

April 10, 2013

MEMORANDUM TO: Gregory Suber, Chief
Low Level Waste Branch
Environmental Protection
and Performance Assessment Directorate
Division of Waste Management
and Environmental Protection

THRU: Chris McKenney, Chief */RA/*
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FROM: Cynthia S. Barr, Sr. Systems Performance Analyst */RA/*
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SUBJECT: TECHNICAL REVIEW OF "TANK 18 AND 19 SPECIAL ANALYSIS
FOR THE PERFORMANCE ASSESSMENT FOR THE F-TANK FARM
AT THE SAVANNAH RIVER SITE", SRR-CWDA-2010-00124, REV. 0,
FEBRUARY 2012 (DOCKET NO. PROJ0734)

The U.S. Nuclear Regulatory Commission (NRC) staff has performed a technical review of the subject document prepared by the United States Department of Energy (DOE) to support Tanks 18 and 19 closure at the F-Area Tank Farm Facility (FTF) at Savannah River Site. This technical review memorandum supports Monitoring Factor 1.1, "Final Inventory and Risk Estimates" listed in NRC staff's monitoring plan for FTF (NRC, 2013). The NRC staff concludes that while the special analysis provides useful information regarding the sensitivity of predicted FTF peak doses to key modeling parameters such as Tank 18 final inventory, Pu solubility, and natural system K_d , due to its limited nature, additional information is needed to address remaining technical issues not addressed by DOE analyses to support the compliance demonstration for Tank 18 and the larger FTF facility.

Enclosure:
Technical Review of Tank 18
and 19 Special Analysis

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Technical Review of Tank 18 and 19 Special Analysis

Date: April 9, 2013

Reviewer: Cynthia S Barr

Documents: *Tank 18 and 19 Special Analysis for the Performance Assessment for the F-Tank Farm at the Savannah River Site*, SRR-CWDA-2010-00124, Rev. 0, February 2012

Summary:

The Tanks 18 and 19 special analysis is used by the U.S. Department of Energy (DOE) to evaluate new information obtained since preparation of the F Tank Farm (FTF) performance assessment (PA) (DOE, 2009). The new information evaluated includes: (i) final inventories for Tanks 18 and 19; and (ii) additional key radioelement sensitivity analyses (Pu solubility and K_d). This sensitivity analysis was performed because earlier analyses showed that considering the final inventory for Tank 18 in the reference case FTF PA model, the peak dose from Tank 18 could be significant at around 5 mSv/yr (500 mrem/yr) over longer simulation timeframes. However, DOE concludes that the additional sensitivity analysis demonstrate that the FTF base case model incorporates conservative approaches/inputs and the peak doses associated with Pu-239 would likely occur even later in time.

NRC Evaluation:

In general, the Tanks 18 and 19 special analysis provides useful information about the sensitivity of dose predictions to key modeling parameters including Pu solubility and natural system K_d . Updated geochemical modeling results (Denham, 2012) are propagated through FTF risk models to provide improved risk estimates for Tank 18. DOE also attempted to address the impact of cement leachate on near-field sorption as recommended in previous NRC comment. The results of the special analysis show that the doses could be above or below the dose standards in 10 CFR Part 61, Subpart C considering longer periods of performance. Therefore, the timing of peak dose may still be important to the compliance demonstration.

In its TER (NRC, 2011), NRC staff stated the following concern related to timing of peak dose:

However, considering the fact that unacceptably high peak doses could occur within the 10,000 year period of compliance with only a factor of 3 to 4 faster time to collective failure of a combination of barriers for Tc or Pu, respectively, and considering the large uncertainty associated with predictions of long-term performance of engineered barriers, NRC staff are not convinced that the high peak doses currently presented in DOE's PA (or lower peak doses of unknown magnitude that might be associated with a more realistic model) could not be realized within a 10,000 year compliance period. (NRC, 2011)

Enclosure

NRC staff thinks DOE has made significant improvements to its initial FTF PA solubility modeling using the latest thermodynamic data available in the literature as discussed in more detail in a separate technical review memorandum to be issued later this year. However, as discussed above, the Tank 18 and 19 special analysis shows that the doses associated with Tank 18 could be above or below the 10 CFR 61.41 performance objective considering longer periods of performance. Additionally, DOE's updated solubility modeling also shows that the solubility of Pu-239 is sensitive to redox conditions. At higher E_h (above around +0.45 V), the updated solubility modeling shows the Pu solubility could be significantly higher than assumed in DOE's PA for oxidized (Region II) conditions (i.e., around $5E-08$ mol/L compared to $3E-13$ mol/L).

Figure 1 shows that the chemical barrier afforded by the tank grout or contaminated zone (shown in purple) is the single-most important barrier affecting the timing of peak Pu-239 dose in DOE's reference case. The green, vertical line in Figure 1 shows the first chemical transition modeled in DOE's PA that delineates the transition from reduced to oxidized conditions. As discussed above, if the E_h of the system is above around +0.45 V in the oxidized (Region II) chemical state, DOE's updated solubility modeling shows that the release of Pu-239 from Tank 18 could be risk-significant much earlier in time at around 10,000 years. Thus, the updated solubility modeling shows that a factor of approximately 3 earlier time-to-failure of the most important barrier to timing of peak dose, the tank grout/contaminated zone chemical barrier, could occur making a peak dose within 10,000 years much more plausible.

Another source of uncertainty in the timing of peak dose related to chemical transition times was addressed by DOE in a Request for Additional Information (RAI) response but was not considered in the Tanks 18 and 19 special analysis. In FTF RAI response (DOE, 2011), DOE performed a "Case G" analysis to study a potential conceptual model referred to as Condition 2 in DOE's PA (SRS-REG-2007-00002, Rev. 1). In Condition 2, a fast flow path exists through the tank grout prior to significant (bulk) grout degradation. Therefore, flow through the system is dominated by fractures or cracks that might form in the system over time (e.g., shrinkage gaps, or cracks that might develop due to thermal or mechanical stresses imposed on the system during curing, or due to corrosion of steel components) and waste release is dominated by water that, after a short period of time, is not assumed to be chemically conditioned via prolonged interactions with the reducing grout. Because the Case G scenario defeats the primary barrier prolonging the peak dose in DOE's FTF PA (i.e., the purple, chemical barrier shown in Figure 1), the peak dose occurs within the 10,000 year performance period¹ at doses

¹ The "NRC Staff Guidance for Activities Related to U.S. Department of Energy Waste Determinations, Draft Final Report for Interim Use" (NUREG-1854) recommends "Generally, a period of 10,000 years after closure is sufficient to capture the peak dose from the more mobile, long-lived radionuclides and to demonstrate the influence of the natural and engineered systems in achieving the performance objectives (NRC, 2000). However, assessments beyond 10,000 years may be necessary to ensure (1) that the disposal of certain types of waste does not result in markedly high impacts to future generations or (2) evaluate waste disposal at arid sites with extremely long groundwater travel times. Periods of performance shorter than 10,000 years are generally not appropriate for disposal facilities for incidental waste, because of the larger fraction of long-lived radionuclides compared to a typical commercial low-level waste (LLW) disposal facility. Presenting and understanding long-term risk (e.g., greater than 10,000 years) can be an important part of performance assessment analyses, even if those risks are not used to demonstrate compliance with the performance objectives of 10 CFR Part 61, Subpart C."

above the 0.25 mSv/yr (25 mrem/yr) dose standard used to evaluate compliance with 10 CFR 61.41.

While DOE characterized Case G as highly unlikely or incredible, the Pu solubility peer review group that evaluated DOE's Pu waste release modeling (Cantrell et al., 2011) stated the following:

Slow carbonation of the grout as a result of diffusion of carbon dioxide into the grout with water seems like a low probability scenario. Fracturing of the grout with preferential flow through the cracks would appear to be a much more likely scenario and should therefore be evaluated.

The peer review group suggests that a key modeling assumption in DOE's FTF reference case—diffusion limited transport of dissolved carbon dioxide through tank grout—seems less likely. The peer review group expectation that fluid flow will occur through cracks in the tank grout might also be extended to the steel liner corrosion modeling, as well as the cementitious material degradation modeling. Case G considers the potential impact of flow through cracks implicitly with earlier steel liner failure and chemical transition times—modeling assumptions that lead to doses above the dose limit of 0.25 mSv/yr (25 mrem/yr) within the 10,000 year performance period. DOE, however, did not address the important peer review group comment regarding flow through cracks in the Tanks 18 and 19 special analysis that might hasten the time to steel liner and chemical barrier failure times (green and purple barriers in Figure 1, respectively). A better understanding of the likelihood and performance impact of a scenario similar to Case G is, therefore, considered by NRC staff to be important to DOE's compliance demonstration. Many of the key monitoring factors developed by NRC staff in its FTF monitoring plan (NRC, 2013) provide additional information to evaluate the likelihood and performance impact of preferential flow through the grouted tanks.

For FTF, NRC staff is also concerned with an alternative scenario in which the water table periodically rises above the bottom of the tanks (or the tank bottoms are within the zone of water table fluctuation), potentially leading to accelerated corrosion and direct contact of saturated groundwater with the contaminated zone. Because Type IV tanks like Tank 18: (i) have bottoms located at or near the elevation of the long-term average water table; (ii) have no vault annulus to grout; (iii) have experienced groundwater in-leakage into tank vaults; and (iv) contain a risk-significant inventory of key radionuclides such as Pu-239 and Np-237, the likelihood of early, unconditioned release of tank waste due to water table rise is of particular concern for Type IV tanks. Thus, in its TER for FTF (NRC, 2011), NRC staff recommended DOE perform experiments to evaluate radionuclide solubility under a range of chemical conditions relevant to the residual sludge in the grouted waste tanks. If DOE can show dissolved concentrations of key radionuclides can be limited to non-risk-significant levels under all relevant chemical conditions, the conceptual model for waste release (and the timing of the release) becomes less important. Experiments to study solubility are the focus of Monitoring Factor 2.1 in NRC's monitoring plan (NRC, 2013).

Additionally, DOE's Tanks 18 and 19 special analysis does not address a key comment made in NRC staff's FTF TER (NRC, 2011) related to Pu sorption in the environment—that travel times for more mobile and less mobile forms of Pu are averaged together leading to longer travel times for the smaller fraction of more mobile Pu. If DOE represents the more mobile form of Pu explicitly in its PA model, rather than averaging the properties of both immobile and

mobile forms together, then travel times of Pu in the natural system to a point of assessment may be relatively rapid for the more mobile fraction of Pu compared to the Pu travel times reflected in the base case and sensitivity studies. It is also important to note that because Tank 18 is close to the FTF boundary and has virtually no vadose zone, travel times from Tank 18 to the 1 m point of compliance are expected to be much shorter than to the 100 m point of compliance making compliance with the 10 CFR 61.42 performance objective potentially bounding if DOE must rely on travel time to the 100 m point of assessment to demonstrate compliance with 10 CFR 61.41.

In lieu of addressing the issue related to K_d averaging, DOE retained a single composite K_d for Pu and revised natural system Pu K_d s only upwards (K_d s of 650 to 1300 L/kg are used in sensitivity cases versus a K_d of 270 L/kg used in DOE's FTF reference case for Pu in sand). This modeling change serves to increase travel times to the point of compliance, partially offsetting the significantly earlier waste release times that result from updated solubility modeling cases.

In select sensitivity cases, DOE initially assigns a value of 1300 L/kg for the Pu sand K_d to reflect the impact grout leachate on sorption of Pu. In other sensitivity cases, DOE assigns a value of 650 L/kg for Pu sand K_d for all time. The new K_d s are based on a statistical analysis of available sorption data for the site (Almond, et al., 2012). Both of these K_d values are significantly higher than the value of 270 L/kg DOE used in the FTF PA. The NRC staff discusses technical concerns related to the updated Pu K_d analysis in a separate technical review memorandum to be issued later this year. For the reasons mentioned above, NRC staff does not think that the range of parameter values evaluated by DOE in its sensitivity analysis encompass the full range of measured values, nor does the modeling approach address the complexities of Pu transport reflected in site-specific studies.

In summary, DOE's updated solubility modeling has not ruled out risk-significant release of Pu-239 under oxidizing conditions. At reasonable, although what is characterized by DOE as "conservative" E_h , Pu-239 can be released at risk-significant quantities leading to doses that are comparable to the reference case dose for Tank 18, around 3 mSv/yr (300 mrem/yr). Experimental verification of the extent to which groundwater is chemically conditioned by the tank grout over time (e.g., the evolution of E_h over time) and the expected change in solubility of key radionuclides in response to these chemical changes is also important. If releases are expected to be risk-significant, it will also be important for DOE to rigorously evaluate the timing of the releases to demonstrate compliance within the performance period. As illustrated in Figure 1, factors that are important to the timing of peak dose in DOE's reference case in order of importance are the following:

- Chemical barriers,
- Natural system (K_d s),
- Steel liner, and
- Basemat.

DOE's updated solubility modeling shows that the chemical barrier for Pu-239 may be much less effective at around 10,000 years or upon transition to oxidized conditions delineated by the vertical, green line in Figure 1 (versus 30,000 years delineated by the vertical red line in Figure

1). DOE did not evaluate alternative conceptual models (e.g., flow through cracks in the tank grout) in its special analysis that may also lead to earlier chemical transitions times (a shift in the vertical green line to earlier times). Therefore, it will be important for DOE to provide additional support for its reference case PA model or to rule out alternative configurations that may lead to unacceptable results.

DOE did evaluate natural system K_d s in the Tanks 18 and 19 special analysis as recommended in NRC staff's FTF TER; however, DOE only revised its natural system K_d s upwards, which serves to further delay the timing of peak dose. DOE did not evaluate the significance of site-specific data on more mobile forms of Pu that if explicitly represented in the FTF PA model may lead to significant doses within the period of performance. For example, if a significant fraction of more mobile Pu-239 is present in the subsurface at FTF, using a K_d of 16 L/kg (SRS-REG-2007-00002, Rev. 1), bulk soil density of 1.65 g/cm³, and a porosity of 0.38, the retardation factor for more mobile forms of Pu-239 can be calculated at a value of 70 using the following equation:

$$R_f = 1 + \frac{\rho}{\eta} * K_d$$

$$70 = 1 + \frac{1.65}{0.38} * 16$$

If groundwater travel times to the 1 and 100 m wells are on the order of around 10 years, travel times to the points of assessment (1 and 100 m exposure points) could be on the order of hundreds, rather than tens of thousands of years currently assumed in DOE's reference case and updated Tanks 18 and 19 special analysis. Therefore, it will be important for DOE to demonstrate that (i) more mobile forms of Pu released from the tanks are not present in risk-significant quantities, or (ii) that geochemical conditions are favorable in the sense that Pu is immobilized and Pu concentrations are reduced to non-risk-significant levels along FTF release paths from the FTF tanks in the natural environment. Evaluation of key radionuclide mobility in the subsurface is listed under MF 4.1 in NRC's FTF monitoring plan (NRC, 2013).

Finally, DOE does not address technical issues raised by NRC associated with steel liner corrosion and basemat sorption in its special analysis. The NRC staff developed monitoring factors 3.1 and 3.5 in its FTF monitoring plan to address these issues (NRC, 2013). It is important to note that the Tank 18 basemat is also an important barrier mitigating Np release and is also listed in NRC staff's FTF monitoring plan (NRC, 2013) under monitoring factor 3.5.

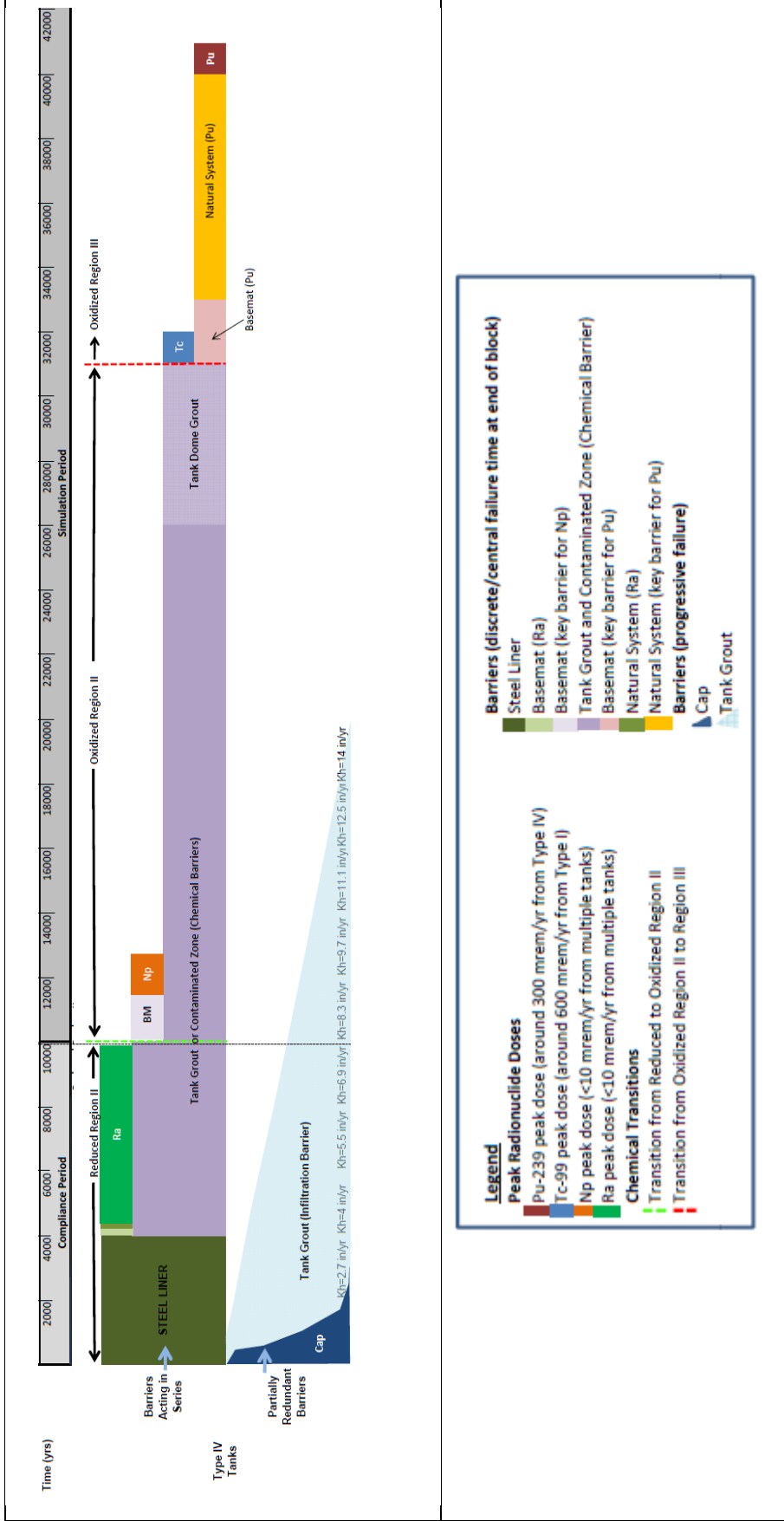


Figure 1 Barriers to Timing of Peak Dose for Type IV Tanks in DOE's Reference Case

NRC staff also has concerns with comments made by DOE in the Tanks 18 and 19 special analysis:

It should be noted that irrespective of the plutonium solubility value, the peak doses associated with Pu-239 are always below the 25 mrem/yr performance objective within the 10,000-year performance period. This sensitivity study also shows that the multiple barriers to Pu-239 release are effective and can prevent a significant release in the 10,000-year time period regardless of the Pu-239 solubility limits.

These additional sensitivity analyses demonstrate that the FTF Base Case model incorporates conservative approaches/inputs and the peak doses associated with Pu-239 would likely occur even farther beyond the 10,000 year performance period if these conservative approaches/inputs were eliminated.

NRC staff does not agree with the statement that sensitivity analyses show that barriers are effective and can prevent a significant release in 10,000 years. First, sensitivity analyses do not show whether a barrier is or is not effective; sensitivity analyses show the impact of a model or model parameter change on a model output. Actual barrier effectiveness is dependent on the field emplaced properties of the barrier(s) and the internal and external forces acting on the barrier(s) that may or may not be represented in a sensitivity study. The NRC staff note that the sensitivity analysis conducted to support the special analysis for Tanks 18 and 19 does not appear to encompass the full range of potential performance of FTF barriers as discussed above. Second, the assumed level of performance of key barriers in DOE's reference case PA is not fully supported. Key barriers to facility performance should be supported with multiple lines of evidence and should consider, as appropriate, various features, events, and processes that might significantly affect the effectiveness of the barrier. While DOE performed updated solubility modeling (Denham, 2012) that generally improves risk estimates for the site, DOE has not performed experimental validation of its modeling results. Furthermore, DOE's updated solubility modeling shows that the Pu dose can be above the Part 61, Subpart C performance objectives but does not consider important factors that might serve to hasten the time of peak (or risk-significant) dose within the performance period.

NRC staff also has concerns with the statement about the conservatism of the base case model for the following reasons: (i) the range of parameter values used in the sensitivity analysis may not reflect the full range of parameter values in site-specific analyses; and (ii) because the sensitivity analyses only evaluate a limited set of potential outcomes, the calculations are inadequate to show whether the base case is or is not conservative. Furthermore, NRC staff also finds statements regarding the timing of peak dose misleading as the updated solubility modeling actually shows that the peak doses could occur much earlier in time. As indicated above, DOE conducted updated modeling to address deficiencies identified by the Pu solubility peer review group with respect to the geochemical modeling in the FTF PA (Denham, 2012) and found that under oxidizing conditions, Pu could be released at risk-significant levels 20,000 years earlier in time than assumed in the basecase model.

The combined impact of *all* potential conservatisms and non-conservatisms in DOE's reference PA model is not clear. Based on consideration of all existing information, NRC staff concludes that the performance objectives in 10 CFR 61.41 and 61.42 may or may not be met considering a 10,000 year period of performance. Therefore, NRC staff continues to recommend waste

release experiments to increase support for key modeling assumptions related to Pu solubility. Additional information related to other key barriers may also be needed pending the results of the solubility experiments.

Teleconference or Meeting:

During the September 26 to 27th onsite observation at FTF, NRC staff sought clarification from DOE on a number of statements made in the Tank 18 and 19 special analysis. DOE clarified that expert panel statements regarding (i) the potential for degraded liner corrosion product sorption and (ii) the expected late timing of the Pu peak dose were actually made in verbal discussions between DOE and the expert panel and were not documented in the Pu solubility peer review report. NRC also discussed the impact of DOE's FTF PA assumption that the tank grout is completely degraded at 500 years in Configurations B through F. Although this might appear to be a conservative assumption, because DOE assumes that infiltrating groundwater flows through the grout monolith (versus through cracks in the grout monolith), a greater mass of tank grout is available to condition infiltrating groundwater to low Eh and high pH thereby maintaining low solubility of several key radionuclides in the waste zone for longer periods of time. If flow were primarily through cracks, then the buffering capacity of the grout along crack faces would become depleted in a shorter period of time and chemical transitions to higher solubility could occur significantly earlier in time. Therefore, NRC staff plan to monitor the extent of groundwater conditioning and chemical transition times in monitoring factors 2.2 and 3.2 of NRC staff's monitoring plan (NRC, 2013).

Follow-up Actions:

There are no follow-up actions.

Open Issues:

There are currently no open issues. The FTF monitoring plan (NRC, 2013) addresses remaining technical issues related to solubility control of Pu (monitoring factor 2.1), groundwater conditioning (monitoring factors 2.2 and 3.2), steel liner corrosion (monitoring factor 3.1), basemat sorption (monitoring factor 3.5), and natural system K_d (monitoring factor 4.1).

Conclusions:

The combined impact of *all* potential conservatisms and non-conservatisms in DOE's reference PA model is not clear; additional information is needed to support the compliance demonstration for Tank 18 and the larger FTF. Therefore, contrary to DOE's conclusion in the Tank 18 and 19 special analysis, NRC staff continues to recommend waste release experiments to better understand Pu solubility under a range of chemical conditions.

NRC staff concludes that while the Tanks 18 and 19 special analysis provides useful information regarding the sensitivity of predicted FTF peak doses to key modeling parameters such as Tank 18 final inventory, Pu solubility, and natural system K_d , due to its limited nature, additional information is needed to address remaining issues identified in NRC staff's technical evaluation report (NRC, 2011) that were not evaluated in the DOE analysis to support the

compliance demonstration for Tank 18 and the larger FTF facility. NRC staff's plan for monitoring factors important to FTF performance is provided in NRC's FTF monitoring plan (NRC, 2013).

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