	6.58E-02	µCi/g
241-TX-117	Sludge (Liquid & Solid)	DE (Solid)	2.40E+01	TS	1.58	110	1.38E-01	µCi/g
241-TX-117	Total		1.96E+02	TS/TE				
241-TX-118	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	3.67E+01	TS	1.45	139	2.64E-01	µCi/mL
241-TX-118	Saltcake Solid	NA (SltCk)	9.40E+00	TE	1.54	125	4.88E-02	µCi/g
241-TX-118	Saltcake Solid	T2-SltCk (Solid)	6.15E+01	TE	1.77	671	5.18E-02	µCi/g
241-TX-118	Total		1.08E+02	TS/TE				
241-TY-101	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	4.92E-02	TE	1.26	15	3.28E-03	µCi/mL
241-TY-101	Saltcake Solid	T1-SltCk (Solid)	1.92E+00	E	1.64	159	7.36E-03	µCi/g
241-TY-101	Sludge (Liquid & Solid)	1CFeCN (Solid)	3.30E+00	E	1.64	273	7.36E-03	µCi/g
241-TY-101	Total		5.26E+00	E/TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TY-102	Saltcake Interstitial Liquid	T1-SltCk (Liquid)	9.51E-02	TE	1.26	29	3.28E-03	µCi/mL
241-TY-102	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	8.98E+00	TS	1.45	34	2.64E-01	µCi/mL
241-TY-102	Saltcake Solid	T1-SltCk (Solid)	1.33E-01	E	1.88	90	7.88E-04	µCi/g
241-TY-102	Saltcake Solid	T2-SltCk (Solid)	1.61E-01	E	1.88	109	7.88E-04	µCi/g
241-TY-102	Total		9.37E+00	E/TS/TE				
241-TY-103	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	1.19E+01	TS	1.45	45	2.64E-01	µCi/mL
241-TY-103	Saltcake Solid	T2-SltCk (Solid)	4.22E+00	E	1.7	150	1.65E-02	µCi/g
241-TY-103	Sludge (Liquid & Solid)	1CFeCN (Solid)	4.78E+00	E	1.7	170	1.65E-02	µCi/g
241-TY-103	Sludge (Liquid & Solid)	TBP (Solid)	6.18E+00	E	1.7	220	1.65E-02	µCi/g
241-TY-103	Total		2.71E+01	E/TS				
241-TY-104	Sludge (Liquid & Solid)	1CFeCN (Solid)	6.02E+00	E	1.65	114	3.20E-02	µCi/g
241-TY-104	Sludge (Liquid & Solid)	TBP (Solid)	2.59E+00	E	1.65	49	3.20E-02	µCi/g
241-TY-104	Supernatant	DW (Liquid)	2.55E-01	E	1.18	5	5.11E-02	µCi/mL
241-TY-104	Total		8.86E+00	E				
241-TY-105	Sludge (Liquid & Solid)	TBP (Solid)	4.40E+01	E	1.53	874	3.29E-02	µCi/g
241-TY-105	Total		4.40E+01	E				
241-TY-106	Sludge (Liquid & Solid)	DE (Solid)	8.18E-01	S	1.4	47	1.24E-02	µCi/g
241-TY-106	Sludge (Liquid & Solid)	TBP (Solid)	2.61E-01	S	1.4	15	1.24E-02	µCi/g

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-TY-106	Total		1.08E+00	S				
241-U-101	Sludge (Liquid & Solid)	R1 (Solid)	1.93E+00	TE	1.77	87	1.25E-02	µCi/g
241-U-101	Total		1.93E+00	TE				
241-U-102	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	3.25E+01	TS	1.48	117	2.78E-01	µCi/mL
241-U-102	Saltcake Solid	S2-SltSlr (Solid)	5.90E+01	TE	1.68	307	1.14E-01	µCi/g
241-U-102	Saltcake Solid	T2-SltCk (Solid)	6.51E+01	TE	1.68	647	5.99E-02	µCi/g
241-U-102	Sludge (Liquid & Solid)	R1 (Solid)	3.98E+00	TE	1.77	163	1.38E-02	µCi/g
241-U-102	Supernatant	T2-SltCk (Liquid)	1.11E+00	TS	1.48	4	2.78E-01	µCi/mL
241-U-102	Total		1.62E+02	TS/TE				
241-U-103	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.07E+01	TS	1.44	119	2.58E-01	µCi/mL
241-U-103	Saltcake Solid	S1-SltCk (Solid)	1.32E+02	TE	1.72	891	8.60E-02	µCi/g
241-U-103	Saltcake Solid	S2-SltSlr (Solid)	6.60E+01	TE	1.72	356	1.08E-01	µCi/g
241-U-103	Sludge (Liquid & Solid)	R1 (Solid)	7.49E-01	TE	1.9	42	9.38E-03	µCi/g
241-U-103	Supernatant	S1-SltCk (Liquid)	5.16E-01	TS	1.44	2	2.58E-01	µCi/mL
241-U-103	Total		2.30E+02	TS/TE				
241-U-104	Sludge (Liquid & Solid)	DE (Solid)	7.21E-01	TE	1.11	201	3.23E-03	µCi/g
241-U-104	Sludge (Liquid & Solid)	MW2 (Solid)	6.69E-03	TE	1.85	4	9.04E-04	µCi/g
241-U-104	Total		7.28E-01	TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-U-105	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	3.28E+01	TS	1.46	130	2.53E-01	µCi/mL
241-U-105	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	8.84E+00	TS	1.46	35	2.53E-01	µCi/mL
241-U-105	Saltcake Solid	S2-SltSlr (Solid)	1.76E+02	TE	1.7	827	1.25E-01	µCi/g
241-U-105	Saltcake Solid	T2-SltCk (Solid)	2.49E+01	TE	1.7	223	6.57E-02	µCi/g
241-U-105	Sludge (Liquid & Solid)	CWR1 (Solid)	6.20E-01	TS	1.7	121	3.01E-03	µCi/g
241-U-105	Total		2.43E+02	TS/TE				
241-U-106	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.79E+01	TS	1.34	154	2.46E-01	µCi/mL
241-U-106	Saltcake Solid	S1-SltCk (Solid)	6.59E+01	TE	1.62	484	8.40E-02	µCi/g
241-U-106	Supernatant	S1-SltCk (Liquid)	1.48E+00	TS	1.34	6	2.46E-01	µCi/mL
241-U-106	Total		1.05E+02	TS/TE				
241-U-107	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	3.66E+01	S	1.43	105	3.49E-01	µCi/mL
241-U-107	Saltcake Interstitial Liquid	T2-SltCk (Liquid)	1.74E+00	S	1.43	5	3.49E-01	µCi/mL
241-U-107	Saltcake Solid	S2-SltSlr (Solid)	1.92E+02	TE	1.77	902	1.20E-01	µCi/g
241-U-107	Saltcake Solid	T2-SltCk (Solid)	4.90E+00	TE	1.77	44	6.29E-02	µCi/g
241-U-107	Sludge (Liquid & Solid)	CWR1 (Solid)	3.01E-01	TS	1.8	57	2.93E-03	µCi/g
241-U-107	Total		2.35E+02	S/TS/TE				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-U-108	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	2.16E+01	TS	1.4	86	2.51E-01	µCi/mL
241-U-108	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	1.73E+01	TS	1.4	69	2.51E-01	µCi/mL
241-U-108	Saltcake Solid	S1-SltCk (Solid)	1.21E+02	TE	1.73	766	9.11E-02	µCi/g
241-U-108	Saltcake Solid	S2-SltSlr (Solid)	1.21E+02	TE	1.73	610	1.14E-01	µCi/g
241-U-108	Sludge (Liquid & Solid)	CWR2 (Solid)	3.60E-02	TE	1.46	110	2.24E-04	µCi/g
241-U-108	Total		2.80E+02	TS/TE				
241-U-109	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	3.55E+01	TS	1.47	137	2.59E-01	µCi/mL
241-U-109	Saltcake Solid	S1-SltCk (Solid)	1.02E+02	TE	1.67	600	1.02E-01	µCi/g
241-U-109	Saltcake Solid	S2-SltSlr (Solid)	1.09E+02	TE	1.67	512	1.28E-01	µCi/g
241-U-109	Sludge (Liquid & Solid)	CWR1 (Solid)	5.29E-01	TS	1.71	103	3.00E-03	µCi/g
241-U-109	Total		2.47E+02	TS/TE				
241-U-110	Sludge (Liquid & Solid)	1C (Solid)	5.48E+00	TS	1.43	120	3.19E-02	µCi/g
241-U-110	Sludge (Liquid & Solid)	CWR1 (Solid)	5.03E-01	S	1.8	149	1.87E-03	µCi/g
241-U-110	Sludge (Liquid & Solid)	R1 (Solid)	5.01E+00	S	1.77	396	7.15E-03	µCi/g
241-U-110	Total		1.10E+01	S/TS				
241-U-111	Saltcake Interstitial Liquid	S1-SltCk (Liquid)	9.73E+00	TS	1.4	41	2.37E-01	µCi/mL

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-U-111	Saltcake Interstitial Liquid	S2-SltSlr (Liquid)	1.71E+01	TS	1.4	72	2.37E-01	µCi/mL
241-U-111	Saltcake Solid	S1-SltCk (Solid)	3.73E+01	TE	1.68	228	9.74E-02	µCi/g
241-U-111	Saltcake Solid	S2-SltSlr (Solid)	7.45E+01	TE	1.63	401	1.14E-01	µCi/g
241-U-111	Sludge (Liquid & Solid)	1C (Solid)	1.70E+00	TS	1.43	49	2.43E-02	µCi/g
241-U-111	Sludge (Liquid & Solid)	R1 (Solid)	1.20E+00	TE	1.77	49	1.38E-02	µCi/g
241-U-111	Total		1.42E+02	TS/TE				
241-U-112	Sludge (Liquid & Solid)	1C (Solid)	1.63E+00	TS	1.43	47	2.43E-02	µCi/g
241-U-112	Sludge (Liquid & Solid)	CWR1 (Solid)	3.10E-01	TS	1.86	58	2.87E-03	µCi/g
241-U-112	Sludge (Liquid & Solid)	R1 (Solid)	1.65E+00	TE	1.86	67	1.32E-02	µCi/g
241-U-112	Total		3.59E+00	TS/TE				
241-U-201	Sludge (Liquid & Solid)	CWR1 (Solid)	4.86E-02	S	1.63	11	2.71E-03	µCi/g
241-U-201	Supernatant	CWR1 (Liquid)	7.32E-02	S	1.26	4	1.83E-02	µCi/mL
241-U-201	Total		1.22E-01	S				
241-U-202	Sludge (Liquid & Solid)	CWR1 (Solid)	4.01E-02	S	1.51	10	2.66E-03	µCi/g
241-U-202	Supernatant	CWR1 (Liquid)	6.95E-02	S	1.28	4	1.74E-02	µCi/mL
241-U-202	Total		1.10E-01	S				
241-U-203	Sludge (Liquid & Solid)	CWR1 (Solid)	3.54E-02	S	1.59	9	2.48E-03	µCi/g
241-U-203	Supernatant	CWR1 (Liquid)	6.40E-02	S	1.28	4	1.60E-02	µCi/mL
241-U-203	Total		9.94E-02	S				

Tank Name	Waste Phase	Waste Type	^{99}Tc (Ci)	Basis	Density (g/mL)	Volume (kL)	Adjusted Conc.	Conc. Units
241-U-204	Sludge (Liquid & Solid)	CWR1 (Solid)	1.64E-02	S	1.47	7	1.59E-03	μCi/g
241-U-204	Supernatant	CWR1 (Liquid)	1.92E-02	S	1.11	4	4.80E-03	μCi/mL
241-U-204	Total		3.56E-02	S				

Table A.2. Hanford Tank Ranking by Total ^{99}Tc Inventory

Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)
1	**AZ-101**	1.23E+03	46	S-111	1.92E+02	91	BY-105	4.63E+01	136	U-101	1.93E+00
2	**AN-105**	1.12E+03	47	TX-110	1.88E+02	92	TY-105	4.40E+01	137	B-101	1.83E+00
3	**SY-103**	1.01E+03	48	**AY-102**	1.82E+02	93	A-102	3.70E+01	138	SX-113	1.55E+00
4	**AP-103**	9.59E+02	49	SX-102	1.82E+02	94	T-105	3.50E+01	139	SX-115	1.25E+00
5	**AP-105**	9.51E+02	50	BY-103	1.78E+02	95	T-101	3.44E+01	140	BX-109	1.09E+00
6	**AW-101**	8.18E+02	51	**AP-107**	1.77E+02	96	S-107	3.06E+01	141	TY-106	1.08E+00
7	**AN-104**	7.94E+02	52	BY-104	1.72E+02	97	SX-114	3.05E+01	142	T-104	9.74E-01
8	**AW-104**	7.59E+02	53	BY-106	1.72E+02	98	A-105	2.82E+01	143	C-102	8.87E-01
9	**AP-101**	7.55E+02	54	S-103	1.69E+02	99	TY-103	2.71E+01	144	T-108	7.72E-01
10	**AP-108**	7.24E+02	55	BY-101	1.68E+02	100	SX-111	2.46E+01	145	U-104	7.28E-01
11	**AP-102**	7.03E+02	56	SX-101	1.64E+02	101	B-110	2.13E+01	146	B-202	6.85E-01
12	**AN-103**	6.81E+02	57	TX-106	1.64E+02	102	AX-104	2.06E+01	147	B-109	4.71E-01
13	**AW-106**	5.93E+02	58	U-102	1.62E+02	103	S-102	2.00E+01	148	B-108	4.17E-01
14	**AN-102**	5.80E+02	59	BY-111	1.59E+02	104	TX-104	1.97E+01	149	B-103	3.34E-01
15	**AN-107**	4.34E+02	60	TX-116	1.56E+02	105	B-104	1.89E+01	150	T-109	3.02E-01
16	S-109	4.18E+02	61	**AP-104**	1.55E+02	106	TX-107	1.67E+01	151	B-102	2.15E-01
17	**AZ-102**	4.01E+02	62	A-106	1.48E+02	107	T-111	1.66E+01	152	C-104	1.73E-01
18	**AW-103**	3.97E+02	63	**AN-106**	1.44E+02	108	BX-112	1.57E+01	153	C-106	1.64E-01
19	**AP-106**	3.68E+02	64	U-111	1.42E+02	109	SX-110	1.52E+01	154	S-112	1.37E-01
20	S-105	3.52E+02	65	TX-111	1.41E+02	110	B-107	1.48E+01	155	U-201	1.22E-01
21	S-108	3.41E+02	66	B-111	1.32E+02	111	A-104	1.41E+01	156	BX-102	1.19E-01
22	**AW-102**	3.21E+02	67	BY-110	1.31E+02	112	TX-101	1.39E+01	157	U-202	1.10E-01
23	**SY-101**	2.98E+02	68	BY-112	1.25E+02	113	SX-108	1.38E+01	158	U-203	9.94E-02
24	TX-113	2.96E+02	69	BY-102	1.18E+02	114	SX-107	1.37E+01	159	BX-103	8.19E-02

Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)	Number Ranking	Tank	^{99}Tc (Ci)
25	AX-101	2.90E+02	70	TX-118	1.08E+02	115	C-112	1.29E+01	160	C-110	5.77E-02
26	S-106	2.85E+02	71	TX-102	1.07E+02	116	SX-112	1.28E+01	161	C-103	4.48E-02
27	SX-103	2.82E+02	72	S-101	1.06E+02	117	BX-106	1.20E+01	162	C-101	4.34E-02
28	U-108	2.80E+02	73	BY-107	1.05E+02	118	U-110	1.10E+01	163	C-108	3.96E-02
29	TX-112	2.64E+02	74	U-106	1.05E+02	119	BX-101	1.03E+01	164	U-204	3.56E-02
30	A-103	2.59E+02	75	BY-109	9.89E+01	120	TY-102	9.37E+00	165	C-109	7.50E-03
31	SX-106	2.59E+02	76	C-105	8.14E+01	121	**AW-105**	9.35E+00	166	C-204	3.18E-03
32	U-109	2.47E+02	77	BY-108	8.00E+01	122	B-112	9.15E+00	167	C-201	2.63E-03
33	**SY-102**	2.44E+02	78	SX-109	7.68E+01	123	TY-104	8.86E+00	168	C-202	2.50E-03
34	TX-105	2.44E+02	79	BX-110	7.59E+01	124	T-102	7.13E+00	169	C-203	2.32E-03
35	U-105	2.43E+02	80	BX-111	7.11E+01	125	AX-102	6.30E+00	170	T-110	1.22E-03
36	TX-115	2.39E+02	81	TX-103	7.07E+01	126	B-106	5.62E+00	171	B-203	4.16E-06
37	S-110	2.37E+02	82	AX-103	7.03E+01	127	TY-101	5.26E+00	172	B-204	3.79E-06
38	U-107	2.35E+02	83	BX-107	6.98E+01	128	C-107	4.00E+00	173	T-204	3.00E-06
39	U-103	2.30E+02	84	BX-105	6.04E+01	129	U-112	3.59E+00	174	T-203	2.96E-06
40	A-101	2.29E+02	85	**AY-101**	5.55E+01	130	T-106	2.85E+00	175	T-201	1.75E-06
41	SX-104	2.09E+02	86	TX-108	5.54E+01	131	B-105	2.75E+00	176	B-201	1.72E-06
42	**AN-101**	2.01E+02	87	S-104	5.12E+01	132	BX-108	2.58E+00	177	T-202	1.65E-06
43	SX-105	1.97E+02	88	T-107	4.83E+01	133	T-103	2.28E+00	Overall ^{99}Tc inventory		2.65E+04
44	TX-114	1.97E+02	89	BX-104	4.65E+01	134	C-111	2.19E+00			
45	TX-117	1.96E+02	90	TX-109	4.65E+01	135	T-112	2.12E+00			

Pink text indicates double-shell tanks (DSTs).

Appendix B

Listing of Reports on Characterization of Residual Sludge in SSTs That Have Been Retrieved

Appendix B

Listing of Reports on Characterization of Residual Sludge in SSTs That Have Been Retrieved

This appendix lists all the reports generated by Pacific Northwest National Laboratory staff on the characterization of residual sludges from single-shell tanks (SSTs) that have had most of their contents removed by various sluicing techniques. A summary of findings on ^{99}Tc is provided.

Some key results found in the reports germane to the fate of ^{99}Tc follow. The percentage of Tc that dissolved from residual sludge from tanks 241-C-103, 241-C-106, 241-C-202, and 241-C-203 ranged from approximately 6% to 10%. Solid-phase characterization results for leached sludges indicate that the recalcitrant forms of Tc are associated with iron oxides. X-ray absorption near edge structure (XANES) analysis of Tc in residual sludge indicates that Tc occurs in Fe oxide particles in their lower, less-soluble oxidation state [Tc(IV)]. Identifying the exact form of the Tc has been challenging. This is because Tc occurs at trace concentrations and the residual sludges are chemically complex assemblages of crystalline and amorphous solids that likely contain contaminants less as discrete phases and more as co-precipitates within oxide phases of the major components (Al, Si, Fe, etc.). Results from some of PNNL's early work using selective extractions suggested that significant fractions of Tc were typically co-precipitated at trace concentrations in Fe oxide phases that could not be identified unambiguously. Leachants used in these studies include DI water, a $Ca(OH)_2$ saturated solution, and a calcite ($CaCO_3$) saturated solution. The aqueous leaching experiments were conducted using both single-contact experiments and sequential-contact experiments. The single-contact experiments typically included 1-day and 1-month contact periods. Additional sequential-contact experiments typically included six stages in which the leachate solution was replaced with fresh leachant after each contact period. The contact periods were typically 1 day for the first five stages and 30 days for the sixth stage. All the leach tests were conducted at a solution-to-solid ratio of approximately 100:1.

In the tank residual waste samples analyzed to date, no Tc was ever detected in any of the numerous Al oxyhydroxide particles analyzed by scanning electron microscopy-energy dispersive X-ray spectrometry. Recently published quantum-mechanical modeling of ^{99}Tc incorporation into hematite indicates that incorporation of small amounts of Tc(IV) (up to at least 2.6 wt%) is energetically feasible whereas incorporation of pertechnetate (TcO_4^-) was unfavorable.[1]

Two archived sludges from tanks that have not been retrieved were also characterized (AY-102 and BX-101). A key finding of these tests is that ^{99}Tc is not completely water-leachable and mobile from tank AY-102. Only 25% of the ^{99}Tc in AY-102 sludge is water-leachable, whereas for tank BX-101 all of the ^{99}Tc is water-leachable. Finally, the drainable liquid from tank AY-102 sludge was tested using the XANES technique to determine redox state of the dissolved Tc. Tc in the drainable liquid from AY-102 sludge was shown by XANES analysis to be either in the oxidized pertechnetate [Tc(VII)] or a Tc(V) valence state soluble species, but not as reduced Tc(IV) species. Presumably the XANES analyst was aware of work on double-shell tank (DST) supernates that suggests that a soluble non-pertechnetate

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[1] Skomurski FN, KM Rosso, KM Krupka, and BP McGrail. 2010. "Technetium incorporation into hematite (α-Fe_2O_3)." *Environ. Sci. Technol.* 44(15): 5855-5861.

(n-Tc) form of ^{99}Tc is present with an assumed Tc(I) valence state. The analyst was Dr. Wayne Lukens, who also has performed much of the XANES studies on model n-Tc species to aid in efforts to determine the ^{99}Tc species in DST supernates. No supernates from AY-102 have been characterized for n-Tc , and based on the available information on what waste type constitutes the AY-102 supernate, this study could not provide any insight on whether AY-102 supernate might contain n-Tc species.

Here is the list of publications. The technical reports have embedded links to pdf files.

- Cantrell KJ, W Um, BD Williams, ME Bowden, BN Gartman, WW Lukens, EC Buck, and EJ Mausolf. 2014. "Chemical Stabilization of Hanford Tank Residual Waste." *Journal of Nuclear Materials* 446(1-3):246-256. doi:10.1016/j.jnucmat.2013.10.060
  Abstract | Details **JOURNAL ARTICLE**
- Cantrell KJ, KC Carroll, EC Buck, D Neiner, and KN Geiszler. 2013. "Single-Pass Flow-Through Test Elucidation of Weathering Behavior and Evaluation of Contaminant Release Models for Hanford Tank Residual Radioactive Waste." *Applied Geochemistry* 28:119-127. doi:10.1016/j.apgeochem.2012.09.004
  Abstract | Details **JOURNAL ARTICLE**
- Cantrell KJ, SM Heald, BW Arey, and MJ Lindberg. 2011. "Inhibited Release of Mobile Contaminants from Hanford Tank Residual Waste." In *Waste Management 2011: Global Achievements and Challenges in Waste Management, February 27-March 3, 2011, Phoenix, Arizona*, p. Paper No. 11447. WM Symposia, Tucson, AZ.
  Abstract | Details **CONFERENCE PAPER**
- Cantrell KJ, WJ Deutsch, and MJ Lindberg. 2011. "Thermodynamic Model for Uranium Release from Hanford Site Tank Residual Waste." *Environmental Science & Technology* 45(4):1473-1480. doi:10.1021/es1038968
  Abstract | Details **JOURNAL ARTICLE**
- Deutsch WJ, KJ Cantrell, KM Krupka, MJ Lindberg, and RJ Serne. 2011. "Hanford tank residual waste – contaminant source terms and release models." *Applied Geochemistry* 26(9-10):1681-1693. doi:10.1016/j.apgeochem.2011.04.025
- Cantrell KJ, KM Krupka, KN Geiszler, BW Arey, and HT Schaef. 2010. H*anford Site Tank 241-C-108 Residual Waste Contaminant Release Models and Supporting Data*. PNNL-19425, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Krupka KM, KJ Cantrell, HT Schaef, BW Arey, SM Heald, WJ Deutsch, and MJ Lindberg. 2010. "Characterizing Solids in Residual Wastes from Single-Shell Tanks at the Hanford Site." *Radwaste Solutions* 17(2):64-75.
- Cantrell KJ, KM Krupka, WJ Deutsch, and MJ Lindberg. 2009. "Contaminant Release from Residual Waste in Single Shell Tanks at the Hanford Site, Washington, USA - 9276." In *WM 2009 Waste Management for the Nuclear Renaissance, March 1-5, 2009, Phoenix, Arizona*. WM Symposia Inc., Tempe, AZ.

- Krupka KM, KJ Cantrell, HT Schaef, BW Arey, SM Heald, WJ Deutsch, and MJ Lindberg. 2009. "Characterization of Solids in Residual Wastes from Single-Shell Tanks at the Hanford Site, Washington, USA - 9277." In *WM2009 Waste Management for the Nuclear Renaissance, March 1-5, 2009, Phoenix, Arizona*. WM Symposia Inc., Tempe, AZ.
- Cantrell KJ, KM Krupka, KN Geiszler, MJ Lindberg, BW Arey, and HT Schaef. 2008. *Hanford Tank 241-S-112 Residual Waste Composition and Leach Test Data*. PNNL-17593, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Cantrell KJ, KM Krupka, WJ Deutsch, MJ Lindberg, HT Schaef, KN Geiszler, and BW Arey. 2008. *Hanford Tank 241-C-103 Residual Waste Contaminant Release Models and Supporting Data*. PNNL-16738, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Cantrell KJ, KM Krupka, WJ Deutsch, MJ Lindberg, HT Schaef, KN Geiszler, and BW Arey. 2008. *Hanford Tank 241-C-103 Residual Waste Contaminant Release Models and Supporting Data*. PNNL-16738, Pacific Northwest National Laboratory, Richland, WA.
- Deutsch WJ, KJ Cantrell, RJ Serne, and EM Pierce. 2008. "Appendix D: Contaminant Release from Residual Waste in Closed Single Shell Tanks and Other Waste Forms Associated with the Tanks." In *The Resource Conservation and Recovery Act Facility Investigation (RFI) Report for Hanford Single-Shell Tank Waste Management Areas, DOE/ORP-2008-01 Revision 0*, vol. Tier 2, ed. FM Mann. Office of River Protection, U.S. Department of Energy, Richland, WA.
  Abstract | Details **BOOK CHAPTER**
- Deutsch WJ. 2008. "Contaminant Release from Residual Waste in Closed Single-Shell Tanks and Other Waste Forms Associated with the Tanks." Chapter 19 in *The Resource Conservation and Recovery Act Facility Investigation (RFI) Report for Hanford Single-Shell Tank Waste Management Areas, DOE/ORP-2008-01 Revision 0*, vol. Tier 1, ed. FM Mann, pp. 19-1 - 19-14. Office of River Protection, U.S. Department of Energy, Richland, WA.
  Abstract | Details **BOOK CHAPTER**
- Deutsch WJ, KJ Cantrell, and KM Krupka. 2007. *Contaminant Release Data Package for Residual Waste in Single-Shell Hanford Tanks*. PNNL-16748, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Deutsch WJ, KM Krupka, MJ Lindberg, KJ Cantrell, CF Brown, and HT Schaef. 2007. *Hanford Tank 241-C-106: Residual Waste Contaminant Release Model and Supporting Data*. PNNL-15187 Rev. 1, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Deutsch WJ, KM Krupka, MJ Lindberg, KJ Cantrell, CF Brown, and HT Schaef. 2007. *Hanford Tanks 241-C-203 and 241 C 204: Residual Waste Contaminant Release Model and Supporting Data*. PNNL-14903 Rev. 1, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**

- Deutsch WJ, KM Krupka, MJ Lindberg, KJ Cantrell, CF Brown, SV Mattigod, HT Schaef, and BW Arey. 2007. *Hanford Tanks 241-C-202 and 241-C-203 Residual Waste Contaminant Release Models and Supporting Data*. PNNL-16229, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (PDF) | Details **FORMAL REPORT**
- Krupka KM, WJ Deutsch, HT Schaef, BW Arey, SM Heald, MJ Lindberg, and KJ Cantrell. 2007. "Characterization of Solids in Residual Wastes from Underground Storage Tanks at the Hanford Site, Washington, U.S.A." In *30th International Symposium on Scientific Basis for Nuclear Waste Management, Boston, November 27 – December 1, 2006. Materials Research Society Symposium Proceedings*, vol. 985, ed. DE Dunn, C Poinssot, and B Begg, pp. 473-482. Materials Research Society, Warrendale, PA.
  Abstract | Details **CONFERENCE PAPER**
- Cantrell KJ, KM Krupka, WJ Deutsch, and MJ Lindberg. 2006. "Residual Waste from Hanford Tanks 241-C-203 and 241-C-204. 2. Contaminant Release Model." *Environmental Science and Technology* 40(12):3755-3761 + supporting info. doi:10.1021/es0511568
  Abstract | Details **JOURNAL ARTICLE**
- Deutsch WJ, KM Krupka, MJ Lindberg, KJ Cantrell, CF Brown, and HT Schaef. 2006. *Hanford Tank 241-C-106: Impact of Cement Reactions on Release of Contaminants from Residual Waste*. PNNL-15544, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Krupka KM, HT Schaef, BW Arey, SM Heald, WJ Deutsch, MJ Lindberg, and KJ Cantrell. 2006. "Residual Waste from Hanford Tanks 241-C-203 and 241-C-204. I. Solids Characterization." *Environmental Science and Technology* 40(12):3749-3754, and sup. info. file.
  Abstract | Details **JOURNAL ARTICLE**
- Lindberg MJ and WJ Deutsch. 2006. *Comparison of Sludge Digestion Methods for High Organic Hanford Tank 241-C-204*. PNNL-16140, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Deutsch WJ, KM Krupka, KJ Cantrell, CF Brown, MJ Lindberg, HT Schaef, SM Heald, BW Arey, and RK Kukkadapu. 2005. *Advances in Geochemical Testing of Key Contaminants in Residual Hanford Tank Waste*. PNNL-15372, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Krupka KM, WJ Deutsch, MJ Lindberg, KJ Cantrell, NJ Hess, HT Schaef, and BW Arey. 2004. *Hanford Tanks 241-AY-102 and 241-BX-101: Sludge Composition and Contaminant Release Data*. PNNL-14614, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**
- Lindberg MJ and WJ Deutsch. 2003. *Tank 241-AY-102 Data Report*. PNNL-14344, Pacific Northwest National Laboratory, Richland, WA.
  Abstract | Full Publication (pdf) | Details **FORMAL REPORT**

Appendix C

Description of the HTWOS Computer Code

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C.1 Code Description

This appendix briefly describes the Hanford Tank Waste Operations Simulator (HTWOS) computer code and references. The HTWOS is a model/computer code that begins with an inventory in each Hanford storage tank and simulates its future form and location by applying a series of assumptions regarding the waste's retrieval out of the tanks, its pretreatment, and its final immobilization.

This model of future tank waste disposition relies on the best basis inventory (BBI) of the current estimates of each storage tank's waste inventory and volume and the processes that will be used to retrieve the waste from the tanks, the processes that will be used to separate it into high-level and low-activity fractions, and finally the processes that will be used to create the final waste forms. The HTWOS model provides estimates of the primary waste form's inventory and volume of waste generated as well as the same for any secondary waste streams and secondary waste forms that are generated. Finally, HTWOS provides an estimate of the tank residual for each tank and an estimate of tank retrieval losses (volumes and composition). As additional tanks are retrieved, the predicted residual inventories left in tanks are replaced by measured values. The predicted inventory is determined by multiplying the volume of waste assumed to be left in the tanks after retrieval by the assumed residual waste concentration.

Essentially, HTWOS is a large mass balance bookkeeping-like computer code. The HTWOS model relies on the design of chemical separations, solidification, and vitrification processes as documented in the detailed flowsheets and designs for the Hanford Tank Waste Treatment and Immobilization Plant (WTP) and the other tank waste treatment options. These various treatment processes are modeled in sufficient detail to provide estimates of process operation, of interactions or recycle between unit operations, and of quantities of primary waste products and composition of secondary waste streams. The model adheres to physical constraints inherent to the equipment and safety limitations as well as the programmatic constraints from current plans or business strategies.

Of course, the estimates are highly dependent on knowledge of the numerous processes involved with waste retrieval from the tanks, waste partitioning during pretreatment, and final waste form generation. To date, numerous scenarios for the fate of current tank waste have been produced that model waste movement from Hanford's single- and double-shell tanks to its final disposition including high-level waste (HLW) glass, low-activity waste (LAW) glass, bulk vitrification glass, supplemental low-temperature LAW waste forms such as Cast Stone, contact-handled and remote-handled transuranic waste, tank retrieval losses, tank residuals, and all secondary waste streams (gaseous, liquid, and solid). McCabe et al. (2013) as well as other Hanford technical staff caution that the HTWOS model does not account for chemical speciation or solubility constraints, so that the fate of each contaminant and other constituents may not be as accurately predicted as it would be with codes that include detailed thermodynamic constraints of solubility. Further, HTWOS does not currently account for carryover of the insoluble solids (which appear to be largely glass-formers) into melter off-gas systems.

Given these cautions, HTWOS has been actively used to estimate the volumes and contaminant inventories in any waste form contemplated to date—as long as process details are available or assumptions are carefully documented. This allows estimates of waste volumes and inventories to be generated for wastes destined for disposal on-site and for wastes that will be sent off-site. HTWOS has been used to evaluate the impacts of treated off-gas fluids recycle to the LAW melters and the benefits of Tc removal from the treated off-gas fluids prior to recycling back to LAW melters. HTWOS runs have been produced for LAW glass feed and treated off-gas fluids' compositions based on weekly batches over the 25-year River Protection Project mission for the primary LAW Facility. For each week's batch, the HTWOS model has provided batch volume and kg-moles of radionuclide and chemical constituents with radionuclides decayed from the HTWOS starting reference date to the batch date as described in Robbins and May (2013).

HTWOS has also been used to predict the composition of LAW and secondary waste streams for weekly waste processing through WTP so that both weekly and extreme high and low LAW and secondary waste simulant compositions could be prepared for use in various low-temperature waste form formulation and performance testing activities (see Westsik et al. 2013 and Russell et al. 2013 for examples).

Three documents that describe the HTWOS architecture and details are Belsher et al. (2012), Kirkbride et al. (2005), and Naiknimbalkar (2005). Figure C.1 displays the processes/facilities and their relationships that are part of the HTWOS mass balance and "fate" model. The diagram was taken from DOE/ORP 2010, Appendix B, but redrawn here to improve legibility.

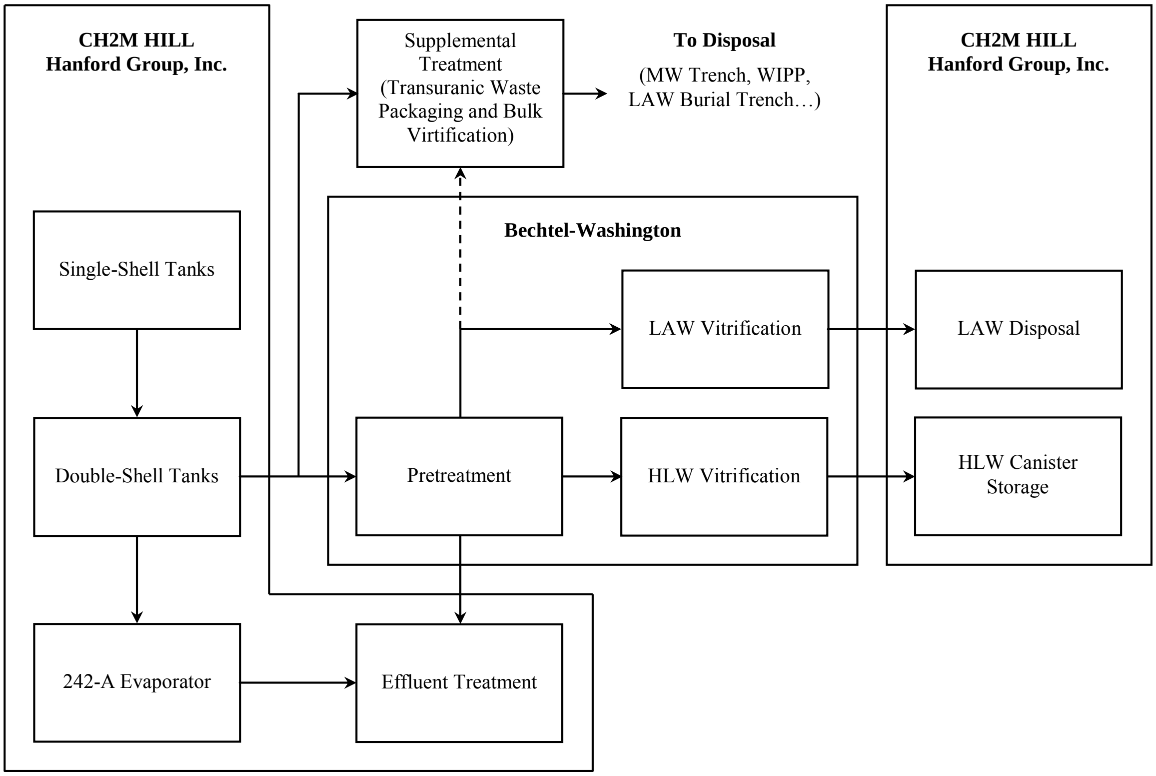

Figure C.1. Schematic of the Systems/Facilities and Waste Flow Included in HTWOS (redrawn from DOE/ORP 2010)

C.2 References

Belsher JD, PA Empey, TM Hohl, RA Kirkbride, JS Ritari, EB West, LM Bergmann, and MN Wells. 2012. *Hanford Tank Waste Operations Simulator (HTWOS) Version 7.4 Model Design Document*. RPP-17152, Rev. 7, Washington River Protection Solutions, Richland, WA.

DOE/ORP. 2010. *RCRA Facility Investigation Report for Hanford Single-Shell Tank Waste Management—Tier 1 & 2*. DOE/ORP-2008-01, Rev. 1 Reissue, Office of River Protection, U.S. Department of Energy, Richland, WA.

Kirkbride RA, GK Allen, PJ Certa, JA Lechelt, and SL Orcutt. 2005. *Hanford Tank Waste Operations Simulator Model Data Package for the Development Run for the Refined Data Case*. RPP-RPT-23412, Rev. 1, CH2M HILL Hanford Group, Inc., Richland, WA.

McCabe DJ, WR Wilmarth, and CA Nash. 2013. *Waste Treatment Technology Process Development Plan for Hanford Waste Treatment Plant Low Activity Waste Recycle*. SRNL-STI-2013-00351, Rev. 0, Savannah River National Laboratory, Aiken, SC.

Naiknimbalkar AN. 2005. *Configuration Management Plan for Hanford Tank Waste Operations Simulator Flowsheet Modeling*. RPP-7630, Rev. 4, CH2M Hill Hanford Group, Inc., Richland, WA.

Robbins RA and TH May. 2013. *Submerged Bed Scrubber Condensate Technetium Removal and Disposal Preconceptual Engineering Study*. RPP-RPT-55213, Rev. 0, Washington River Protection Solutions, LLC, Richland, WA.

Russell RL, JH Westsik, Jr, DJ Swanberg, RE Eibling, A Cozzi, MJ Lindberg, GB Josephson, and DE Rinehart. 2013. *Letter Report: LAW Simulant Development for Cast Stone Screening Test*. PNNL-22352, Pacific Northwest National Laboratory, Richland, WA. Full Publication (pdf)

Westsik Jr., JH, GF Piepel, MJ Lindberg, PG Heasler, TM Mercier, RL Russell, AD Cozzi, WE Daniel, RE Eibling, EK Hansen, MR Reigal, and DJ Swanberg. 2013. *Supplemental Immobilization of Hanford Low-Activity Waste: Cast Stone Screening Tests*. PNNL-22747 and SRNL-STI-2013-00465 Rev. 0, Pacific Northwest National Laboratory, Richland, WA and Savannah River National Laboratory, Aiken, SC.

Appendix D

Discussion and List of Resources for ^{99}Tc in Hanford Mass Balance "Pools"

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Discussion and List of Resources for ^{99}Tc in Hanford Mass Balance "Pools"

This appendix briefly describes some of the efforts and processes used to estimate the ^{99}Tc inventories that have been released to the Hanford vadose zone. Figure D.1 is a schematic showing the entire Hanford irradiated fuel separations process and where each waste stream was sent. Efforts have been expended to estimate both the release of ^{99}Tc from single-shell tanks (SSTs) and ^{99}Tc purposefully released to the now-inactive cribs, trenches, and ponds. Much effort has been placed on developing conceptual models for key contaminants for three regions in the Central Plateau and for the 300 Area. However, the 300 Area does not contain a significant inventory of ^{99}Tc, rather the key contaminant is uranium. Thus, the conceptual model and mass balance studies performed in the 300 Area are not discussed here.

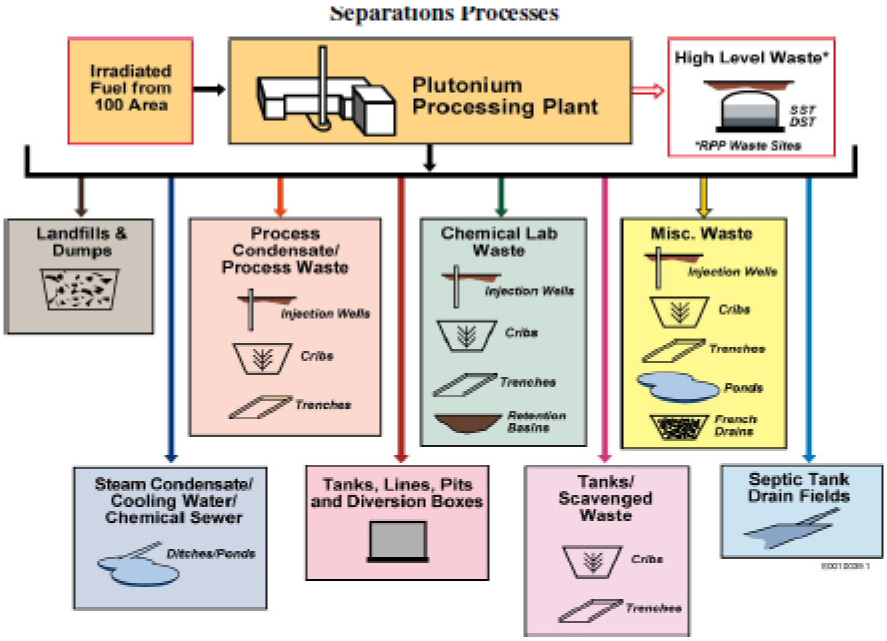

Figure D.1. Schematic of the Hanford Separations Process and Fate of the Wastes Produced

D.1 Conceptual Models for Central Plateau Sub-regions

The three areas in the Central Plateau for which contaminant conceptual models and inventories have been generated are the B-Complex in 200-E Area and the T-Complex, and to a lesser extent the S-Complex in 200-W Area. The key objective of these efforts was to evaluate available data pertaining to the boxes in Table 6.1, which evaluate releases to the ground (Box #3 in the table) and more specifically to determine the inventories in the three sub-boxes, 3A, 3B, and 3CA—inventory still in the vadose zone, inventory presently in the aquifer, and inventory that may have escaped to the Columbia River.

Conceptual models identify the most likely sources (disposal facilities or SSTs) and hypothesize the migration pathways by which contaminants reached the water table at evaluated locations.

Conceptual models attempt to estimate the activity of key mobile contaminants such as ^{99}Tc that still remain in the vadose zone (from ground surface to the water table; with the deep vadose zone portion highlighted) and within the current groundwater plumes. The methodology used to create conceptual models starts with review and assembly of pertinent background information on the suspected facilities that received the wastes. Key background information includes locations of the facilities, periods of operation, volumes of waste disposed, types of waste streams disposed, and total masses of constituents released to the vadose zone sediments.

The second step is to assemble the available data on the distribution (vertical and lateral) of the key contaminants and species that control the mobility of the contaminants in the vadose zone sediments. Plots of the available data for localized subregions when available are superimposed on cross-sections of the stratigraphy, and available field surveys (gamma logs and soil resistivity) are used to help construct models of the lateral and vertical extent of the key contaminants within the vadose zone. A similar effort is then performed using the available groundwater monitoring data to develop time-series contaminant plume maps. Using a three-dimensional aquifer conceptual model with 10 m by 10 m grid cells and the aquifer thicknesses for each year from the geologic conceptual model, the mass of selected contaminants is calculated. The masses within the annually averaged groundwater plumes are then calculated and compared to evaluate whether mass has been increasing or decreasing over the last decade and where the mass appears to be entering the water table. The shapes of the time series groundwater plumes are used to reveal the net direction of flow of the groundwater. The vadose zone contaminant distribution data and groundwater data are then combined to create integrated conceptual models of where the bulk of the contaminants remain, where they enter the water table, and how the groundwater plumes have migrated over time. The estimated masses of contaminants remaining in the vadose zone and currently present in the groundwater plumes are compared to the estimates of the total masses disposed or released to the subsurface since the beginning of waste disposal activities. These mass-balance estimates provide baseline information on two key issues:

1. How well has the mass (and thus distribution in the vadose zone and aquifer) of the contaminant been identified?
2. Can the contaminant mass distribution in the deep vadose zone and aquifer be used to project future risks and guide the selection of remediation alternatives?

Figure D.2 is an example (from the draft S-Complex conceptual site model (CSM) report) schematic of the “pools” or boxes in which contaminant mass data or estimates are needed and the logic flow of input information. The B-Complex conceptual model has been the most successful effort because this region has the most data and a relatively shallow aquifer below the key vadose zone sources and a detailed report is available:

- Serne RJ, BN Bjornstad, JM Keller, PD Thorne, DC Lanigan, JN Christensen, and GS Thomas. 2010. *Conceptual Models for Migration of Key Groundwater Contaminants Through the Vadose Zone and Into the Upper Unconfined Aquifer Below the B-Complex*. PNNL-19277, Pacific Northwest National Laboratory, Richland, WA. | Full Publication (pdf)

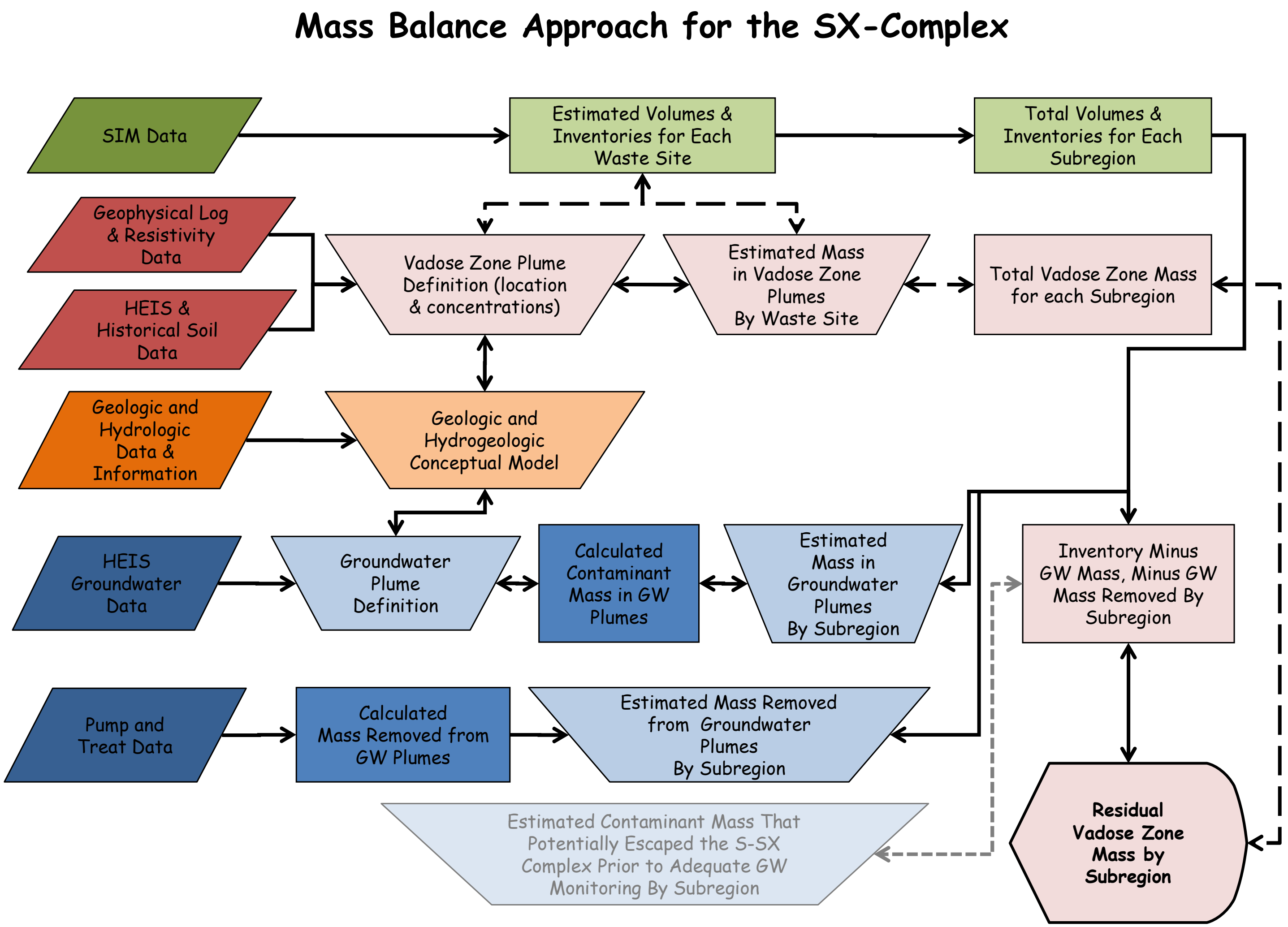

Figure D.2. Schematic of Inputs Used and Mass "Pools" Used or Calculated to Develop Conceptual Models for Contaminant Fate

A brief summary of the findings from the B-Complex report follows. The B-Complex contains three major crib and trench disposal sites and three SST farms that have released nearly 346 million liters of waste liquids containing 145 Ci of ^{99}Tc. After a thorough review of available vadose zone sediment and pore water data, groundwater plume information, field gamma logging results, and field electrical resistivity information, conceptual models were developed for which waste sites have been the significant sources of the contaminants in the groundwater. This included estimating the masses of these contaminants remaining in the vadose zone and currently present in the groundwater in comparison to the totals released. This allowed mass balance calculations to be made on how consistent our knowledge is on the deep vadose zone and groundwater distribution of contaminants. Strengths and weaknesses of the conceptual models are discussed as well as implications on future groundwater and deep vadose zone remediation alternatives.

The hypothesized conceptual models attribute the source of most of the ^{99}Tc currently in the groundwater to the BY Cribs. Mass-balance estimates suggest that there are much larger masses of ^{99}Tc remaining in the deep vadose zone within ~20 ft of the water table than is currently in the groundwater plumes below the B-Complex. This hypothesis needs to be carefully considered before future remediation efforts are chosen. The masses of these contaminants in the groundwater plumes have been increasing over the last decade, and the groundwater plumes are migrating to the northwest toward the Gable Gap. The groundwater flow rate appears to fluctuate in response to seasonal changes in hydraulic gradient. The flux of contaminants out of the deep vadose zone from the three proposed sources also appears to be transient such that the evolution of the contaminant plumes is transient.

The available vadose zone sediment characterization data show that high concentrations of ^{99}Tc reside deep in the vadose zone below the BY Cribs within both the Hanford formation H2 and the Cold Creek Unit (CCU_z) sediments. In the region of the BY Cribs, the fine-grained CCU_z subunit is absent to very thin (1 to 1.5 ft thick). It appears that water (steady-state natural recharge or transient natural or human-induced) that is fluxing through these deep sediments is carrying the ^{99}Tc down to the water table. Concentrations of ^{99}Tc in the sediments below the BY Cribs range from 120 to 200 pCi/g within 90 ft of the water table. Residual ^{99}Tc concentrations in deep vadose zone sediments at all other boreholes within or proximal to other inactive disposal facilities in the B-Complex in the same geologic units are at least one order of magnitude lower. However, at two new boreholes (299-E33-343 and 299-E33-345) far from the footprints of waste disposal facilities, water-extractable ^{99}Tc also reaches concentrations of 230 pCi/g in the CCU_z unit. Based on the available deep vadose zone sediment samples from below the BY Cribs' footprint and the assumption that the areas under all eight BY Cribs have similar ^{99}Tc concentrations, the deep vadose zone inventory below the BY Cribs is estimated to be up to 55 Ci. No other deep vadose zone region, for which sediment data are available, calculates to have ^{99}Tc activity greater than 4 Ci. Based on the high volume of waste disposed of at the BY Cribs compared to any other facility in the B-Complex, the lack of or thin manifestation of the CCU_z lateral spreading layer, and the observed high concentrations of ^{99}Tc in the deep vadose zone, the BY Cribs are considered the most important source for the ^{99}Tc currently within the groundwater plume below the B-Complex and the plume's extension out north of Gable Gap.

Time series groundwater plumes also show that the BY Cribs are the location where the high groundwater concentrations for ^{99}Tc originate. The recent time series groundwater plume maps also show a second location near the new borehole 299-E33-343 where a small ^{99}Tc "hot spot" has formed. This coincides with the occurrence of high concentrations of both ^{99}Tc and uranium in the CCU_z sediments, which are only ~16 ft above the water table at this location. The source of this secondary elevated ^{99}Tc mass has not been absolutely determined. The simplest hypothesis is that the ^{99}Tc originated from the tank BX-102 overfill event. The BX-102 fluids then spread laterally with a vertical component driven by gravity. Gravity and the stratigraphic dip of the vadose zone sediments have managed to transport the Tc into the very moist CCU_z sediments, where it and uranium from BX-102 migrate east all the way to 299-E33-345.

Once the ^{99}Tc enters the water table at the two designated locations, it appears to be migrating toward the northwest and out of the B-Complex into the Gable Gap region. Given the fact that ^{99}Tc exhibits little to no sorptive tendencies with all Hanford Site sediments, its ultimate fate will be continued migration with the regional groundwater to the northwest out of the B-Complex with little to no retardation. The leading edge of the ^{99}Tc 900 pCi/L isopleth is moving toward the northwest and near the Gable Gap advanced ~920 ft between 2000 and 2009. However, the ^{99}Tc secondary plume farther north and east of Gable Gap, likely a remnant from earlier times, is moving more slowly (900 pCi/L isopleth has advanced to the north only ~350 ft in the same period), suggesting that dispersion and dilution to concentrations well below the drinking water standard are occurring far inland from the Columbia River.

The conceptual model for both ^{99}Tc and uranium suggests that significant amounts continue to migrate through the deep vadose zone into the groundwater. The total activity of ^{99}Tc currently in the B-Complex groundwater plume is estimated at 6.06 Ci, which represents no more than 5% of the total activity of ^{99}Tc released to the subsurface below the B-Complex. The conceptual models cannot assess how much of the ^{99}Tc may have migrated out of the B-Complex region before regular groundwater monitoring was performed (i.e., before 1993, when ^{99}Tc measurements commenced). Thus, some of the difference between the mass balance estimates (~65 out of 145 Ci have been accounted for in the proposed ^{99}Tc conceptual model) may have "escaped" the region in past groundwater. The mass balance estimates show that the sum of ^{99}Tc activity deep below the BY Cribs and in the CCU_z sediments below wells E33-343 over to E33-345 is much larger (3.4 to 20.9 Ci) than the estimated activity of ^{99}Tc currently in the groundwater plume directly below B-Complex (0.76 Ci). Succinctly, there is from 8 to 30 times more ^{99}Tc in the deep vadose sediments than is currently present in the ^{99}Tc groundwater plume directly below the B-Complex. These ^{99}Tc estimates are speculative because of sparser data availability for the deep vadose zone sediments' ^{99}Tc concentrations and uncertainty in the lateral spreading area with high ^{99}Tc concentrations below BY Crib footprints. Future fate and transport modeling can be used to assess risks associated with these estimates currently in the deep vadose zone and to predict the timing for deep vadose zone ^{99}Tc to reach the water table.

Similar preliminary conceptual models for the T-Complex and S-Complex have been developed but remain unpublished pending further vadose zone contaminant characterization. Table D.1 shows the overall volumes of liquid waste released to the vadose zone and the total ^{99}Tc inventory in the wastes that were released.

Table D.1. Comparison of Total Volumes Released to Ground and ^{99}Tc Inventory in the Disposed Liquid Wastes by Central Plateau Region

Region	Total Volume of Liquid Waste Released (ML)	Total ^{99}Tc Inventory in Waste (Ci)
B-Complex	346	145
S-Complex	460	35
T-Complex	725	50

No CSMs have been generated for A-Complex, C-Complex, or U-Complex.

D.2 Releases of ^{99}Tc from SSTs and Their Infrastructure

Perhaps of more interest to staff dealing strictly with Hanford tank farm issues is the status of SST leaks and infrastructure (piping, junction boxes, etc.) release of liquids to the subsurface and the possibility of leaks during retrieval. Finally, there is the issue of residual sludge left in the bottoms of tanks after retrieval. The current reference that tabulates the status of SST integrity, the timing of past releases of fluids, and the estimated release volumes is a series of status reports generally called the Hanlon reports (e.g., Hanlon 2006), which are updated when significant new evaluations have been made. A second resource is Field and Jones 2006, which revisited the timing of past releases and the estimated release volumes. Field and Jones (2006) release volume estimates are identical to those used in the current version of the Soil Inventory Model (SIM) (Corbin et al. 2005). Attempts to revisit and improve the understanding of past SST releases to the subsurface have been ongoing. A formal protocol (see Figure D.3) is in place to revisit the past estimates for the volume, timing of the release, type of waste released (chemical composition), and total inventory of key mobile contaminants. Essentially, a team of tank farm contractor experts, staff from the Office of River Protection, and the two regulators (Washington State Department of Ecology and Environmental Protection Agency-Richland Field Office) convene and evaluate all available data and then produce a consensus document.

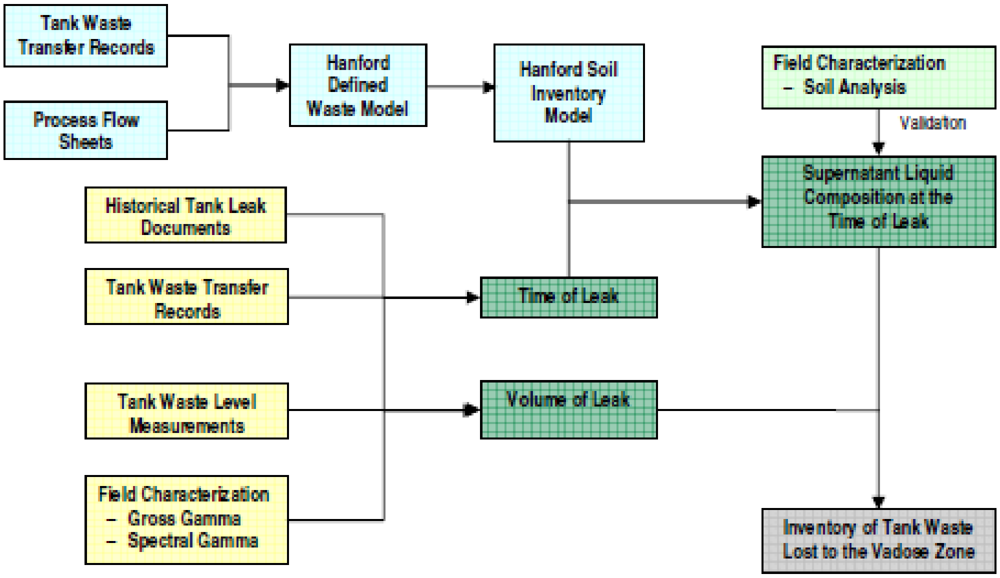

Figure D.3. Logic Diagram for the SST Leak Reassessment Efforts (from DOE/ORP 2010)

The protocol is documented in:

- Field JG, JP Harris, and ME Johnson. 2007. *Process to Assess Tank Farm Leaks in Support of Retrieval and Closure Planning.* RPP-32681, Rev. 0, CH2M Hill Hanford Group, Inc., Richland, WA.

To date the following SST farms have been reevaluated using the protocol shown in Figure D.3 and reports produced:

- Aruwah MR, JG Field, and LA Fort. 2011. *Hanford B-Farm Inventory Assessments Report*. RPP-RPT-49089, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG. 2010. *Hanford SX-Farm Leak Assessments Report.* RPP-ENV-39658, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG. 2010. *Hanford TY-Farm Leak Assessments Report*. RPP-RPT-42296, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG, JM Barton, BN Hedel, LA Fort, and MI Wood. 2011. *Hanford BY-Farm Leak Assessments Report*. RPP-RPT-43704, Rev. OA, Washington River Protection Solutions LLC, Richland, WA.
- Field JG and LA Fort. 2013. *Hanford 241-T Farm Leak Assessment Report*. RPP-RPT-55084, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG and LA Fort. 2013. *Hanford 241-TX Farm Leak Assessment Report*. RPP-RPT-50870, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG, LA Fort, BN Hedel, and MI Wood. 2011. *Hanford 241-U Farm Leak Assessment Report*. RPP-RPT-50097, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG, LA Fort, and MI Wood. 2011. *Hanford BX-Farm Leak Assessments Report.* RPP-RPT-47562, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Field JG, LA Fort, A Shrum, and MI Wood. 2011. *Hanford 241-S Farm Leak Assessment Report*. RPP-RPT-48589, Rev. 0, Washington River Protection Solutions LLC, Richland, WA.
- Johnson ME. 2008. *Hanford A and AX-Farm Leak Assessments Report: 241-A-103, 241-A-104, 241-A-105, 241-AX-102, 241-AX-104, 241-A-105, 241-AX-102, 241-AX-104 and Unplanned Waste Releases*. RPP-ENC-37956, Rev.1, CH2M Hill Hanford Group, Inc. Richland, WA.
- Fort LA, JG Field, and BN Hedel. 2011. *Hanford C-Farm Leak Assessments Report*. RPP-ENV-33418, Rev. 2, Washington River Protection Solutions, LLC, Richland, WA.

In addition to these SST farm assessments, there is a monthly Hanford tank report that tabulates the status of SST integrity, the timing of past releases of fluids, the current estimated volumes remaining in each tank, and the retrieval activities. This series of monthly status reports in the past generally were referred to as the “Hanlon reports” (e.g., HNF-EP-0182, Rev. 19; Hanlon 2006) and now are authored by Rogers (e.g., HNF-EP-0182-Rev.309; Rogers 2014). A second resource is Field and Jones 2006, which revisited the timing of past releases and the estimated release volumes. Field and Jones (2006) release volume estimates are identical to those used in the current version of the SIM (Corbin et al. 2005).

Other valuable resources that discuss the impacts of past SST leaks and long-term leaching of residual sludge left in SSTs after retrieval (and regulatory acceptance) on potential groundwater risks include Chapter 9, *Tank Waste Inventory In The Vadose Zone*, Chapter 27, *Future Impacts*, and Chapter 28, *Cumulative Impacts from the Hanford Site*, all part of DOE/ORP-2010. One of the most important findings is that the biggest source of future groundwater impacts related to Hanford SSTs comes from wastes already released (past tank leaks and infrastructure releases) and not from the residual wastes left in the tanks after retrieval. Figure D.4 shows the estimated release volume (1000 gal) and ^{99}Tc inventory (Ci) in the release for the four largest known SST releases. These four releases account for about 85% to 90% of the total estimated ^{99}Tc released (100 Ci; see Table 6.1) from SSTs and their infrastructure to the vadose zone sediments.

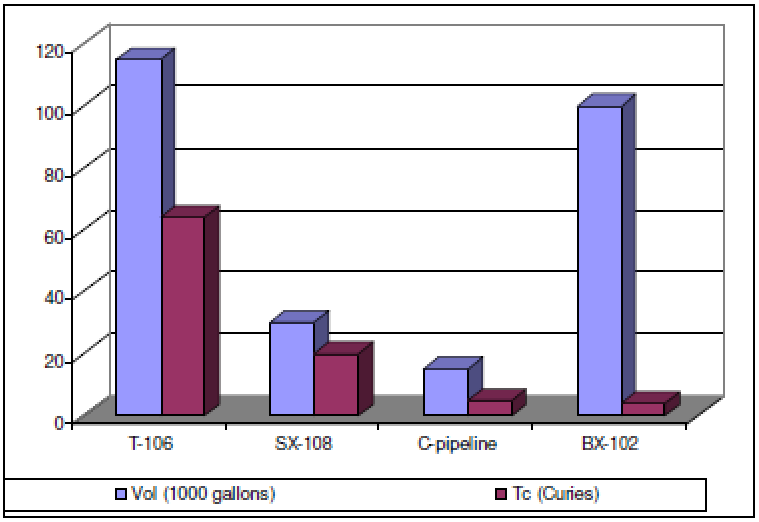

Figure D.4. Estimated Volume and ^{99}Tc Inventory in the Four Largest SST Releases (from DOE/ORP 2010)

<u>Other SST Resources Cited:</u>

- Corbin RA, BC Simpson, MJ Anderson, WF Danielson III, JG Field, TE Jones, and CT Kincaid. 2005. *Hanford Soil Inventory Model, Rev 1*. RPP-26744, CH2M Hill Hanford Group, Inc., Richland, WA.
- DOE/ORP 2005-01, *Initial Single-Shell Tank Performance Assessment for the Hanford Site*, DOE/ORP-2005-01, Office of River Protection, U.S. Department of Energy, Richland, WA.
- DOE/ORP. 2010. *RCRA Facility Investigation Report for Hanford Single-Shell Tank Waste Management—Tier 1 & 2*. DOE/ORP-2008-01 Rev. 1 Reissue, Office of River Protection, U.S. Department of Energy, Richland, WA.
- Field JG and TE Jones. 2006. *Tank Farm Vadose Zone Contamination Volume Estimates.* RPP-23405, Rev. 2, CH2M Hill Hanford Group, Inc., Richland, WA.
- Hanlon BM. 2006. *Waste Tank Summary Report for Month Ending June 30, 2006.* HNF-EP-0182-Rev. 19, CH2M Hill Hanford Group, Inc., Richland, WA.

- Rogers MJ. 2014. *Waste Tank Summary Report for Month Ending December 31, 2013*. HNF-EP-0182-Rev. 309, Washington River Protection Solutions, LLC, Richland, WA.

D.3 Useful References for Finding Input Data for Contaminant Conceptual Model Development and ^{99}Tc Inventories

One roll-up of ^{99}Tc inventories in the various "boxes" or "pools" that constitute the Hanford Site was provided in DOE/ORP 2010 and is reproduced here as Table D.2.

Table D.2. Estimate of ^{99}Tc Inventories and Waste Volumes at the End of WTP Mission (from DOE/ORP 2010, Table 2.1)

Facility	Volume	Tc-99 (Ci)
Ponds	~200,000,000,000 gallons[(a)]	~2.5[(a)]
Cribs/trenches	~300,000,000 gallons[(a)]	~600 [(a)]
Tank releases	~1,000,000 gallons[(a)]	~100 [(a)]
Tanks (as of 10/1/2007)	~50,000,000 gallons[(b)]	~26,500[(b)]
Residual waste – Future	~300,000 gallons[(b)]	~68[(b)]
Treated low-activity waste – Future	~360,000 cubic meters[(b)]	~25,000[(b)]
Geologic repository – Future	~13,000 cubic meters[(b)]	~1500[(b)]

(a) SIM (Corbin et al. 2005)

(b) HTWOS (Kirkbride et al. 2005)

Current volumes and estimates of chemical and radionuclide contaminants released to each crib/trench/pond, etc. can be found in:

- Corbin RA, BC Simpson, MJ Anderson, WF Danielson III, JG Field, TE Jones, and CT Kincaid. 2005. *Hanford Soil Inventory Model, Rev 1*. RPP-26744, CH2M Hill Hanford Group, Inc., Richland, WA. This document was also released at the same time as PNNL-15367 by Pacific Northwest National Laboratory, Richland, WA. This document has electronic appendices with all the data. However, SIM (Soil Inventory Model) is no longer being updated.

Groundwater annual reports are good resources for current estimates of the location and movement of ^{99}Tc plumes as well as discussions on probable sources, current and future remediation plans, and in some cases discussion of impacts of human health and the environment:

http://www.hanford.gov/page.cfm/SoilGroundwaterAnnualReports

Six annual reports (2007 through 2012) are currently on this web site. Earlier reports can be found on redirected URL links accessed by clicking the following links. After being redirected, scroll down and click on the link "View Document Here."

- Earlier year annual reports such as PNNL—Pacific Northwest National Laboratory. 1996. *Hanford Site Groundwater Monitoring for Fiscal Year 1996.* PNNL-11470, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www2.hanford.gov/arpir/?content=findpage&AKey=D199132964.

- PNNL. 1997. *Hanford Site Groundwater Monitoring for Fiscal Year 1997*. PNNL-11793, Pacific Northwest National Laboratory, Richland, WA.
- PNNL. 1998. *Hanford Site Groundwater Monitoring for Fiscal Year 1998*. PNNL-12086, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www2.hanford.gov/arpir/?content=findpage&AKey=D199091099.
- PNNL. 1999a. *Hanford Site Groundwater Monitoring for Fiscal Year 1998.* PNNL-12086, Pacific Northwest National Laboratory for the U.S. Department of Energy, Richland Operations Office, Richland, WA.
- PNNL. 1999b. *Hanford Site Groundwater Monitoring for Fiscal Year 1999*. PNNL-13116, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www2.hanford.gov/arpir/?content=findpage&AKey=D2736610 and http://www2.hanford.gov/arpir/?content=findpage&AKey=D2736978.

These groundwater annual reports are available directly on the PNNL publications web site:

- PNNL. 2000. *Hanford Site Groundwater Monitoring for Fiscal Year 2000*. PNNL-13404, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-13404.pdf.
- PNNL. 2001. *Hanford Site Groundwater Monitoring for Fiscal Year 2001*. PNNL-13788, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-13788.pdf.
- PNNL. 2002. *Hanford Site Groundwater Monitoring for Fiscal Year 2002*. PNNL-14187, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-14187.pdf.
- PNNL. 2003. *Hanford Site Groundwater Monitoring for Fiscal Year 2003*. PNNL-14548, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-14548.pdf.
- PNNL. 2004. Hanford Site Groundwater Monitoring for Fiscal Year 2004. PNNL-15070, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-15070.pdf.
- PNNL. 2005. *Hanford Site Groundwater Monitoring for Fiscal Year 2005*. PNNL-15670, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-15670.pdf.
- PNNL. 2006. *Hanford Site Groundwater Monitoring for Fiscal Year 2006*. PNNL-16346, Pacific Northwest National Laboratory, Richland, WA. Available at: http://www.pnl.gov/main/publications/external/technical_reports/PNNL-16346.pdf.

Commonly sought Hanford Environmental Information System (HEIS) well-monitoring data, tracking contaminants from about 1950 to the present, can be accessed at http://environet.hanford.gov/eda/.

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