SRR-CWDA-2012-00080

Revision 0

Barrier Analysis Report from the Performance Assessment for the H-Area Tank Farm at the Savannah River Site

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ACRONYNMS / ABBREVIATIONS

CZ	Contamination Zone
HELP	Hydrologic Evaluation of Landfill Performance
HTF	H-Area Tank Farm
K_d	distribution coefficient
mL/g	milliliters per gram
MOP	Member of the Public
PA	Performance Assessment

1.0 INTRODUCTION

The H-Area Tank Farm (HTF) Performance Assessment (PA) was prepared to support the removal from service of the HTF underground radioactive waste tanks and ancillary equipment. The PA provides the technical basis and results used in subsequent documents to demonstrate compliance with the pertinent requirements for removal from service and final closure of the HTF. [SRR-CWDA-2010-00128]

This report documents the barrier analysis from the HTF PA Revision 0 and draws conclusions based on the results, as is relevant to future revisions of the HTF PA. For the purpose of this report, a barrier is a major feature of the HTF closure system that functions to inhibit the release or transport of radionuclides. Specifically, this analysis examines the performance of the following five engineered and natural barriers: the closure cap, the waste tank grout and cementitious material, the contamination zone (CZ) grout, the waste tank liner, and the vadose zone under the tanks.

The barrier analysis from the HTF PA Revision 0 evaluated the barrier capabilities offered by the HTF closure system and provided a systematic analysis of each barrier relative to waste release and migration. The barrier analyses assessed the contributions of individual barriers by comparing flux results from representative contaminants under various barrier conditions.

1.1 Barrier Analysis Scope

This analysis divided the HTF closure system into five barriers. Each barrier was modeled as having degraded conditions and the model results were compared to results from models that simulated undegraded or nominal conditions. These comparisons focused on the timing and magnitude of fluxes of released radionuclides that represented both quick releasing and slow releasing contaminants. These degraded modeling cases did not consider the causative factors for barrier degradation (i.e., degradation was non-mechanistic), thus the assumed degradation of each barrier was independent of any other barrier in the analysis.

The barrier analysis was carried out for the waste tanks listed in Table 1.1-1. Time histories displaying radionuclide fluxes from Type II tanks (both initially degraded and initially intact) and from Type IV tanks, which are both predominant contributors to dose, will be the focus of this analysis. The Type II tanks are unique in that they are modeled with an initial radionuclide inventory in their primary sand pad and annulus (Tank 16 also includes an initial inventory in the secondary sand pad).

Table 1.1-2 lists the radionuclides selected for the barrier analyses, along with a description of their significance. These radionuclides were chosen based on either (1) the relative impact on dose or (2) the transport characteristics (i.e., the radionuclide has distribution coefficient (K_d) values and solubility limits that are representative of a number of radionuclides).

Table 1.1-1: Summary of Waste Tanks Selected for Barrier Analysis

Representative Waste Tank	Waste Tank Type	Initially Failed Liner?
Tank 9	Type I (submerged)	No
Tank 12	Type I (submerged)	Yes
Tank 13	Type II (submerged)	No
Tank 15	Type II (submerged)	Yes
Tank 21	Type IV	No
Tank 36	Type IIIA	No

[SRR-CWDA-2010-00128]

Table 1.1-2: Summary of Radionuclides Selected for Barrier Analysis

Radionuclide of Interest	Half-Life (years)	Significant Characteristics
Tc-99	2.11E+05	Significant dose contributor, long-lived, redox sensitive, K_d/solubility controlled
Ra-226	1.60E+03	Significant dose contributor, short-lived, not redox sensitive, K_d /solubility controlled, generated through ingrowth from the Pu-238 → U-234 → Th-230 chain
Pu-239	2.41E+04	Long-lived, redox sensitive, K_d/solubility controlled
I-129	1.57E+07	Significant dose contributor, long-lived, not redox sensitive, no solubility controls
Np-237	2.14E+06	Long-lived, K_d/solubility controlled, generated through ingrowth from Cm-245 → Pu-241 → Am-241 chain

[SRR-CWDA-2010-00128]

Each modeling run performed in support of the barrier analyses used the same initial inventory as the Base Case (Case A) for each waste tank (i.e., the initial inventory estimates are the same). The analysis point for each barrier was the time histories for each radionuclide flux below the waste tank (i.e., at the water table for unsaturated waste tanks and exiting the basemat for submerged waste tanks).

Table 1.1-3 describes the nominal (N), partially degraded (P), and fully degraded (F) condition for each barrier (vadose zone beneath the waste tanks, closure cap, waste tank liner, CZ, and the waste tank grout and other cementitious material). The initial conditions for each barrier analysis were assumed to be independent of the other barriers. For the vadose zone, the "fully degraded" assumed modified K_d values for radionuclides in the soil, as shown in Table 1.1-5.

Table 1.1-4 lists the ten Barrier Cases and the physical condition of the materials for each of the ten run configurations. The barrier analyses includes Case A, which uses the nominal chemical and hydraulic properties and a degraded run configuration (Barrier Case 2) where every barrier other than the CZ is modeled as fully degraded. There are also specific cases associated with each barrier to evaluate the capabilities of each configuration by holding other barrier conditions constant while varying the condition of the barrier being assessed. K_d values applied to Vadose Zone nominal and failed conditions are defined in Table 1.1-5. Additional information regarding the material properties for the various barriers was provided in Sections 4.2.1 and 4.2.2 of the HTF PA Revision 0. [SRR-CWDA-2010-00128]

Table 1.1-3: Barrier Analysis Variability

Barrier	N (Nominal)	P (Partially Degraded)	F (Fully Degraded)
Closure Cap	Infiltration profile per Case A (HTF PA Table 4.2-23)	N/A	Infiltration constant at 16.45 inches per year [WSRC-STI-2007-00184]
Tank Grout and Cementitious Materials [a] (K_d controlled)	Hydraulic properties (e.g., failure date) and chemical properties unchanged per Case A	N/A	Hydraulic properties of failed grout and cementitious materials at time = 0, initial chemical properties unchanged. High flow throughout grout causes grout to impart reducing capacity onto CZ. Chemical transitions are a function of the "failed" flow fields.
CZ (Solubility controlled)	CZ initial solubility limits are those associated with Case A	N/A	Solubility controls are removed for Tc-99 and Ra-226, and set to facilitate faster releases for the remaining radionuclides (for example the solubility control for plutonium was changed from 8.2E-14 mol/L to 5.7E-05 mol/L).
Waste Tank Liner [b]	Later liner failure (based on grouted CO_2 diffusion coefficient of 1E-06 cm^2/s)	Early liner failure (based on grouted CO_2 diffusion coefficient of 1E-04 cm^2/s)	No Liner at time = 0
Vadose Zone [c] (K_d controlled)	Native soil K_d values are set equal to Case A values	N/A	Native soil K_d values are as defined in Table 1.1-5.

[SRR-CWDA-2010-00128]

a Includes basemat, wall, and roof cementitious materials

b The liner barrier analysis does not apply to waste tanks with initially failed waste tank liners (e.g., Tank15 for this analysis).

c The vadose zone is the unsaturated native soil zone beneath the basemat of the waste tanks; therefore, this analysis only applies to the unsaturated waste tanks, which include Types IV and III/IIIA tanks.

N/A = Not applicable

Table 1.1-4: Barrier Case Analysis Configuration Summary

Configuration	**PORFLOW Base Case**	**Fully Degraded**	**Waste Tank Liner[a]**			**CZ**		**Vadose Zone**		**Closure Cap**
Barrier Case	**Case A**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	No Cap[a]
Closure Cap	N	F	F	F	N	F	N	N	F	F
Tank Grout and Cementitious materials [b]	N	F	F	F	N	F	N	N	F	N
CZ	N	N	N	N	N	F	F	N	N	N
Liner [c]	N	F	N	P	P	F	N	N	F	N
Vadose Zone	N	F	F	F	N	F	N	F	N	N

[SRR-CWDA-2010-00128]

a The "No Cap" configuration represents a configuration that simulates closure without a closure cap.
b Includes basemat, wall, and roof cementitious materials
c For Tank 15, liner is failed at time zero, therefore nominal liner barrier analysis and partial liner barrier analysis are not applicable.

N = Nominal
P = Partially degraded
F = Fully degraded

Table 1.1-5: K_d Values Applied to Vadose Zone Barrier Cases

Radionuclides	**Nominal Values [a]**		**Failed Values**	
	Sandy Soil (mL/g)[b]	**Cement Leachate Impacted Sandy Soil (mL/g)[c]**	**Minimum - Sandy Soil (mL/g)[b]**	**Minimum - Cement Leachate Impacted Sandy Soil (mL/g)[c]**
I-129	0.3	0	0.07	0.01[d]
Np-237	3	5	0.75	1.12
Pu-239	290	580	72.5	145
Ra-226	25	75	1.25	3.75
Tc-99	0.6	0.1	0.15	0.01

[SRR-CWDA-2010-00128]

a From HTF PA Table 4.2-29
b Values applied to unsaturated waste tanks (Type III, IIIA, and IV tanks) upon transition to non-cement leachate impacted soil
c Applied to unsaturated waste tanks (Type III, IIIA, and IV Tanks) initially
d The failed, cement leachate impacted sandy soil K_d for I-129 was simulated using the value 0.01 mL/g, instead of 0 mL/g. However, this low value had little impact on the results; therefore, the simulation was not rerun.

In addition to the modeling cases that were specifically designed in support of the barrier analyses, the barrier analyses also used results from the alternative tank configuration cases (Cases B and C) modeled for the HTF PA. Table 1.1-6 summarizes the conditions of these modeling cases. An assumed "fast flow path" is modeled by modifying the hydraulic conditions of a portion of the waste tank properties such that a channel of faster flow rates exists through various barriers within the waste tank. For completeness, this table also includes the other HTF PA alternative configuration cases. [SRR-CWDA-2010-000128]

Table 1.1-6: Waste Tank Case Summary

Case	Assumed Fast Flow Paths	Degradation of Cementitious Materials	Liner Failure Time[a]	CZ/Chemical Transition Driver
A	None	Degradation curve based on Table 4.2-34 of the HTF PA Rev 0	Later failure date (based on grouted diffusion coefficient of 1.0E-06 cm^2/s CO_2) in Table 4.2-36	Full Grout Capacity
B	Channel with no flow impedance through grout	Degradation assumed to be a step change at year 501	Early failure date (based on grouted diffusion coefficient of 1.0E-04 cm^2/s CO_2) in Table 4.2-36	Full Grout Capacity
C	Channel with no flow impedance through grout	Degradation curve based on Table 4.2-34 of the HTF PA Rev 0	Early failure date (based on grouted diffusion coefficient of 1.0E-04 cm^2/s CO_2) in Table 4.2-36	CZ Reducing Capacity
D	Channel with no flow impedance through grout and basemat	Degradation assumed to be a step change at year 501	Early failure date (based on grouted diffusion coefficient of 1.0E-04 cm^2/s CO_2) in Table 4.2-36	Full Grout Capacity
E	Channel with no flow impedance through grout and basemat	Degradation curve based on Table 4.2-34 of the HTF PA Rev 0	Early failure date (based on grouted diffusion coefficient of 1.0E-04 cm^2/s CO_2) in Table 4.2-36	CZ Reducing Capacity

[Source: SRR-CWDA-2010-00128]

Note Case E is a combination of Cases C and D. Case E uses flow path from Case D and remaining transitions from Case C.

D = diffusion coefficient

a Grouted diffusion coefficient reported in cm^2/sec and Tanks 12, 14, 15, and 16 were modeled with a failed liner at the time of closure for all cases.

Chemical transition times for the different barrier cases are discussed and presented in Section 2. Time histories displaying flux below the waste tanks for each of the barrier cases are included in Appendix I of HTF PA Revision 0. The barrier analysis results presented in Section 3 illustrate the importance of each barrier with respect to the release and transport of radionuclides. To support the evaluation of the barriers, the flux results from Appendix I are re-plotted for the radionuclides and waste tanks of interest such that only the run configurations pertaining to the specific barrier are included. These barrier-specific plots are included in Appendix N of the HTF PA Revision 0; however, data used to support the analysis of the performance of each barrier are reproduced in Section 3.

2.0 BARRIER ANALYSIS TRANSITION TIMES

The release of contaminants from residual waste in closed high level waste tanks will depend on the chemical composition of pore fluids passing through the CZ. The composition of these fluids will vary, causing solubilities of key radionuclides to vary, as infiltration water flows through the tank fill grout. Infiltration from the surface or groundwater encounters numerous barriers, in nominal or degraded conditions, that influence radionuclide transport. Fluids that pass through the waste tanks interact with the grout, driving changes to grout mineralogy and causing fluids emerging from the grout into the CZ to have compositions that reflects these interactions. Release of contaminants from the waste tanks is controlled primarily by solubility of assumed contaminant-bearing solid phases in the varying fluid composition. The contaminants that are released from the CZ travel through the waste tank basemat, which delays their travel based on the K_d value, which is also dependent on the chemistry of the pore fluid that travels through the basemat. Therefore, the various barrier cases will influence the times that the cementitious barriers (tank grout, CZ, annulus, basemat, etc.) transition from one chemical state to another. The transition times also vary by waste tank type because of the differing thickness of the cementitious material used in each.

In addition to the chemical transition times, the liner failure time and the hydraulic degradation of the waste tank components are also important to radionuclide transport and the timing of radionuclide releases from the waste tanks. A selection of the transition times for the waste tanks are included in Figures 2.0-1 through 2.0-17, and illustrate the chemical transition times, the hydraulic degradation transition times, and the liner failure times for all barrier cases. Transitions for the annulus, waste tank grout, and CZ for Type II tanks (e.g. Tank 13) (Figures 2.0-1 through 2.0-6), Type II tanks with initially degraded liner (e.g. Tank 15) (Figures 2.0-7 through 2.0-12), and Type IV tanks (e.g. Tank 21) (Figures 2.0-13 through 2.0-17) are provided for reference for the discussion in Section 3. Note the chemical and hydraulic degradation of the other cementitious components (roof, wall, and basemat) can be important to radionuclide transport; however, the transition times are not presented here because in general, their transitions are secondary to the transitions in the annulus (Type II tanks), waste tank grout, and CZ.

Figure 2.0-1: Waste Tank Grout Chemical Transitions - Type II Tank

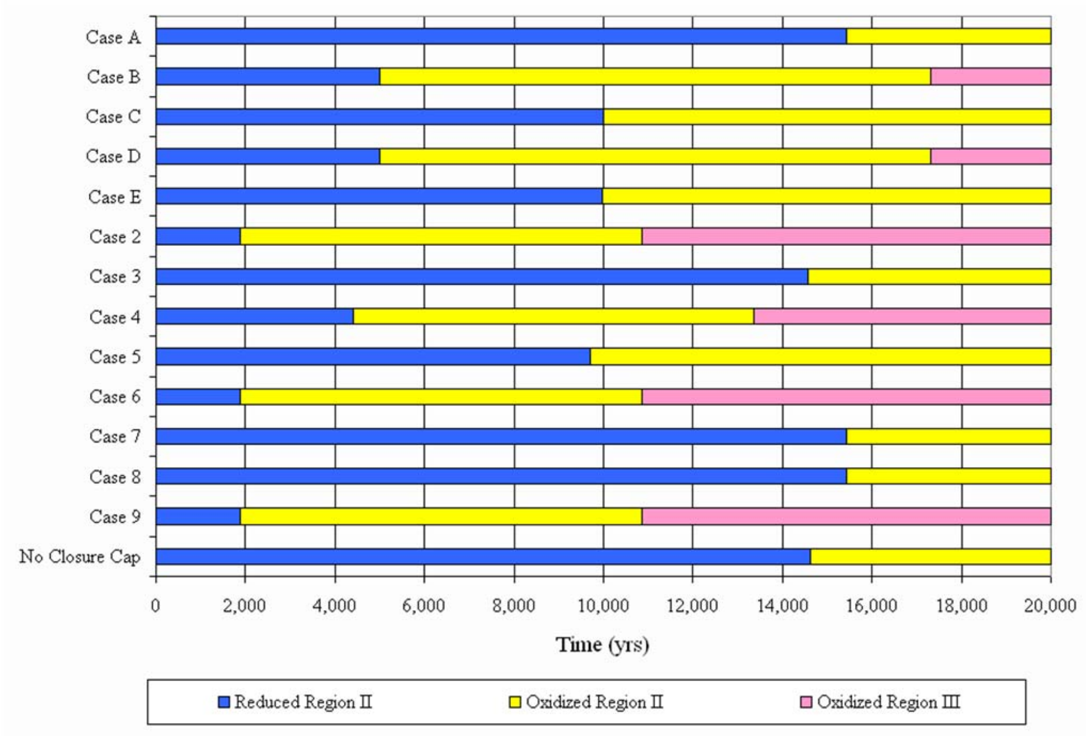


Figure 2.0-2: CZ Chemical Transitions - Type II Tank

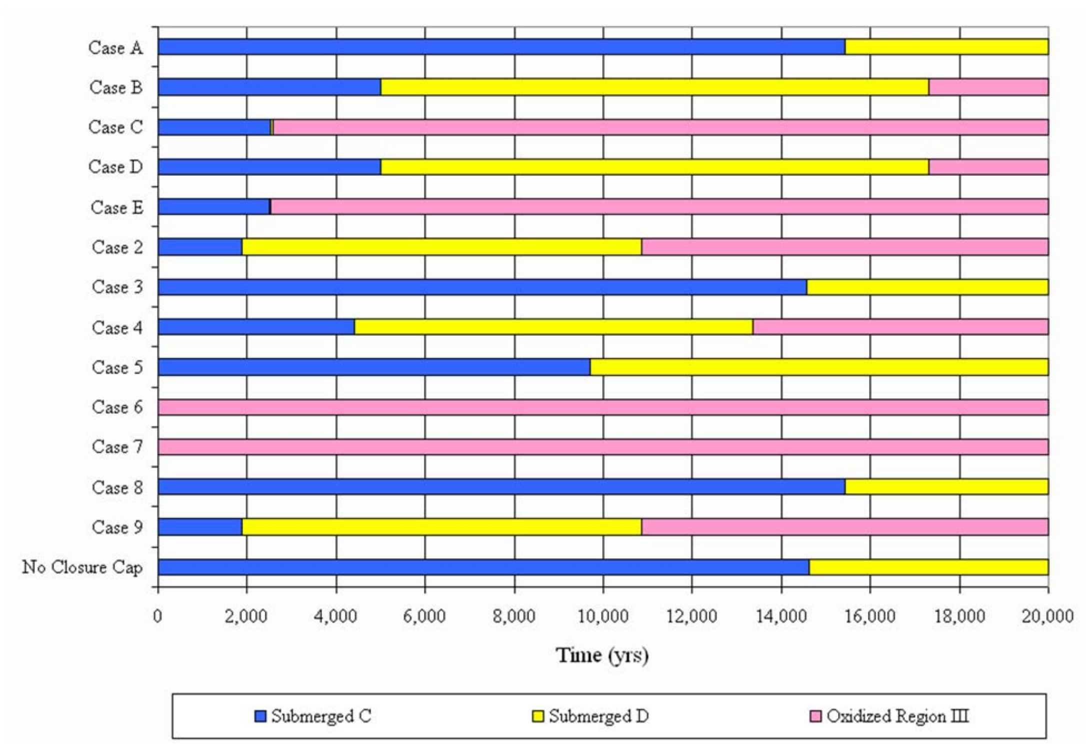


Figure 2.0-3: Annulus Chemical Transitions - Type II Tank

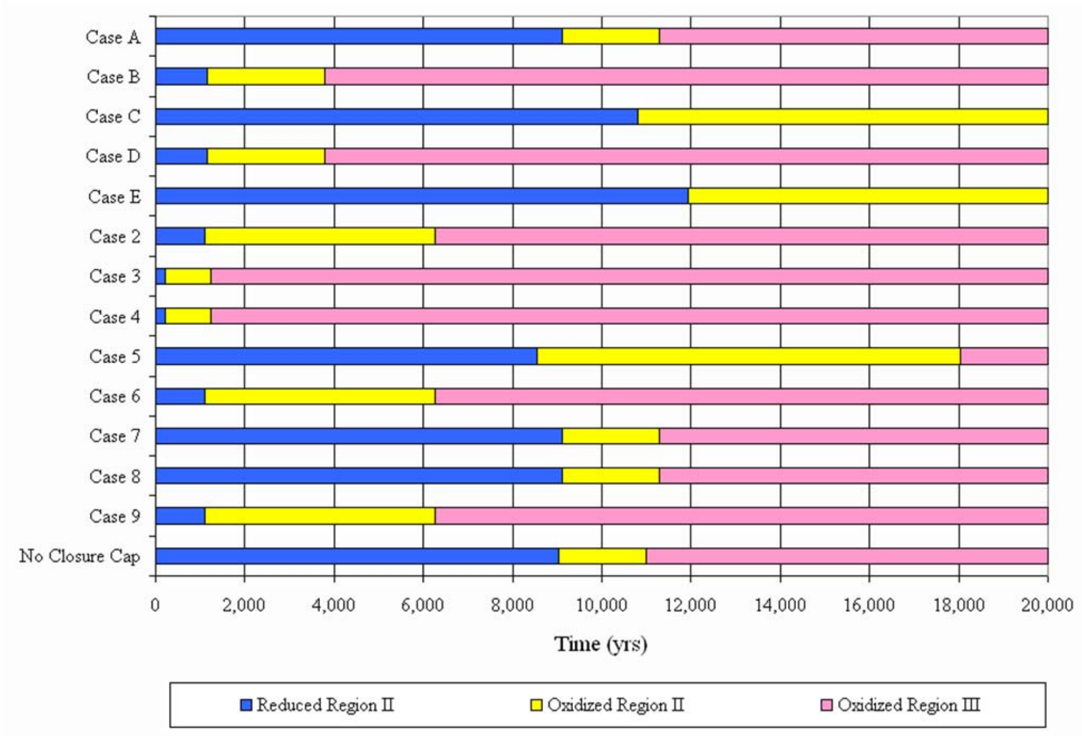


Figure 2.0-4: Waste Tank Grout and Annulus Hydraulic Degradation - Type II Tank

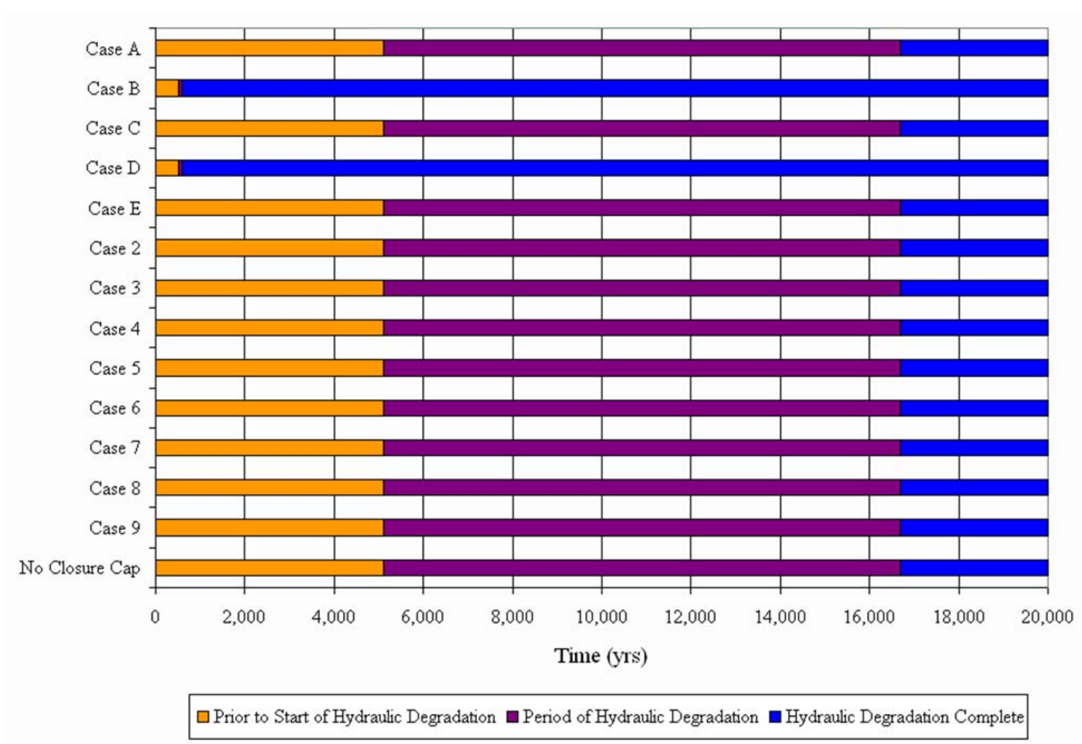


Figure 2.0-5: CZ Hydraulic Degradation - Type II Tank

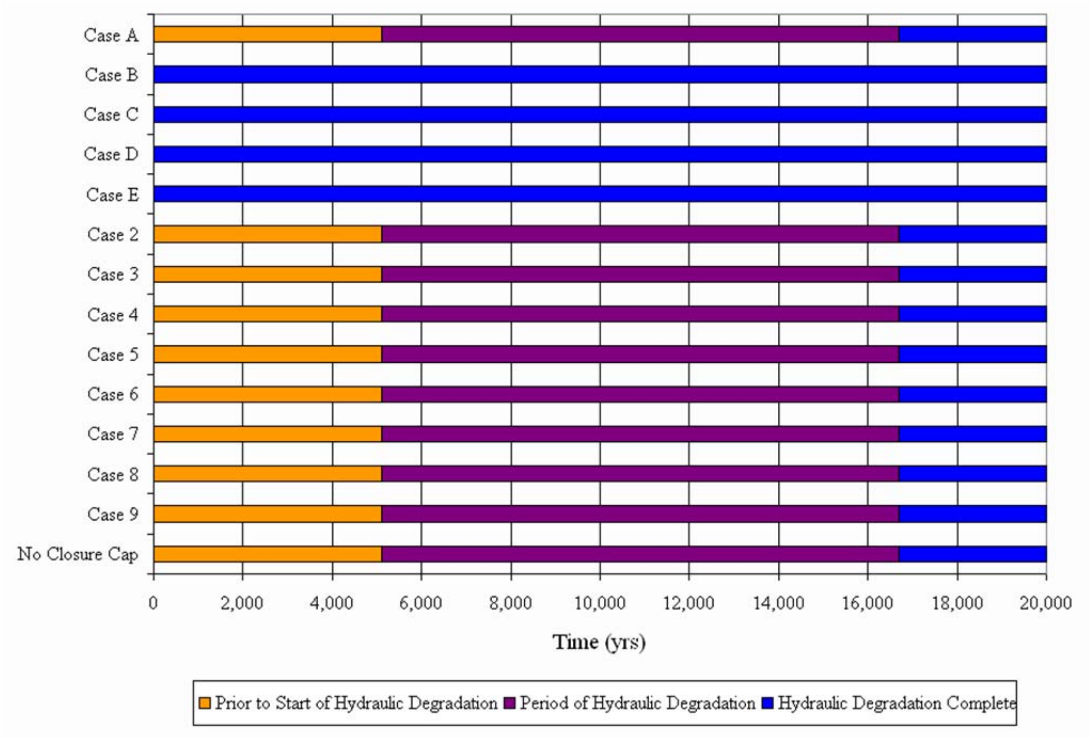


Figure 2.0-6: Waste Tank Liner Failure - Type II Tank

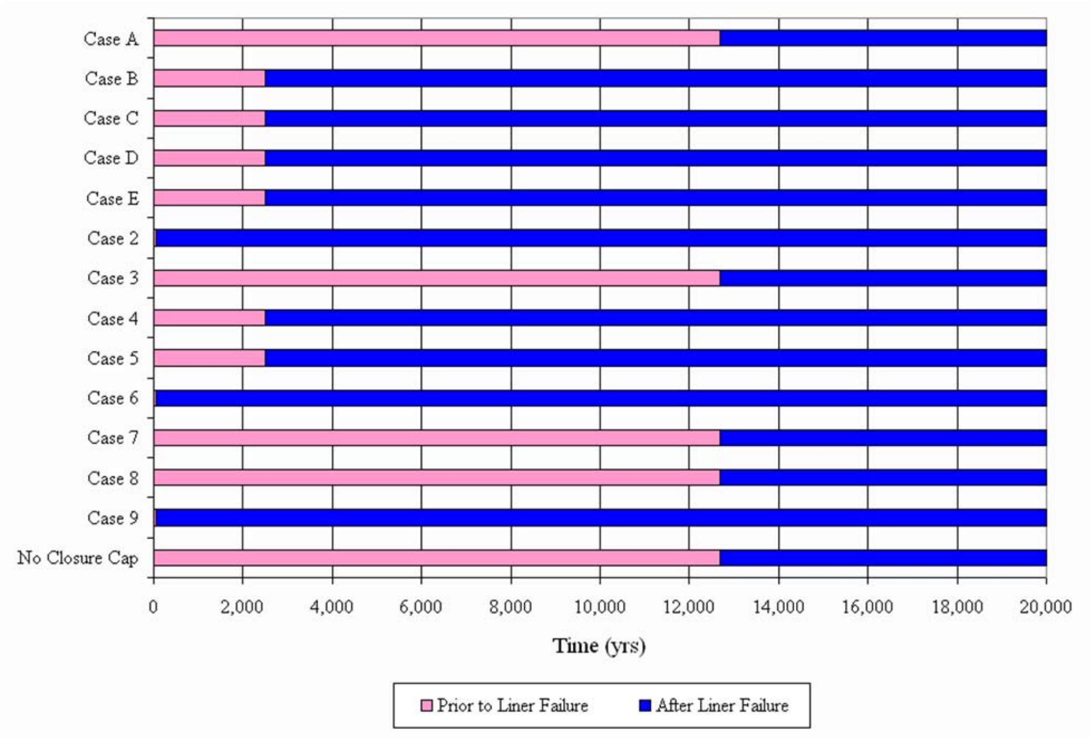


Figure 2.0-7: Waste Tank Grout (No Liner) Chemical Transitions - Type II Tank

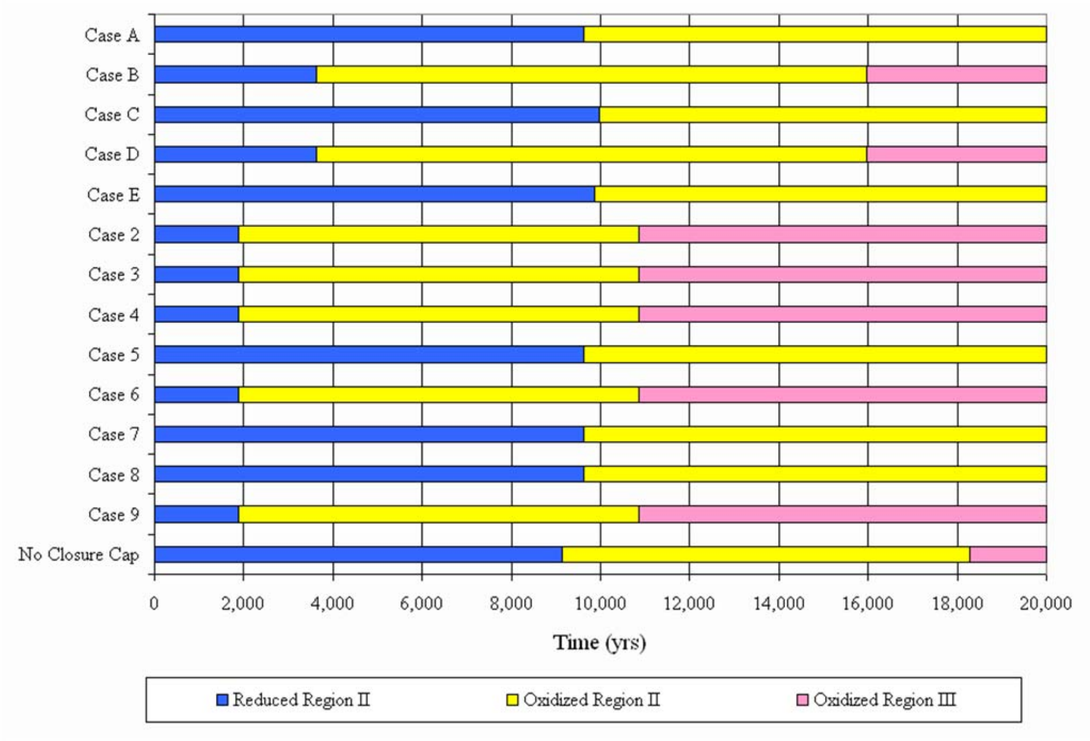


Figure 2.0-8: CZ (No Liner) Chemical Transitions - Type II Tank

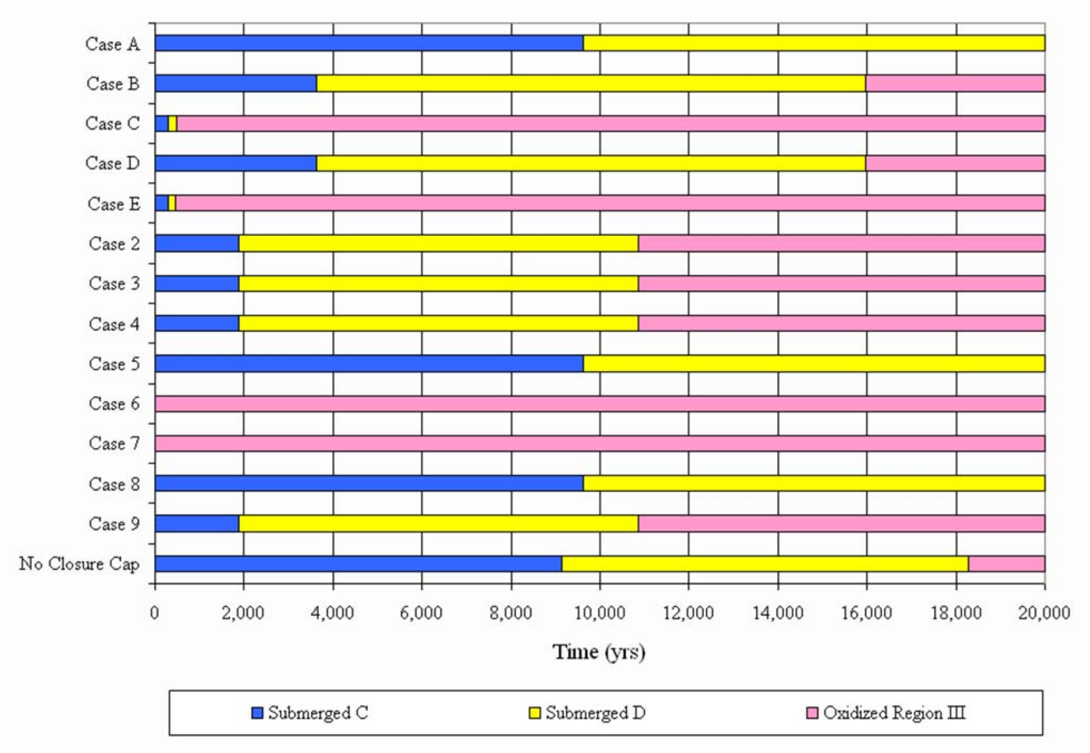


Figure 2.0-9: Annulus (No Liner) Chemical Transitions - Type II Tank

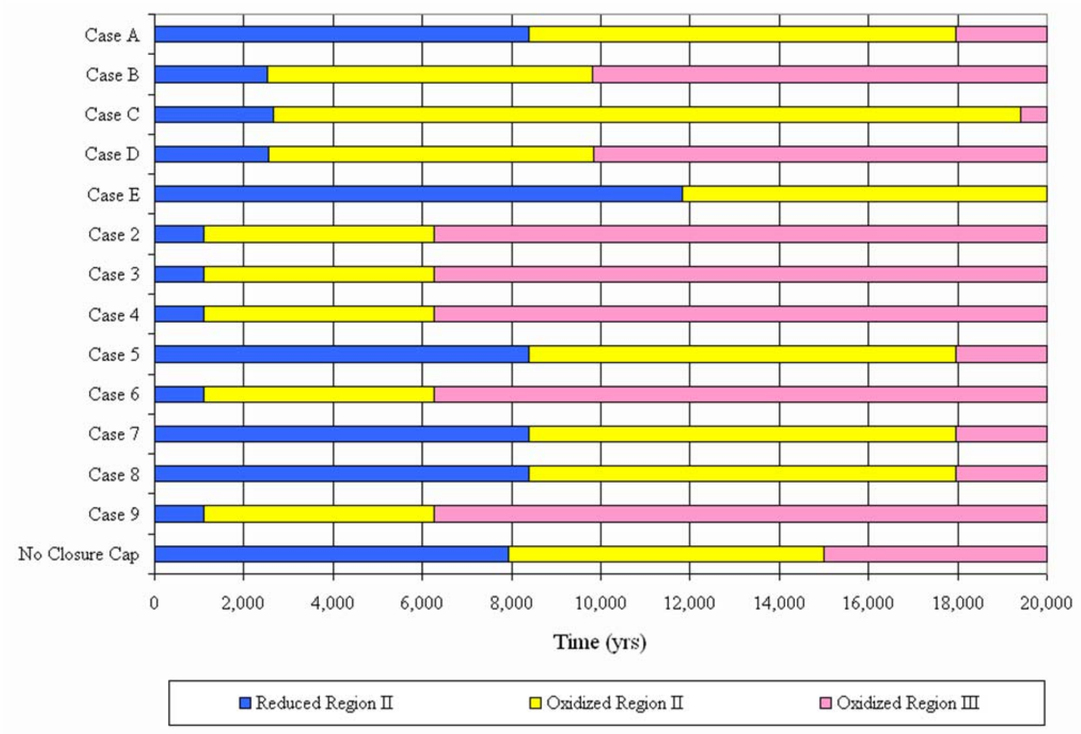


Figure 2.0-10: Waste Tank Grout (No Liner) Hydraulic Degradation - Type II Tank

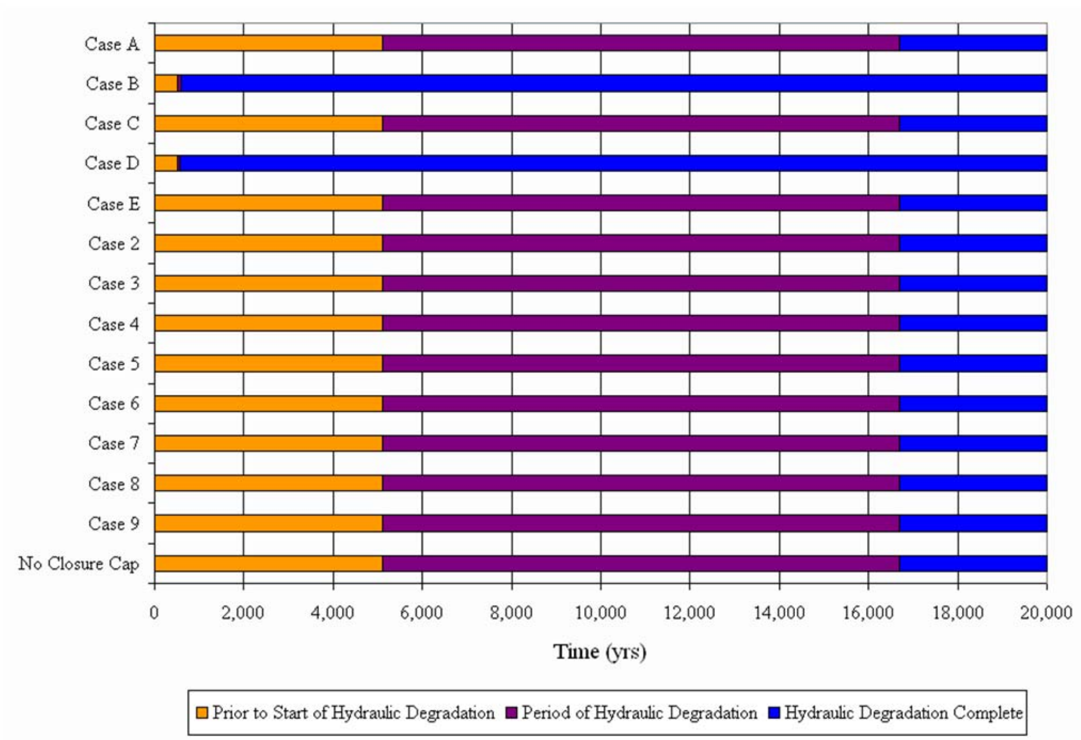


Figure 2.0-11: CZ (No Liner) Hydraulic Degradation - Type II Tank

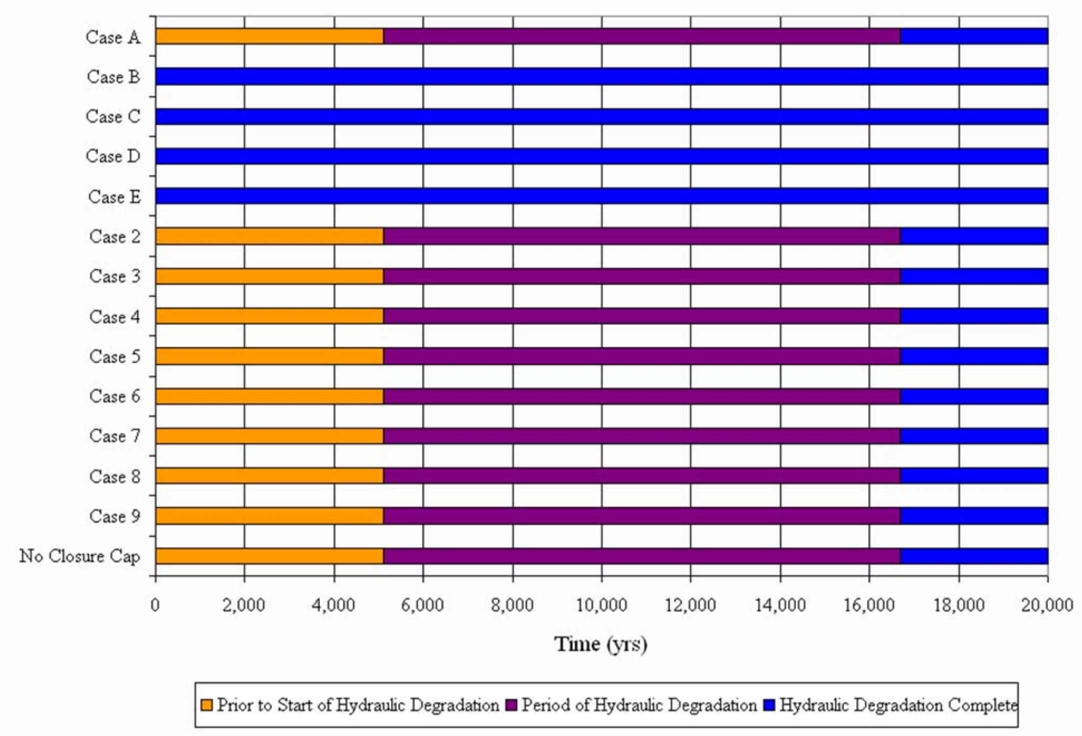


Figure 2.0-12: Annulus (No Liner) Hydraulic Degradation - Type II Tank

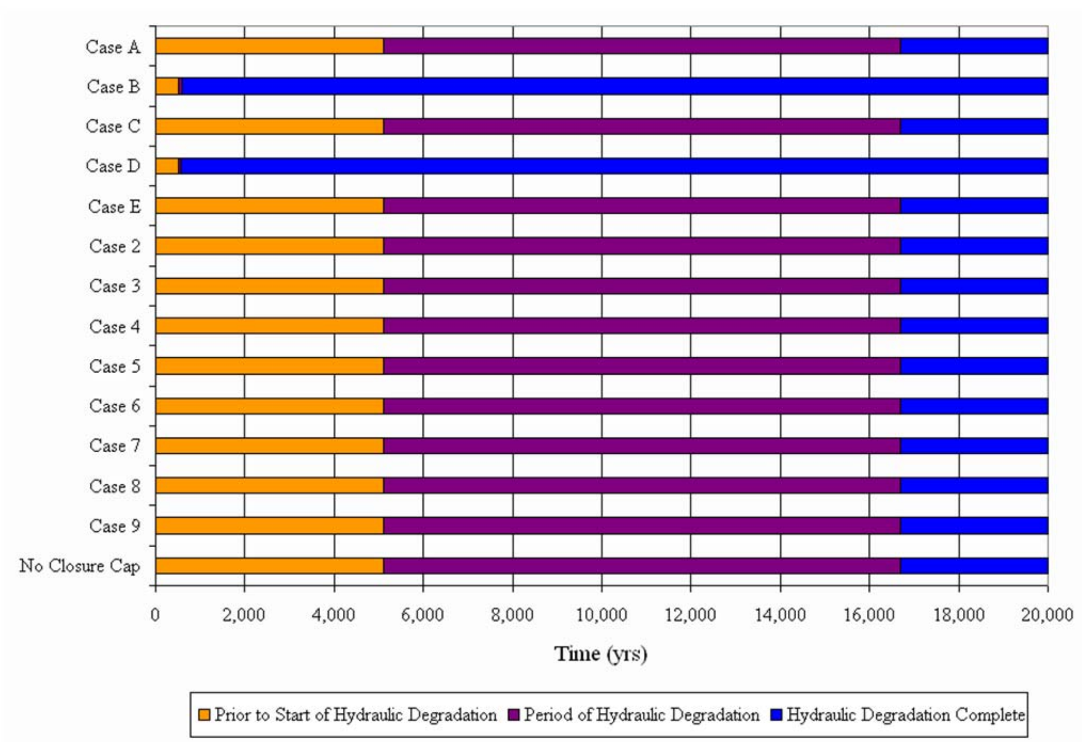


Figure 2.0-13: Waste Tank Grout Chemical Transitions - Type IV Tank

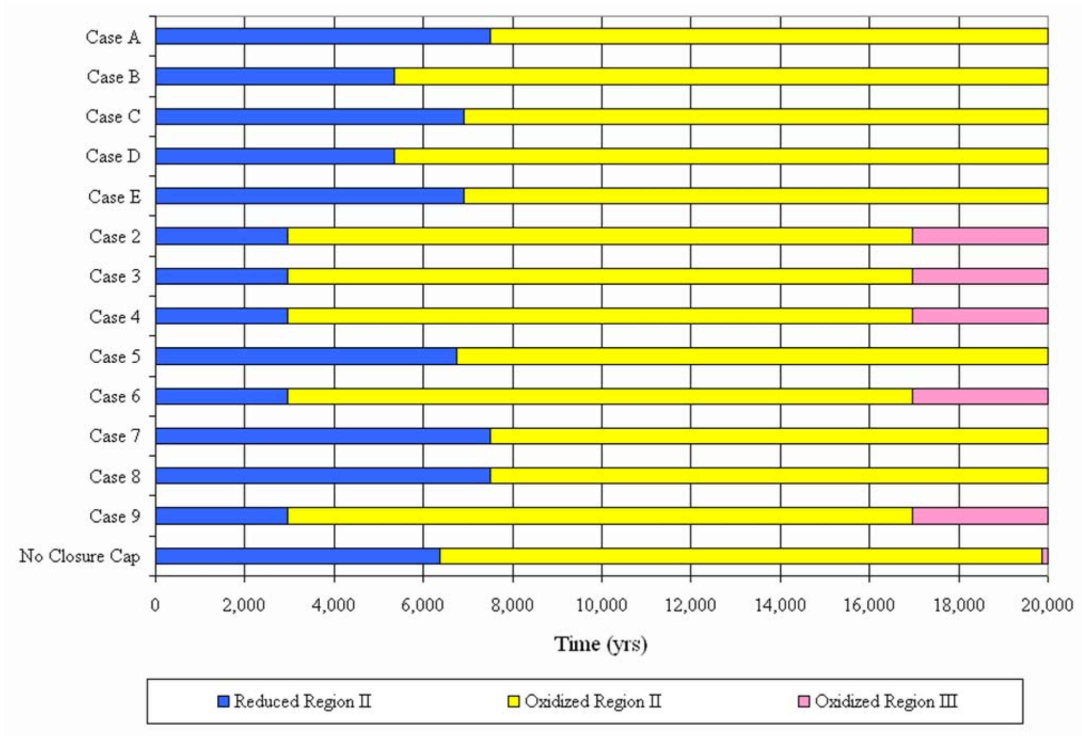


Figure 2.0-14: CZ Chemical Transitions - Type IV Tank

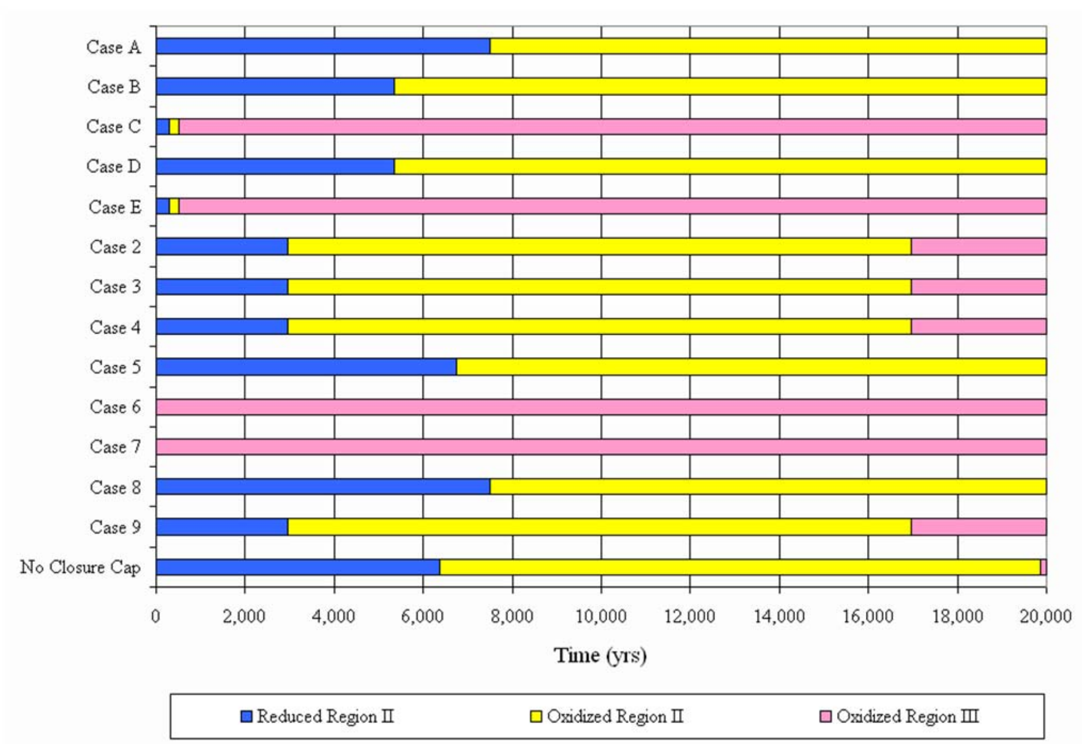


Figure 2.0-15: Waste Tank Grout Hydraulic Degradation - Type IV Tank

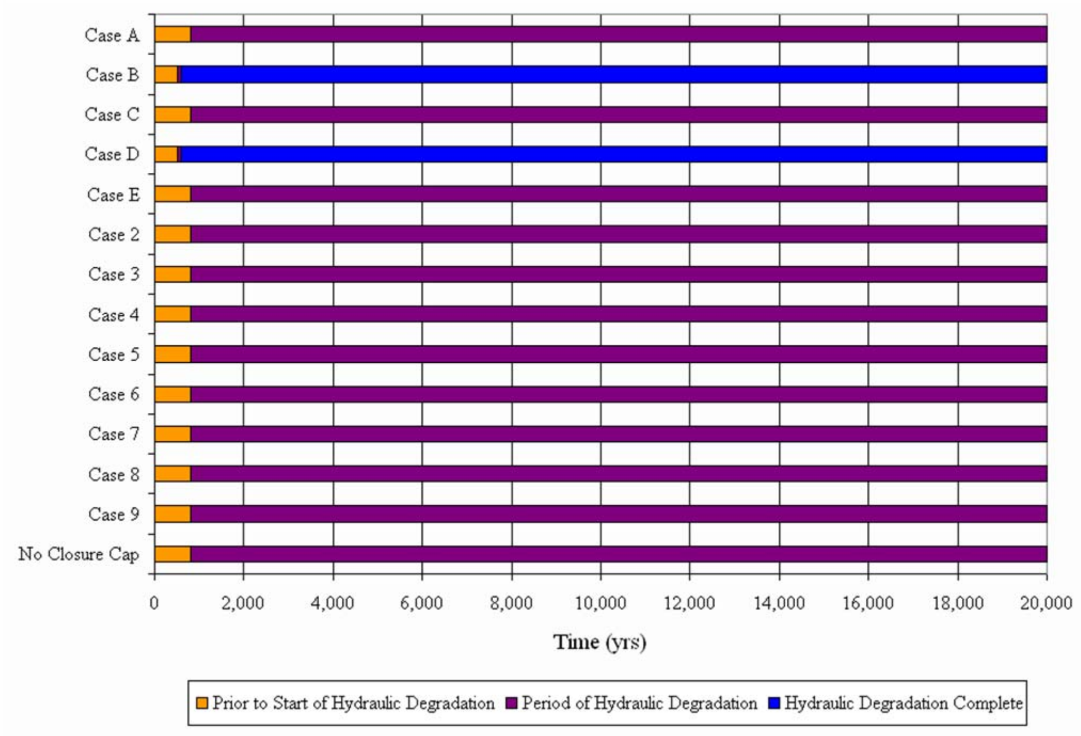


Figure 2.0-16: CZ Hydraulic Degradation - Type IV Tank

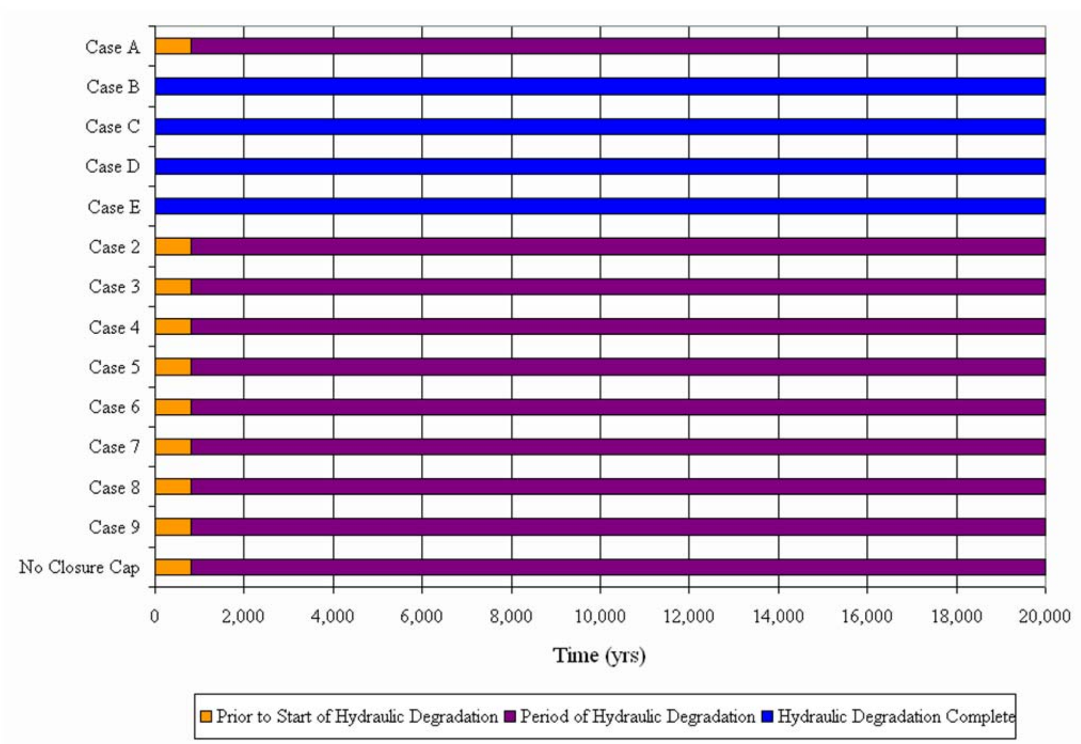
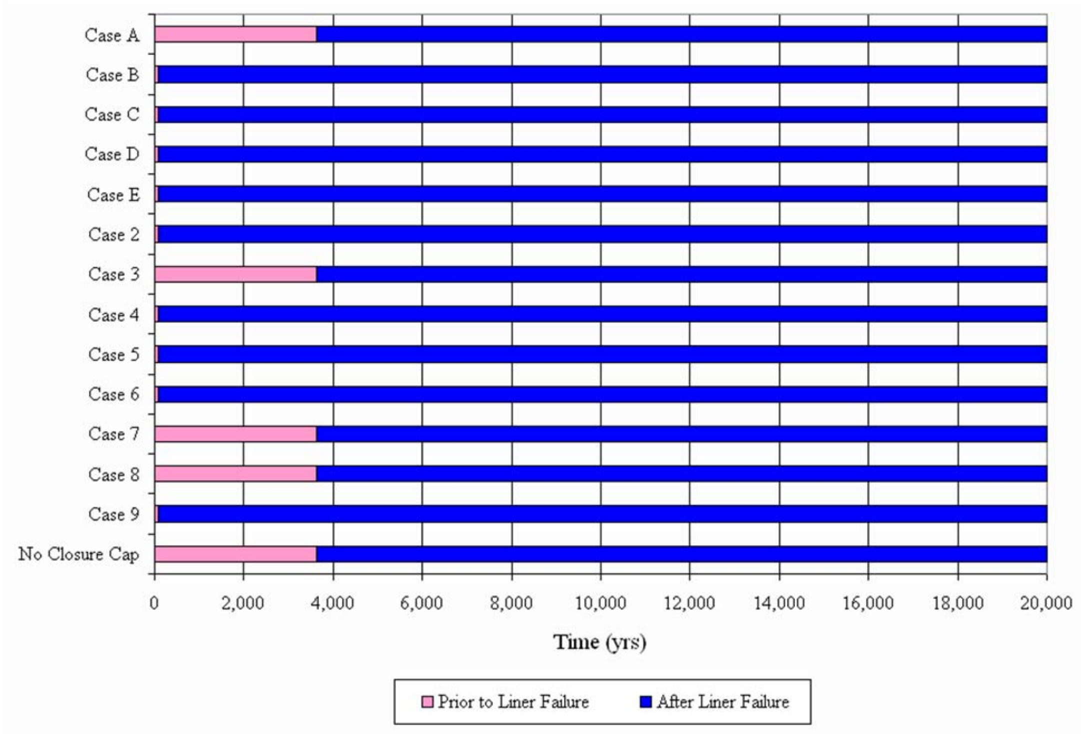


Figure 2.0-17: Waste Tank Liner Failure - Type IV Tank



3.0 ANALYSIS BY BARRIER

This section demonstrates the importance of each barrier under varying possible conditions by analyzing the results from the different barrier analysis configurations with respect to timing and magnitude of peak fluxes below the waste tanks. The simulations were run for 20,000 years to evaluate long term barrier performance. This section also considers each barrier's ability to prevent or retard radionuclide migration, by considering the behavior of the radionuclide under failed and nominal barrier conditions.

The following five barriers are considered here: the closure cap, waste tank grout and cementitious material, CZ, waste tank liner, and the vadose zone. The faster-moving radionuclides, Tc-99, Ra-226, and I-129 are graphed on a single plot for each barrier separately from the slower-moving radionuclides, Np-237 and Pu-239. Flux results for these slower radionuclides were generally smaller values; therefore, the Y-axes for these figures vary according to the magnitudes of the plotted results. [SRR-CWDA-2010-00128]

3.1 Closure Cap (Case A vs. No Cap)

Closure cap barrier capability is estimated by comparing the timing and magnitude of peak fluxes and the time history curves for Case A and the No Cap Barrier Case. Using the Hydrologic Evaluation of Landfill Performance (HELP) model, the closure cap reaches an approximate steady state infiltration rate in year 2,625 of 11.5 in/yr for Case A, compared to 16.45 in/yr without a closure cap. [WSRC-STI-2007-00184 Figure 29 and Table 31] The No Cap Barrier Case is analogous to modeling a "soils only" closure cap with no impermeable barrier, lateral drainage, or erosion control layers. This analysis evaluates fluxes out of the waste tanks with the increased infiltration rate to understand the impact of removing the engineered closure cap barrier on the transport behavior of specific radionuclides.

Table 3.1-1 presents a comparison of the transitions times for select barriers within the Case A and the No Cap models. This table provides a summary of the relevant information depicted in Section 2.

Table 3.1-1: Select Transition Times by Waste Tank –Case A and No Cap Case

	Year of Occurrence	
Type of Transition	**Case A**	**No Cap**
All Tank Types		
Closure cap reaches approximate steady state infiltration rate (11.5 in/yr for Case A and 16.45 in/yr for No Cap)	2,625	0
Type II Tank - Tank 13 (Figures 2.0-1 through 2.0-6)		
Liner fails hydraulically	12,687	12,687
Annulus grout (Submerged C to Submerged D)	9,126	9,022
Annulus grout (Submerged D to Oxidized Region III)	11,291	10,996
Tank Grout and CZ (Submerged C to Submerged D)	15,418	14,626
Tank Grout and CZ (Submerged D to Oxidized Region III)	>20,000	>20,000
Type II Tank (No Liner) - Tank 15 (Figures 2.0-7 through 2.0-12)		
Liner fails hydraulically	N/A	N/A
Annulus grout (Submerged C to Submerged D)	8,392	7,919
Annulus grout (Submerged D to Oxidized Region III)	17,949	14,991
Tank Grout and CZ (Submerged C to Submerged D)	9,615	9,137
Tank Grout and CZ (Submerged D to Oxidized Region III)	>20,000	18,274
Type IV Tank - Tank 21 (Figures 2.0-13 through 2.0-17)		
Liner fails hydraulically	3,638	3,638
Basemat (Oxidized Region II to Oxidized Region III)	3,936	3,850
Tank Grout and CZ (Reducing Region II to Oxidized Region II)	7,491	6,363
Tank Grout and CZ (Oxidized Region II to Oxidized Region III)	>20,000	19,878

Note: Information extracted from HTF PA PORFLOW results. [SRR-CWDA-2010-00128]
N/A = Not Applicable

Figures 3.1-1 through 3.1-3 compare Case A fluxes with the No Cap fluxes from Tank 13 (submerged Type II tank with initially intact liner), Tank 15 (submerged Type II tank with initially degraded liner), and Tank 21 (unsaturated Type IV tank), respectively. The figures illustrate the impact of the closure cap on the fast-moving radionuclides: Tc-99, Ra-226, and I-129. All three figures indicate that inclusion of the closure cap (Case A) provides a small delay and reduction in radionuclide releases to the saturated zone. Removal of the closure cap in the No Cap Barrier case allows for increased flow to the vadose zone leading to an increase in the mass released from the CZ. The increase in flow also results in slightly earlier chemical transition times in the different waste tank components (Table 3.1-1). The releases from the annulus, sand pads (Type II tanks only), waste tank grout, and CZ are most influenced by earlier transition times. The waste tank grout transition time coincides with the CZ transition times.

Figure 3.1-1: Fast-Moving Radionuclide Fluxes Tank 13 (No Cap)

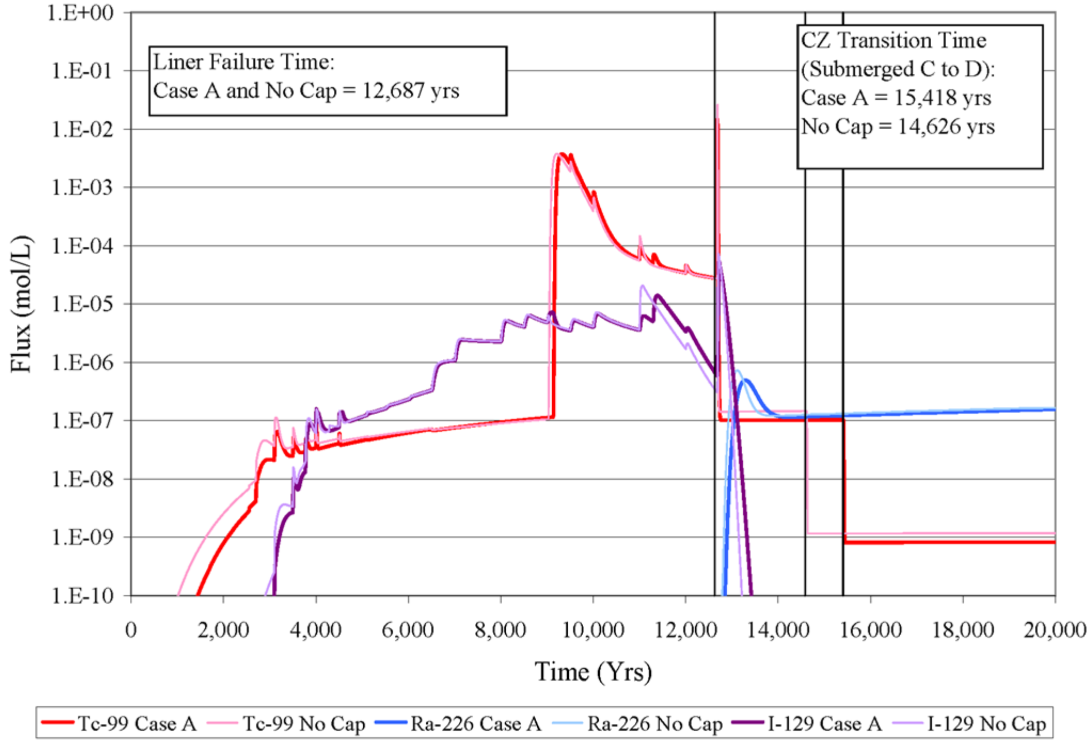


Figure 3.1-2: Fast-Moving Radionuclide Fluxes Tank 15 (No Cap)

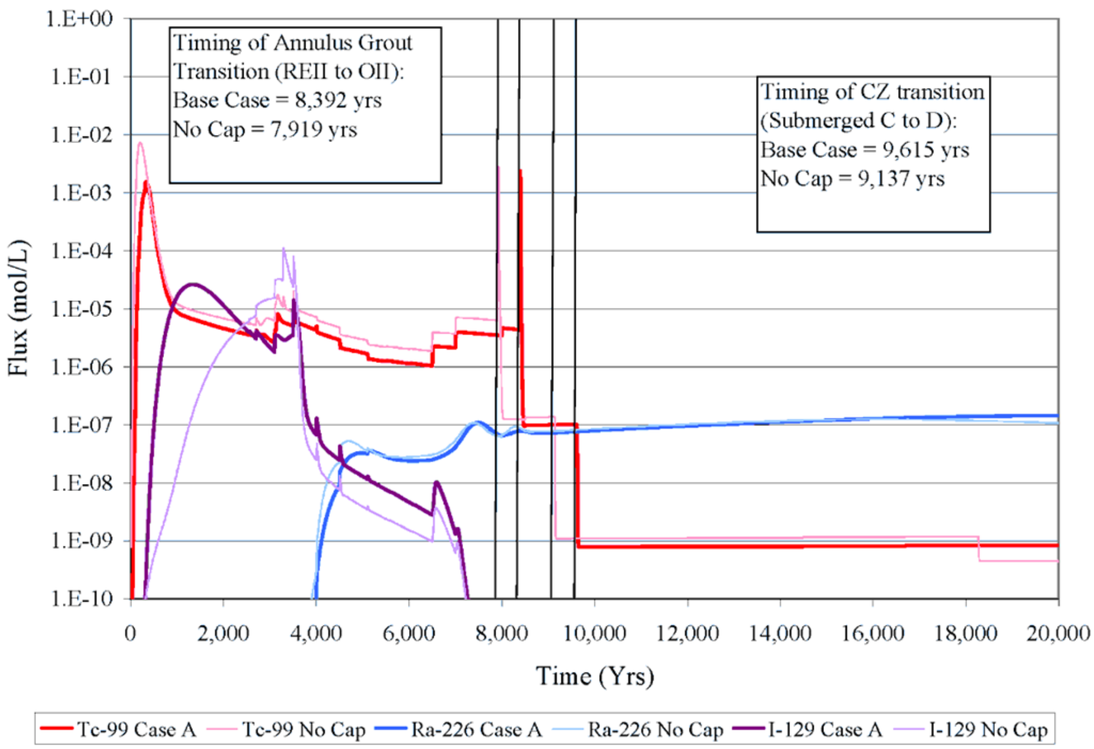
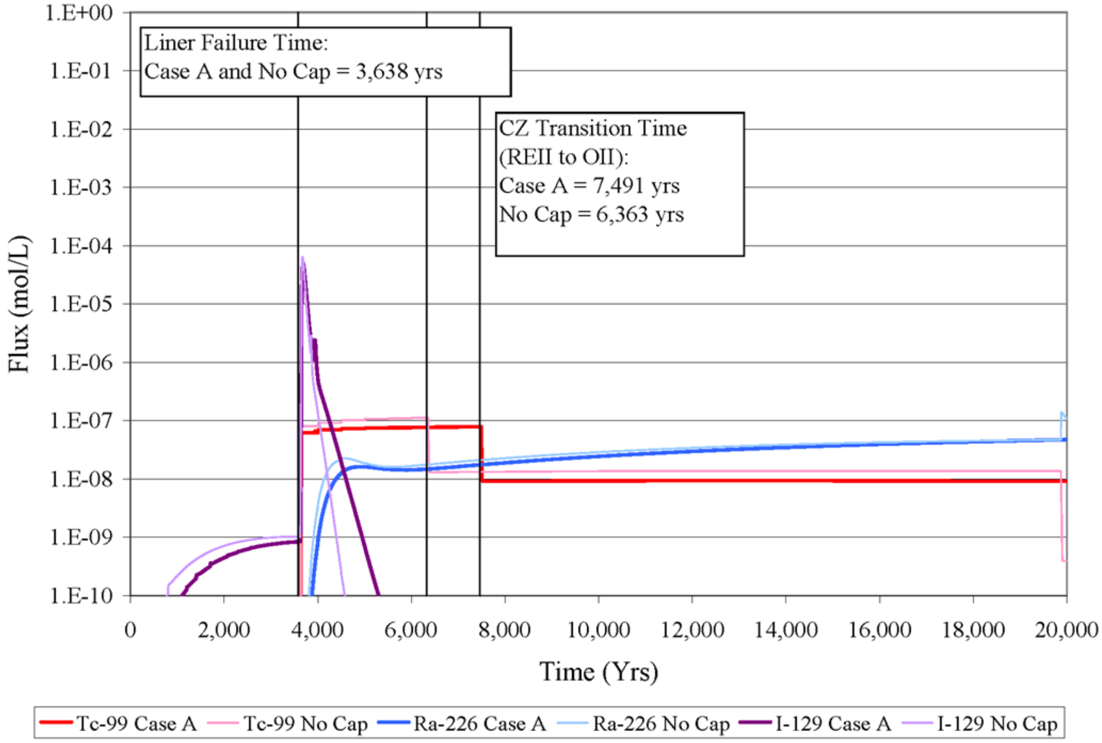


Figure 3.1-3: Fast-Moving Radionuclide Fluxes Tank 21 (No Cap)



Comparison of the timing and magnitude of the peak fluxes for each radionuclide from the three waste tanks presented in Figures 3.1-1 through 3.1-3 indicate that for Tc-99, the No Cap Barrier Case has the most impact on releases from Tank 15, the submerged Type II tank with an initially degraded liner (Figure 3.1-2). The Tc-99 peak flux from this waste tank occurs approximately 200 years earlier than in Case A and is higher by less than an order of magnitude. In contrast, the I-129 peak flux is delayed several thousand years, but peaks higher from Tank 15 in the No Cap Barrier Case compared to the Case A, while Ra-226 behaves similarly in all waste tanks types (e.g., initial Ra-226 flux occurs earlier and the peak is slightly higher in magnitude in the No Cap Barrier Case).

The closure cap is moderately effective as a barrier for those mobile radionuclides not greatly influenced by sorption onto oxidized cementitious material (e.g., Tc-99, Ra-226, and I-129). The impact of the closure cap is greater for the slow-moving radionuclides, Pu-239 and Np-237; however, because these radionuclides take time to move through the system, the impact is only relevant later (> 5,000 years). Plutonium and neptunium move slowly through the system due to high K_d values in cementitious materials. Figure 3.1-4 and Figure 3.1-5 compare fluxes from Tank 15 and Tank 21, respectively, for Case A to the No Cap case. Tank 13 tank fluxes for Np-237 and Pu-239 are so low that they are not plotted here. The increased flow in the No Cap Barrier Case causes earlier and higher magnitude fluxes of Pu-239 and Np-237 out of the waste tanks. As the cementitious materials become more oxidizing with time, more of the highly sorbed Pu-239 and Np-237 are released in the No Cap Barrier Case. While the less sorptive fast-moving radionuclides show only a moderate impact from the closure cap, Pu-239 and Np-237 show about an order of magnitude increase in flux or more without the closure cap.

Figure 3.1-4: Slow-Moving Radionuclide Fluxes Tank 15 (No Cap)

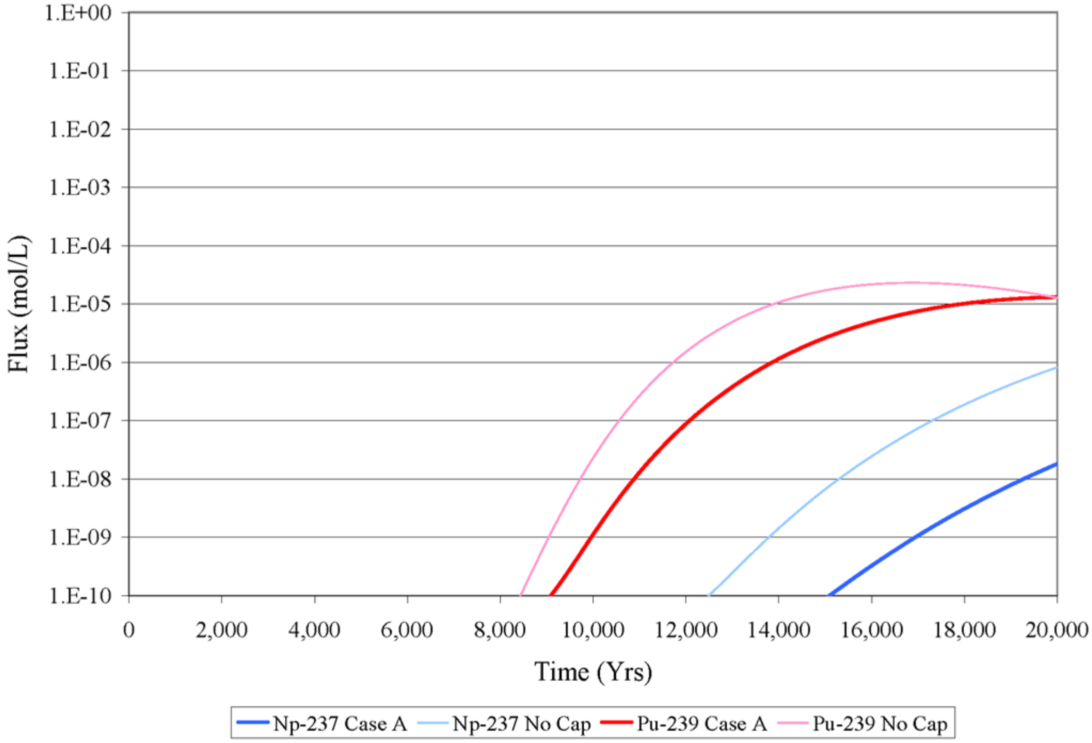
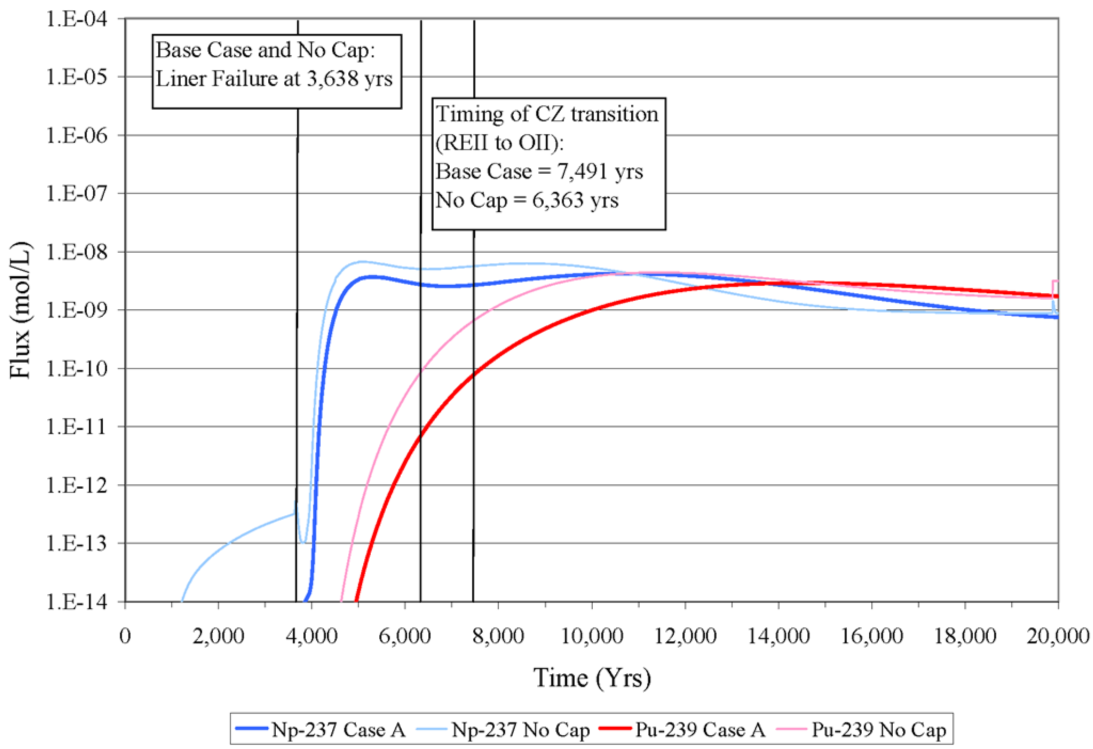


Figure 3.1-5: Slow-Moving Radionuclide Fluxes Tank 21 (No Cap)



This analysis indicates that the presence of the closure cap has the greatest impact on waste tanks with initially degraded liners. Therefore, the importance of the closure cap relies on the performance of the steel liners.

3.2 Grout and Cementitious Material (Alternate Configuration Case B vs. Case C)

The radionuclide flux results from selected waste tanks for the alternative scenarios Case B and Case C (as described in the HTF PA) were used to evaluate the impact of the grout as a barrier to radionuclide migration. Both Case B and Case C were modeled with a fast flow path through the grout and early failure of the waste tank liners based on the times presented in Table 4.2-36 of the HTF PA. [SRR-CWDA-2010-00128] As previously described, an assumed "fast flow path" is modeled by modifying the hydraulic conditions of a portion of the tank properties such that a channel of faster flow rates exists through various barriers within the waste tank. Case B differs from Case C in that the waste tank grout, annulus grout, and other cementitious barriers in the former case hydraulically failed at year 501, while Case C gradually fails these barriers (see HTF PA Table 4.2-34). Additionally, for Case B the full reducing capacity of the grout is imparted on the CZ, as would be the case with waste tank grout that is hydraulically degraded; whereas Case C was modeled as having the reducing capacity of the CZ only. The dominant flow for Case C is through the fast flow channel on the edge of the grout, bypassing the influence of this reducing zone. Comparing these cases provided insight to the effectiveness of the intact waste tank grout on the fluxes.

Figures 3.2-1 through 3.2-3 compare Case B fluxes with Case C fluxes from Tank 13 (submerged Type II tank with initially intact liner), Tank 15 (submerged Type II tank with initially degraded liner), and Tank 21 (unsaturated Type IV tank), respectively. The figures illustrate the impact of (1) the grout reducing capacity and (2) grout integrity on the migration of fast-moving radionuclides (Tc-99, Ra-226, and I-129). Relevant transition times for the two cases are compared by waste tank in Table 3.2-1. This table provides a summary of the relevant information depicted in Section 2 (Figures 2.0-1, 2.0-3, 2.0-7, 2.0-9, and 2.0-13).

Releases of fast-moving radionuclides (Tc-99, Ra-226, and I-129) from Tank 13 are compared for Cases B and C in Figure 3.2-1. A vertical line on the graph indicates the liner failure time for both cases at 2,506 years. Significant releases of Tc-99 and I-129 occur prior to the liner failure in Case B, but not Case C, due to the early hydraulic degradation (at 500 to 585 years, Figure 2.0-4) of the annulus grout in Case B. This allows rapid transport of inventory out of the annulus. In Case B, transport of Tc-99 out of the annulus is further enhanced due to a decrease of Tc-99 sorption (because of the chemical transition at 1,143 years, Figure 2.0-3)). When the liner fails, a pulse of radionuclides leaves the waste tank (from the CZ). Releases for Case B following liner failure are greatly suppressed by several orders of magnitude relative to Case C, especially for Tc-99 and I-129. In the absence of high flow rates in the waste tank grout in Case C, upward diffusion of Tc-99, I-129 and Ra-226 from the CZ becomes an important process, promoting storage of these radionuclides within the chemically reducing grout. This explains the much higher release rates over time in Case C after liner failure, as transport of the radionuclides finds another path of exit, from the grout through the annulus and wall.

Figure 3.2-1: Fast-Moving Radionuclide Fluxes Tank 13 (Grout)

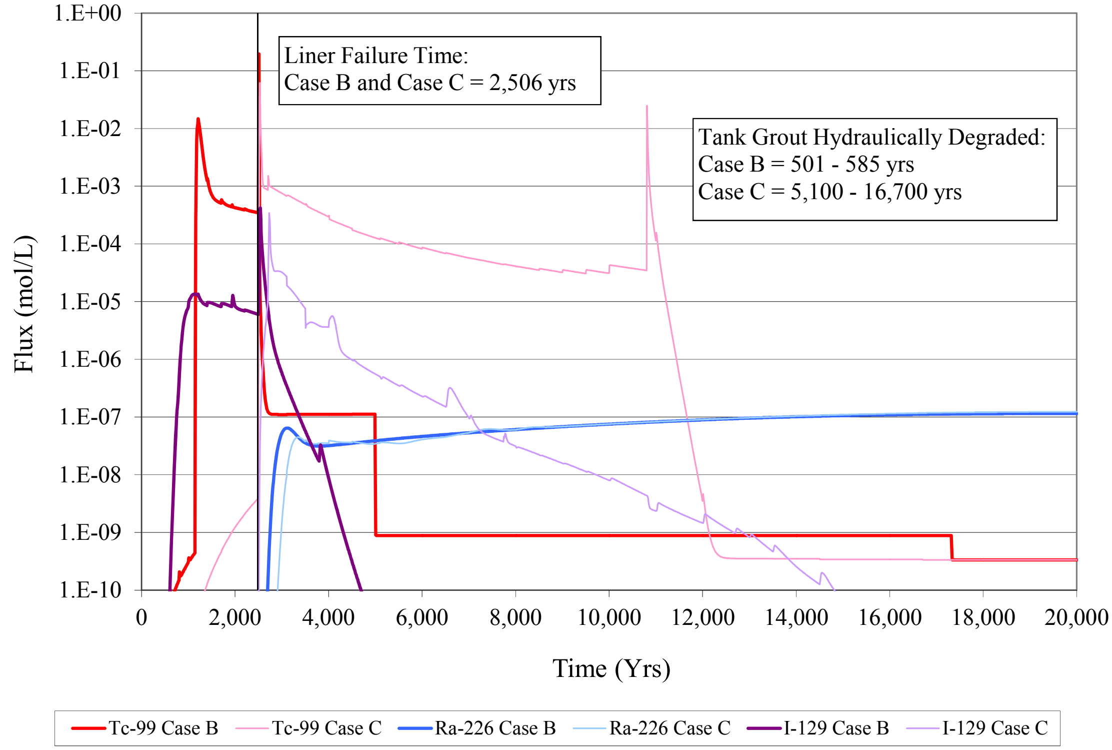


Figure 3.2-2: Fast-Moving Radionuclide Fluxes Tank 15 (Grout)

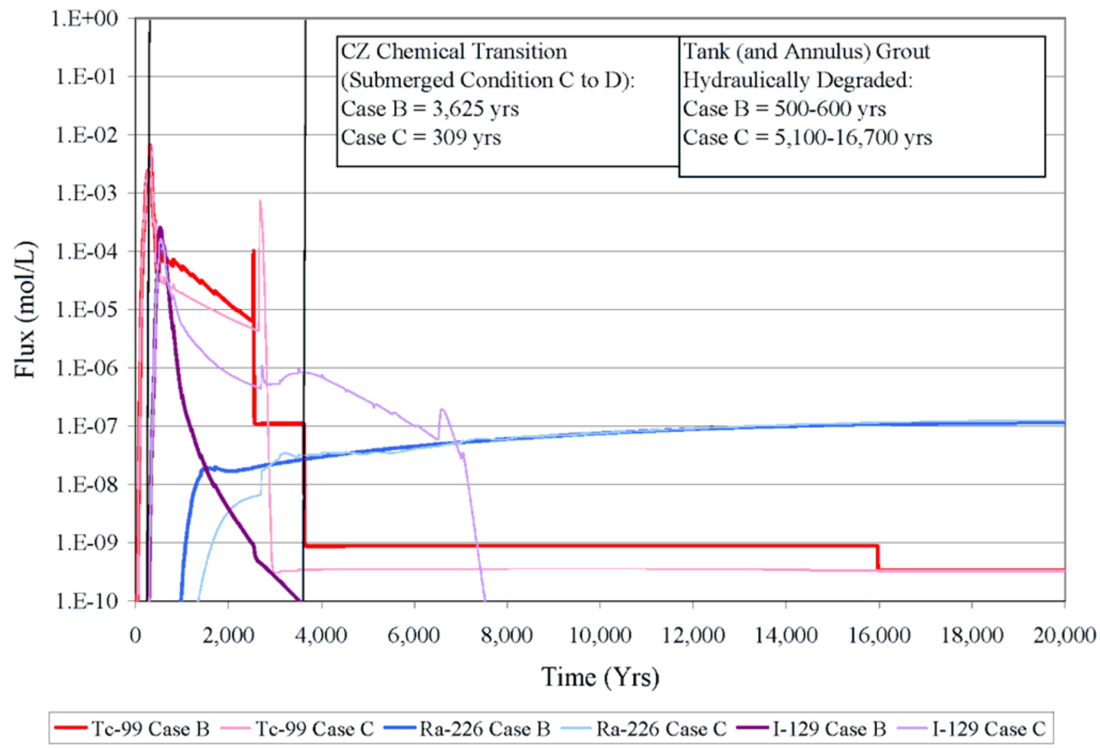


Figure 3.2-3: Fast-Moving Radionuclide Fluxes Tank 21 (Grout)

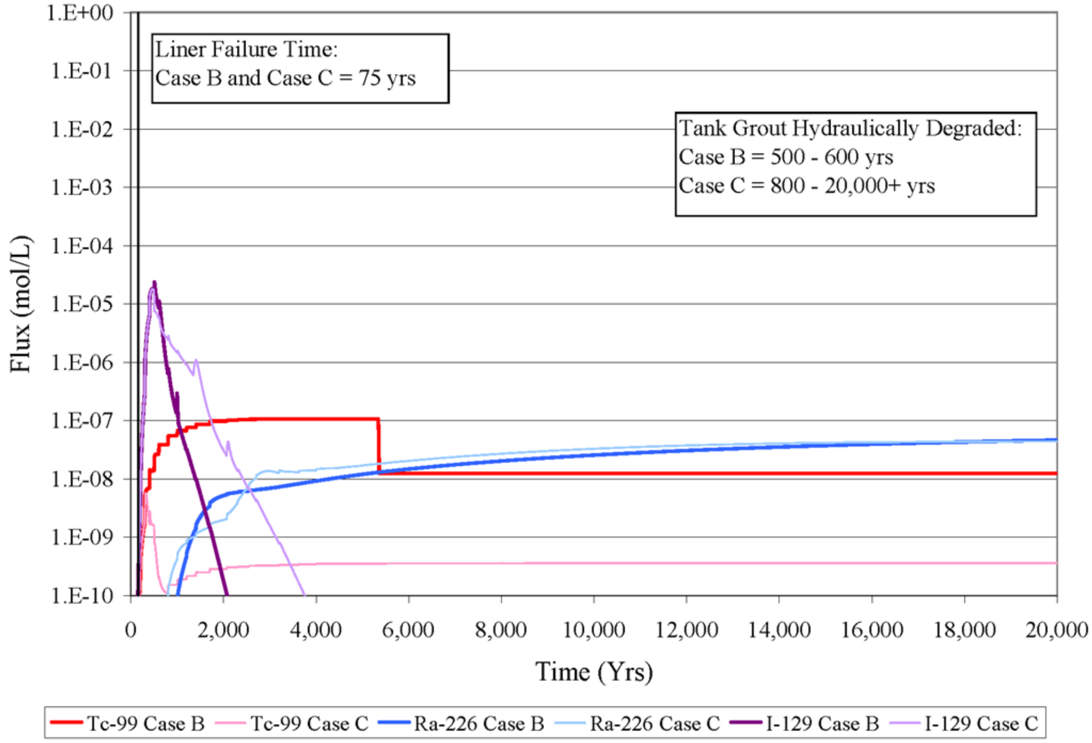


Table 3.2-1: Select Transition Times by Waste Tank - Cases B and C

	Year of Occurrence	
Type of Transition	**Case B**	**Case C**
Type II Tank - Tank 13 (Figures 2.0-1 through 2.0-6)		
Liner fails hydraulically	2,506	2,506
Basemat (Oxidized Region II to Oxidized Region III)	585	2,719
Cementitious material degrades hydraulically	500-585	2,550-5,100
Waste tank grout degrades hydraulically	500-585	5,100-16,700
Annulus grout (Reducing Region II to Oxidized Region II)	1,143	10,805
Waste tank grout (Reducing Region II to Oxidized Region II)	4,990	9,993
CZ (Submerged C to Submerged D)	4,990	2,518
CZ (Submerged D to Oxidized Region III)	17,323	2,575
Type II Tank (No Liner) - Tank 15 (Figures 2.0-7 through 2.0-12)		
Liner fails hydraulically	N/A	N/A
Basemat (Oxidized Region II to Oxidized Region III)	89	89
Cementitious material degrades hydraulically	500	2,550-5,100
Waste tank grout degrades hydraulically	500-600	5,100-16,700
Annulus grout (Reducing Region II to Oxidized Region II)	2,530	2,657
Waste tank grout (Reducing Region II to Oxidized Region II)	3,625	9,965
CZ (Submerged C to Submerged D)	3,625	309

	Year of Occurrence	
Type of Transition	**Case B**	**Case C**
CZ (Submerged D to Oxidized Region III)	15,969	493
Type IV Tank - Tank 21		
Liner fails hydraulically	75	75
Basemat (Oxidized Region II to Oxidized Region III)	988	1,350
Cementitious material degrades hydraulically	500-600	400-800
Waste tank grout degrades hydraulically	500-600	800->20,000
Annulus grout (Reducing Region II to Oxidized Region II)	NA	NA
Waste tank grout (Reducing Region II to Oxidized Region II)	5,346	6,896
CZ (Reducing Region II to Oxidized Region II)	5,346	302
CZ (Oxidized Region II to Oxidized Region III)	>20,000	501

Note: Information extracted from HTF PA Tables 4.4-4, 4.4-5 and 4.4-9. [SRR-CWDA-2010-00128]
N/A = Not Applicable

Further, Case B Tc-99 releases from the CZ for Tank 13 (Figure 3.2-1) are controlled by the submerged Condition C solubility limit until 4,990 years, at which time the CZ transitions to the lower technetium solubilities prevalent in submerged Condition D solubility limit. For the remainder of the Case B simulation, Tc-99 releases are held at the solubility limit in the CZ. The submerged Condition C solubility limit to Condition D solubility limit transition occurs immediately following the liner failure in Case C, but releases for Case C are mostly coming from the annulus and the waste tank grout until after 12,000 years (Table 3.2-1). Therefore, the step is overshadowed by the change in flow fields accompanying the loss of liner integrity. For Case C, the Tc-99 in the grout is due to upward diffusion from the CZ before liner failure.

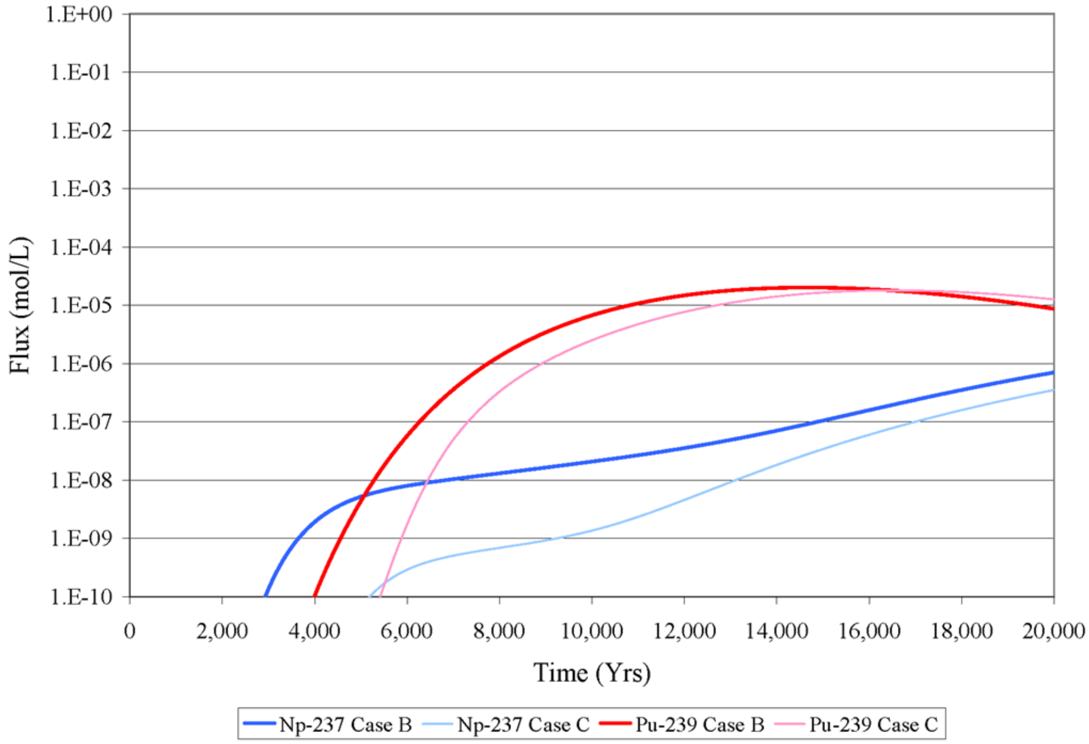
The late Tc-99 spike (at 10,800 years) in Case C is due to the chemical transition of the annulus grout from Reducing Region II to Oxidizing Region II (wherein the Tc-99 K_d reduces significantly from 5,000 mL/g to 0.8 mL/g), effectively flushing the available Tc-99 out of the annulus. Once the Tc-99 is gone from the annulus, the releases revert to the solubility controlled releases out of the CZ (Table 3.2-1).

Figure 3.2-2 shows Tank 15 (which has a degraded liner at year zero), wherein the annulus grout transitions at approximately the same time for both cases. The degraded grout in Case B results in slightly higher I-129 and Ra-226 peaks, but more dramatic is the faster transport out of the system for I-129, which has no solubility limits. Tc-99 in Tank 15 is controlled mostly by solubility limits due to the timing of annulus and CZ transitions forinventory from the sand pads and annulus, both which vary by case (Table 3.2-1).

Figure 3.2-3 illustrates the fast-moving radionuclide release behavior from Tank 21 (Type IV tank). Degraded grout in Case B promotes faster transport of I-129 out of the system, while Tc-99 curves are again predominantly controlled by the solubility limits in the CZ (and there is no sand pad or annulus inventory to be a second source of Tc-99).

Releases of slow-moving radionuclides, Np-237 and Pu-239, from Tank 15 are displayed in Figure 3.2-4. The figure indicates releases from Case B, the case with early grout degradation, are earlier and one to two orders of magnitude greater than releases in Case C. Slow-moving radionuclide releases from Tank 13 and Tank 21 show the same behavior.

Figure 3.2-4: Slow-Moving Radionuclide Fluxes Tank 15 (Grout)



The timings of the chemical transitions (i.e., Reduced Region II → Oxidized Region II → Oxidized Region III) are driven by flow through the cementitious materials. By increasing such flow, either through degradation of the hydraulic properties (e.g., compare Case B to Case C) or by assuming a fast flow path (compare Case A to Case C), these transitions occur earlier, thus expediting the release of solubility-dependent contaminants.

3.3 Contamination Zone (Barrier Case A vs. Case 7, and Case 2 vs. Case 6)

The CZ barrier analysis modeled the CZ without the solubility limits applied to radionuclides to evaluate the retarding affect this barrier has on Case A releases. In Barrier Case 7, all of the barriers are assumed to have nominal conditions except the CZ. Within the CZ, Barrier Case 7 removes all solubility controls for Tc-99, Ra-226, and I-129. Similarly, Barrier Case 6 also removes all solubility controls for Tc-99, Ra-226, and I-129; however this barrier case also assumes that all other barriers are fully degraded. Barrier Case 7 is compared to Case A to evaluate the CZ impact on releases of fast-moving radionuclide under otherwise nominal conditions; Barrier Case 6 is compared to Barrier Case 2 to evaluate the CZ impact on the releases of slow-moving radionuclides under degraded conditions. The fully degraded configuration (Barrier Case 2) modeled the CZ under nominal (i.e., Case A) conditions.

Table 3.3-1 presents a comparison of the transitions times for select barriers within the Case A, Case 6, and Case 7 models. This table provides a summary of the relevant information depicted in Section 2.

Table 3.3-1: Select Transition Times by Waste Tank – Barrier Cases 2, 6, and 7

	Year of Occurrence		
Type of Transition	**Case 2**	**Case 6**	**Case 7**
Type II Tank - Tank 13 (Figures 2.0-1 through 2.0-6)			
Liner fails hydraulically	0	0	12,687
Annulus grout (Submerged C to Submerged D)	1,092	1,092	9,126
Tank Grout (Submerged C to Submerged D)	1,890	1,890	15,418
CZ (Submerged C to Submerged D)	1,890	0	0
Annulus grout (Submerged D to Oxidized Region III)	6,273	6,273	11,291
Tank Grout (Submerged D to Oxidized Region III)	10,860	10,860	>20,000
CZ (Submerged D to Oxidized Region III)	10,860	>20,000	>20,000
Type II Tank (No Liner) - Tank 15 (Figures 2.0-7 through 2.0-12)			
Liner fails hydraulically	N/A	N/A	N/A
Annulus grout (Submerged C to Submerged D)	1,092	1,092	8,392
Tank Grout (Submerged C to Submerged D)	1,890	1,890	9,615
CZ (Submerged C to Submerged D)	1,890	0	0
Annulus grout (Submerged D to Oxidized Region III)	6,273	6,273	17,949
Tank Grout (Submerged D to Oxidized Region III)	10,860	10,860	>20,000
CZ (Submerged D to Oxidized Region III)	10,860	>20,000	>20,000
Type IV Tank - Tank 21 (Figures 2.0-13 through 2.0-17)			
Liner fails hydraulically	0	0	3,638
Tank Grout (Reducing Region II to Oxidized Region II)	2,954	2,954	7,491
CZ (Reducing Region II to Oxidized Region II)	2,954	0	0
Tank Grout (Oxidized Region II to Oxidized Region III)	16,971	16,971	>20,000
CZ (Oxidized Region II to Oxidized Region III)	16,971	>20,000	>20,000
Basemat (Oxidized Region II to Oxidized Region III)	165	165	3,936

Note: Information extracted from HTF PA PORFLOW results. [SRR-CWDA-2010-00128]
N/A = Not Applicable

When the radionuclide aqueous concentration in the CZ reaches the solubility concentration limit (or solubility limit), the radionuclide will precipitate to a solid form, and thus becomes unavailable for aqueous form transport out of the CZ. Low solubility limits in the CZ promote increased precipitation and decreased releases. Thus, it is expected that when the solubility limits in the barrier cases are removed, effectively promoting dissolution and minimizing precipitation, radionuclide releases will increase. The fast-moving radionuclide releases for Tank 13, Tank 15, and Tank 21 are provided in Figures 3.3-1, 3.3-2, and 3.3-3, respectively. The releases from Tank 15 are provided in Figure 3.3-4 as an example of the release behavior for the slow-moving radionuclides.

Figure 3.3-1: Fast-Moving Radionuclide Fluxes Tank 13 (CZ)

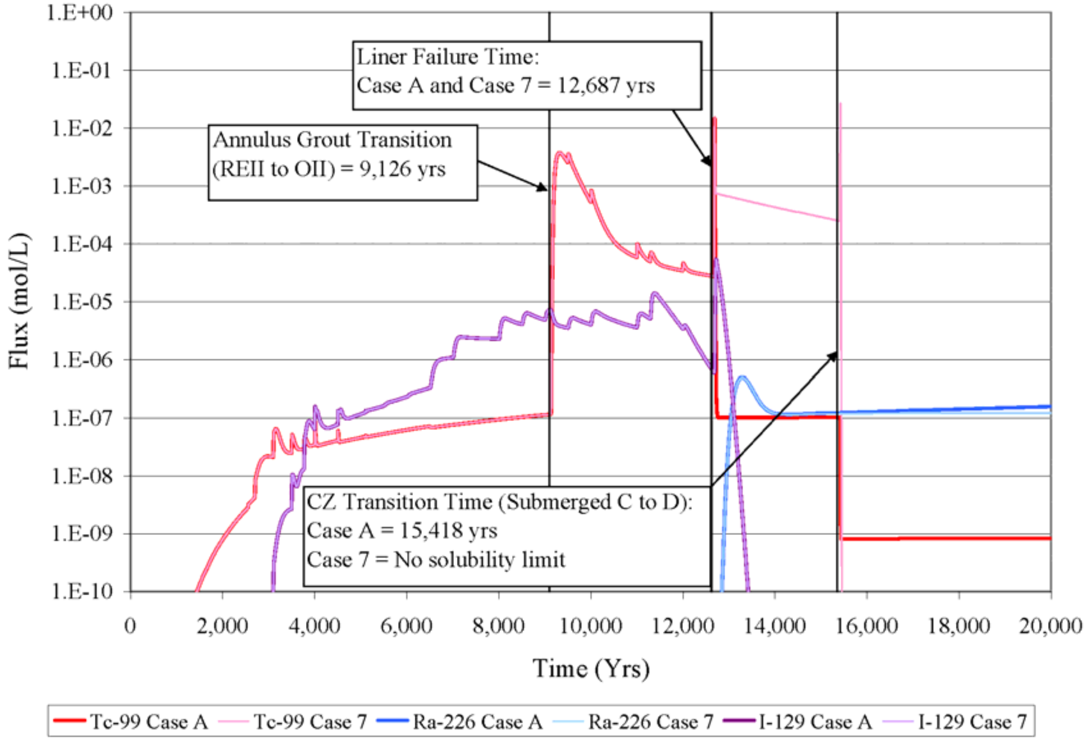


Figure 3.3-2: Fast-Moving Radionuclide Fluxes Tank 15 (CZ)

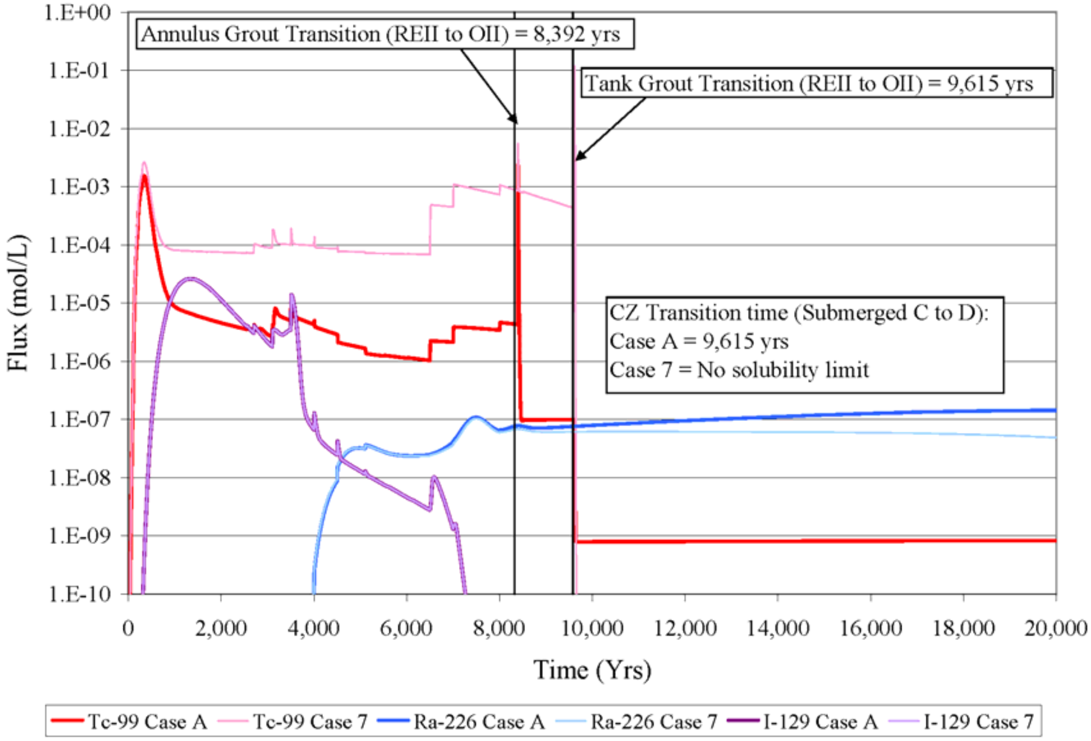


Figure 3.3-3: Fast-Moving Radionuclide Fluxes Tank 21 (CZ)

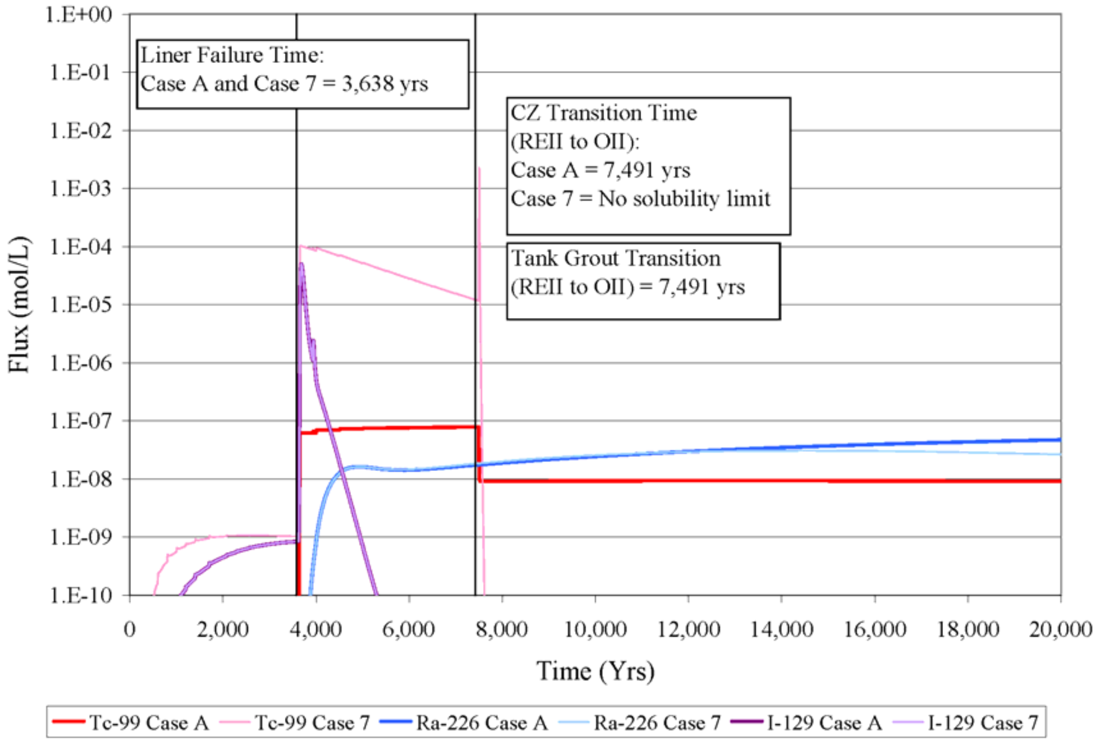
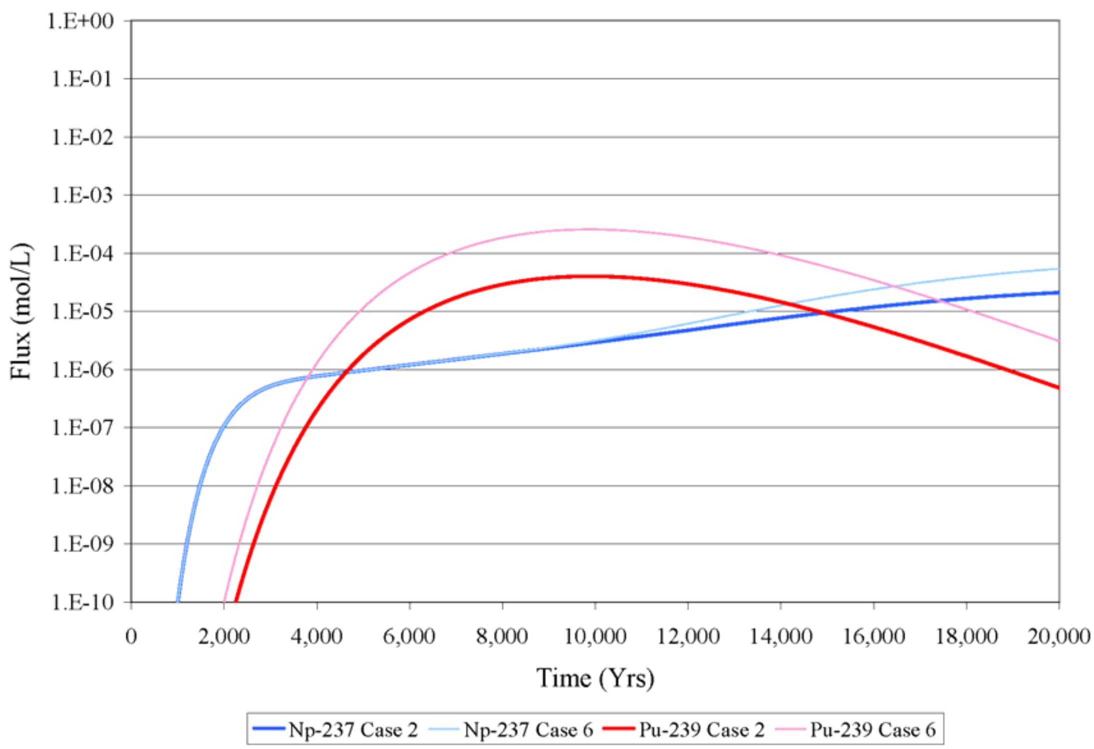


Figure 3.3-4: Slow-Moving Radionuclide Fluxes Tank 15 (CZ)



As expected, the radionuclides with solubility controls in the CZ are impacted in thebarrier cases (i.e., releases increase), and most notably Tc-99 and Pu-239 (and Np-237 later). As defined in the HTF PA, I-129 is modeled with instantaneous releases (i.e., no solubility limit); therefore I-129 is not impacted by this barrier case.

In Figure 3.3-1, Tc-99 releases from Case A and Case 7 are identical prior to liner failure (12,687 years) because releases originate in the sand pad and annulus, and are not subject to the solubility controls in the CZ. The gradual release of Tc-99 from the sand pad and annulus is depleted by the time the liner fails and CZ-sourced Tc-99 releases become dominant. At this time, the Tc-99 releases in Barrier Case 7 are more than three orders of magnitude greater than Case A, indicating that Tc-99 concentrations are controlled by the solubility constraints in the CZ in Tank 13 at this late time (Table 3.3-1).

Both Figure 3.3-2 and Figure 3.3-3 show that Tc-99 releases from Tank 15 (Type II tank with initially degraded liner) and Tank 21 are controlled at early times by the solubility limit. The Case A releases are suppressed by more than two orders of magnitude. Pu-239 releases from Tank 15 indicate an order of magnitude increase when solubility controls are removed (Figure 3.3-4).

Although Ra-226 is relatively insensitive to the chemical condition of the CZ, the release of Ra-226 is strongly dependent on the availability of the release of the radionuclides whose decay produces Ra-226, most notably Pu-238. Plutonium is solubility controlled; therefore, it is possible that given a large enough initial inventory of Pu-238, Ra-226 could be affected. This also explains the late time increase in Np-237 releases relative to the Case A (Figure 3.3-4). While Np-237 is relatively insensitive to the chemical condition of the CZ, the concentrations of Np-237's parent radionuclides (Cm-245→Pu-241→Am-241→Np-237) frequently reach their respective solubility limits.

After liner failure, changing the solubility limits applied within the CZ have the potential to significantly control the release of contaminants, most notably those radionuclides with concentrations that are well above the solubility limits, as illustrated by the Tc-99 and Pu-239 releases. Other barriers that influence the flow rate and chemical composition of pore fluids passing through the tank system contribute to changes in solubility limits.

3.4 Liner (Barrier Case A vs 5, and Case 2 vs 4)

The liner barrier analysis compared two barrier cases against the Case A and the fully degraded configuration (Barrier Case 2):

- Barrier Case 4: Nominal CZ, liner fails early based on grouted CO_2 diffusion coefficient of 1E-04 cm^2/s, all other barriers degraded
- Barrier Case 5: Nominal CZ, liner fails early based on grouted CO_2 diffusion coefficient of 1E-04 cm^2/s, all other barriers intact

For the Case A model, liner failure times vary with waste tank type due to differences in liner properties between the waste tank type (see Table 3.4-1). [SRR-CWDA-2010-00128] The Case A is compared to Barrier Case 5 to illustrate the impacts of early liner failure on the fast-moving radionuclides. Note, this analysis does not apply to waste tanks with initially failed liners (e.g., Tank 15), as the nominal conditions for these liners are the same as the failed conditions. The degraded configuration (Barrier Case 2) is compared to Barrier Case 4 in order to evaluate the impacts of liner failure on slow-moving radionuclides. Barrier Case 2 was modeled with no liner present starting at year zero and the CZ set to nominal conditions (see HTF PA Tables 4.2-5 and 4.2-6 for solubility tables). [SRR-CWDA-2010-00128]

Table 3.4-1: Carbon Steel Liner Life Estimates by Waste Tank Type

Waste Tank Type	**Liner Failure Year for Modeling Based on Grouted Waste Tank Liner Condition**	
Liner Condition	CO_2 Diffusion Coefficient = 1.0E-06 cm^2/s (Nominal)	CO_2 Diffusion Coefficient = 1.0E-04 cm^2/s (Partially Failed)
Type I	11,397	1,142
Type II	12,687	2,506
Type III/IIIA	12,751	2,077
Type IV	3,638	75

[SRR-CWDA-2010-00128 (Table 4.2-36)]

For Type I, Type III, and Type IIIA waste tanks, releases from the waste tanks initiate following liner failure. For the Type II waste tanks, inventory in the sand pads and annulus allow for early releases. Similarly, although flow through the liners is relatively limited for all waste tank types, the thinner basemat of the Type IV tanks, combined with no secondary liner results in relatively greater flow through the Type IV tanks. Because peak dose to the Member of the Public (MOP) is controlled primarily by Type II and IV waste tanks (see HTF PA Section 5.5), this barrier analysis focused on evaluating the liner failure time impacts to these waste tank types.

Table 3.4-2 presents a comparison of the transitions times by waste tank for Barrier Cases 4 and 5. This table provides a summary of the relevant information depicted in Section 2.

Table 3.4-2: Select Transition Times by Waste Tank – Barrier Cases 4 and 5

	Year of Occurrence	
Type of Transition	**Case 4**	**Case 5**
Type II Tank - Tank 13 (Figures 2.0-1 through 2.0-6)		
Liner fails hydraulically	2,506	2,506
Annulus grout (Submerged C to Submerged D)	216	8,542
Tank Grout and CZ (Submerged C to Submerged D)	4,396	9,709
Annulus grout (Submerged D to Oxidized Region III)	1,244	18,051
Tank Grout and CZ(Submerged D to Oxidized Region III)	13,366	>20,000
Basemat (Oxidized Region II to Oxidized Region III)	78	3,574
Type IV Tank - Tank 21 (Figures 2.0-13 through 2.0-17)		
Liner fails hydraulically	75	75
Tank Grout and CZ (Reducing Region II to Oxidized Region II)	2,954	6,744
Tank Grout and CZ (Oxidized Region II to Oxidized Region III)	16,971	>20,000
Basemat (Oxidized Region II to Oxidized Region III)	240	2,524

Note: Information extracted from HTF PA PORFLOW results. [SRR-CWDA-2010-00128]
N/A = Not Applicable

Figures 3.4-1 and 3.4-2 present the impacts of early liner failure (Barrier Case 5) on the transport of fast-moving radionuclides, Tc-99, I-129, and Ra-226, in Tank 13 (Type II tank, intact liner) and Tank 21 (Type IV tank). In Figure 3.4-1, Case A releases of Tc-99 prior to liner failure (12,687 years) are primarily from the sand pad and annulus inventory. The first Tc-99 peak at 9,380 occurs due to the annulus chemical transition from Submerged C (Reducing Region II), where Tc-99 is bound in the annulus due to a high K_d (5,000 mL/g), to Submerged D (Oxidizing Region II), where Tc-99 is released because Tc-99 has a lower K_d (0.8 mL/g) in this chemical environment (Table 3.1-1). The second Tc-99 peak (12,710 years) occurs immediately after liner failure, which follows the annulus transition from Submerged D (Oxidizing Region II) to Oxidizing Region III, where Tc-99 has an even lower K_d (0.5 mL/g) (Table 3.1-1). This transition results in additional Tc-99 being flushed from the annulus (and any Tc-99 in the waste tank grout from upward diffusion from the CZ). For the remainder of the simulation Tc-99 releases are limited by the solubility controls applied to the CZ.

Failing the Tank 13 liner more than 10,000 years earlier in Barrier Case 5 causes the peak Tc-99 flux to occur more than 4,000 years earlier (Figure 3.4-1). For Barrier Case 5, the cause of the Tc-99 peak differs from the Case A, in that the Barrier Case 5 peak is from the annulus transition from Submerged C (Reducing Region II) to Submerged D (Oxidizing Region II) (at 8,542 years), while the Case A peak is from liner failure (Table 3.4-2). The magnitude of the peak flux in Barrier Case 5 is slightly higher. Similarly, the I-129 peak flux is higher in magnitude (less than one order of magnitude) and occurs more than 9,000 years earlier in Barrier Case 5. In contrast, the peak flux from Ra-226 for Case A is higher in magnitude, relative to Barrier Case 5, despite occurring nearly 6,000 years later, due to the contributions from in-growth as parent radionuclides decay.

Figure 3.4-2 shows the fast-moving radionuclide releases from Tank 21. Liner failure in Case A occurs at 3,683 years, and at 75 years in Barrier Case 5. Tc-99 releases begin immediately after liner failure in each case. There is little difference in the peak or trend of Tc-99 releases because both release curves are solely controlled by the solubility limits in the CZ, and unlike Type II tanks, there is no other source of Tc-99 inventory in the Type IV tanks. In contrast, peak fluxes for I-129 in Barrier Case 5 occurs earlier following liner failure by more than 3,000 years, and the peak is nearly an order of magnitude lower. The flux for Ra-226 begins to appear earlier in Barrier Case 5, but follows the same trend as Case A out to 20,000 years.

These figures illustrate the importance of liner failure on the release of fast and slow moving radionuclides. As seen previously, early liner failure also contributes to changing solubilities and early release of radionuclides from the CZ.

Figure 3.4-1: Fast-Moving Radionuclide Fluxes Tank 13 (Liner Failure)

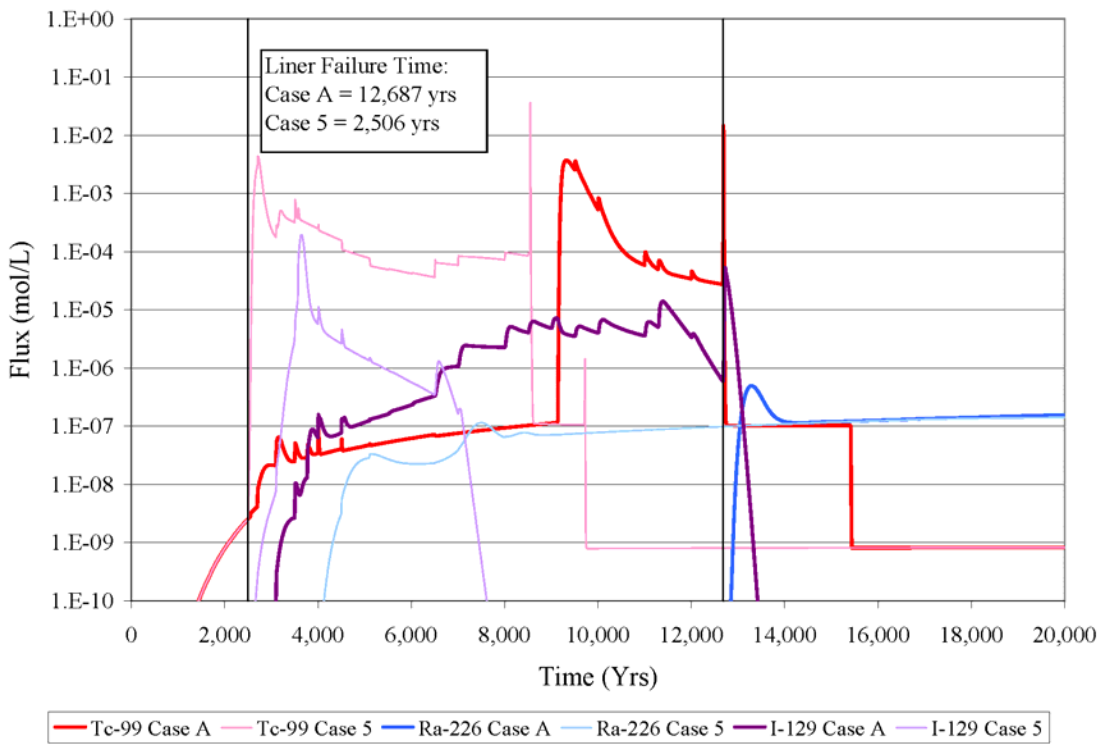
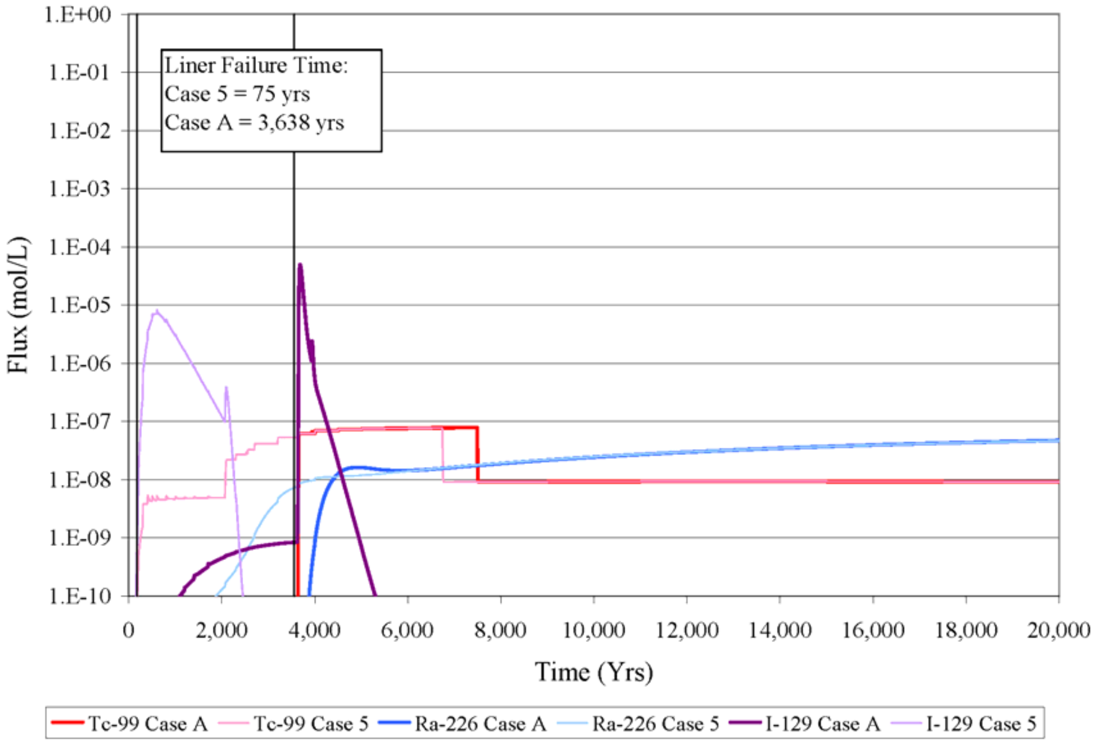
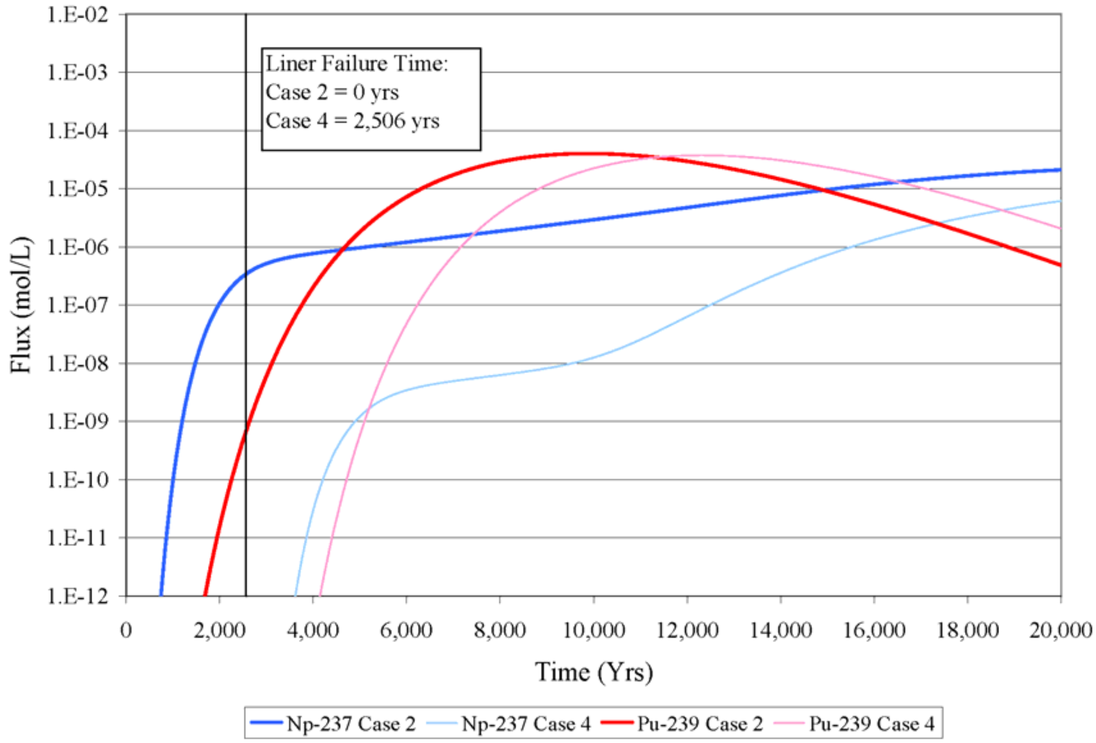


Figure 3.4-2: Fast-Moving Radionuclide Fluxes Tank 21 (Liner Failure)



The effect of early liner degradation on slow-moving radionuclides (Pu-239 and Np-237) in Tank 13 is illustrated in Figures 3.4-3. The liner is failed at the onset of the simulation period in the degraded case (Barrier Case 2), and is failed at 2,506 years in Barrier Case 4. The magnitude of the peak Pu-239 flux is nearly the same in both runs, while the Np-237 peak flux is approximately one order magnitude less in Barrier Case 4 at the end of the simulation period. Differences in the fluxes at earlier times are more pronounced. The timing of liner failure delays the releases of both Pu-239 and Np-237 in Barrier Case 4. Note that early liner degradation has very little effect on the fluxes of the slow-moving radionuclides in Tank 21.

Figure 3.4-3: Slow-Moving Radionuclide Fluxes Tank 13 (Liner Failure)



3.5 Vadose Zone (Barrier Case A vs 8, and. Case 2 vs 9)

The barrier capability of the vadose zone is estimated by comparing the timing and magnitude of peak fluxes and the time history curves for a vadose zone under degraded conditions compared to one with nominal conditions. The vadose zone is a natural barrier of material immediately below the basemat. [SRR-CWDA-2010-00128] Submerged waste tanks are not included in this analysis because the native soil below the waste tank basemat is saturated. Therefore, only non-submerged, unsaturated waste tanks represented by Tank 21 and Tank 36 are presented in this analysis. The nominal condition for the native soil in the unsaturated waste tanks initially uses cement leachate impacted soil K_ds. The "failed" vadose zone assumes lower K_d values for each element. Using lower K_d values maximizes radionuclide migration through the vadose zone. The K_d values used for nominal and failed vadose zone conditions are presented in Table 1.1-5 for the radionuclides of interest: Tc-99, I-129, Ra-226, Pu-239, and Np-237. Note that the failed, cement leachate impacted sandy soil K_d for I-129 was simulated using the value 0.01 mL/g, instead of 0 mL/g. However, as the following section indicates, varying the I-129 soil K_d between 0.01 and 0 mL/g has little impact on the results.

Barrier Cases 2 and 8 apply failed conditions for the vadose zone (Table 1.1-4). The Case A compares to Barrier Case 8 as these cases use the nominal settings for the other barriers; whereas Barrier Case 2 compares to Barrier Case 9 as these cases use the degraded settings for the other barriers.

Figures 3.5-1 and 3.5-2 present the impact of failing the vadose zone on the fast-moving radionuclide releases in Tank 21 and 36, respectively. Figures 3.5-3 and 3.5-4 present the impact of failing the vadose zone on the slow-moving radionuclides in Tank 21 and 36, respectively.

The vadose zone has no appreciable impact on radionuclides with little to no affinity for sorbing to soil (e.g., Tc-99, I-129, and Np-237). For those radionuclides that do sorb, the vadose zone provides a moderate (e.g., Ra-226) to significant (e.g., Pu-239) impact, commensurate with their relative sorptive capacity of the radionuclide and the thickness of the native soil zone below the waste tank (more impact is apparent in Tank 36 due to a thicker vadose zone). Under nominal conditions, sorption in the vadose zone is increased for these radionuclides. Consequently, because less sorption is occurring in the failed case, radionuclides influenced by sorption onto soil have higher early releases. However, over time the failed cases become depleted, allowing the nominal case, which has more available mass, to overtake the failed case (e.g., Pu-239). Figures 3.5-1 and 3.5-2 indicate that the vadose zone delays initial Ra-226 releases between 500 and 1,000 years, whereas peak releases of Pu-239 are delayed by as much as 3,000 years (see Figure 3.5-3 and 3.5-4). The vadose zone dampens the peak Ra-226 fluxes by less than one order of magnitude, while the Pu-239 peak flux is not significantly different.

Figure 3.5-1: Fast-Moving Radionuclide Fluxes Tank 21 (Vadose Zone)

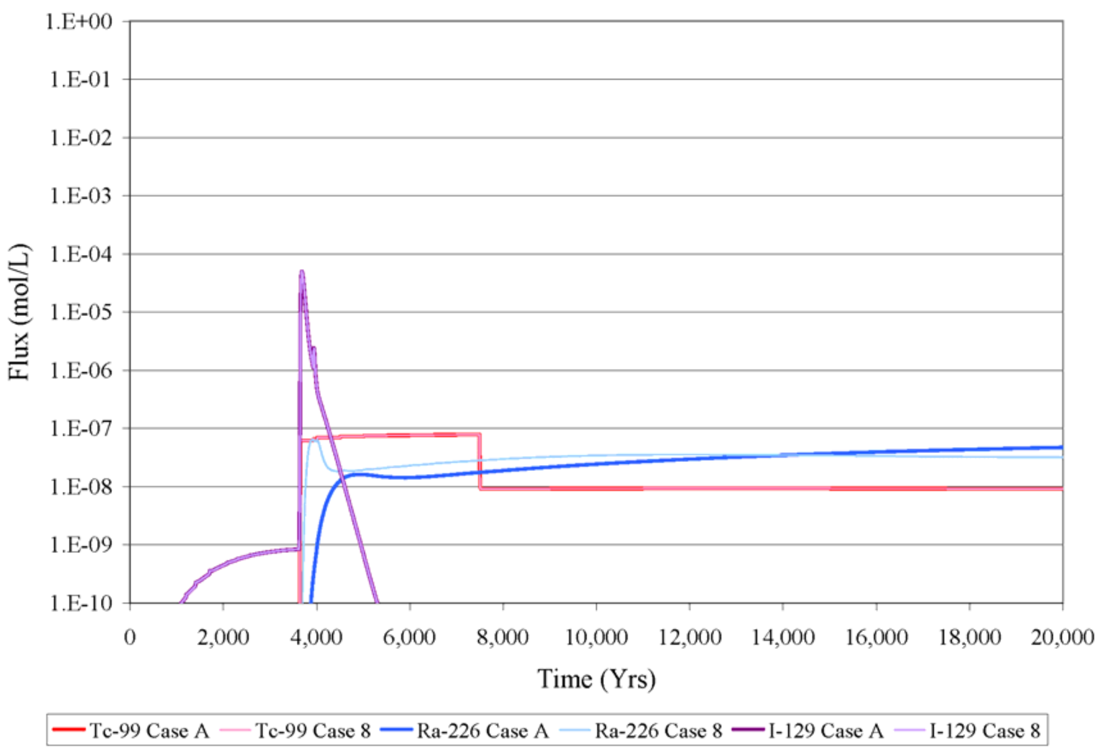


Figure 3.5-2: Fast-Moving Radionuclide Fluxes Tank 36 (Vadose Zone)

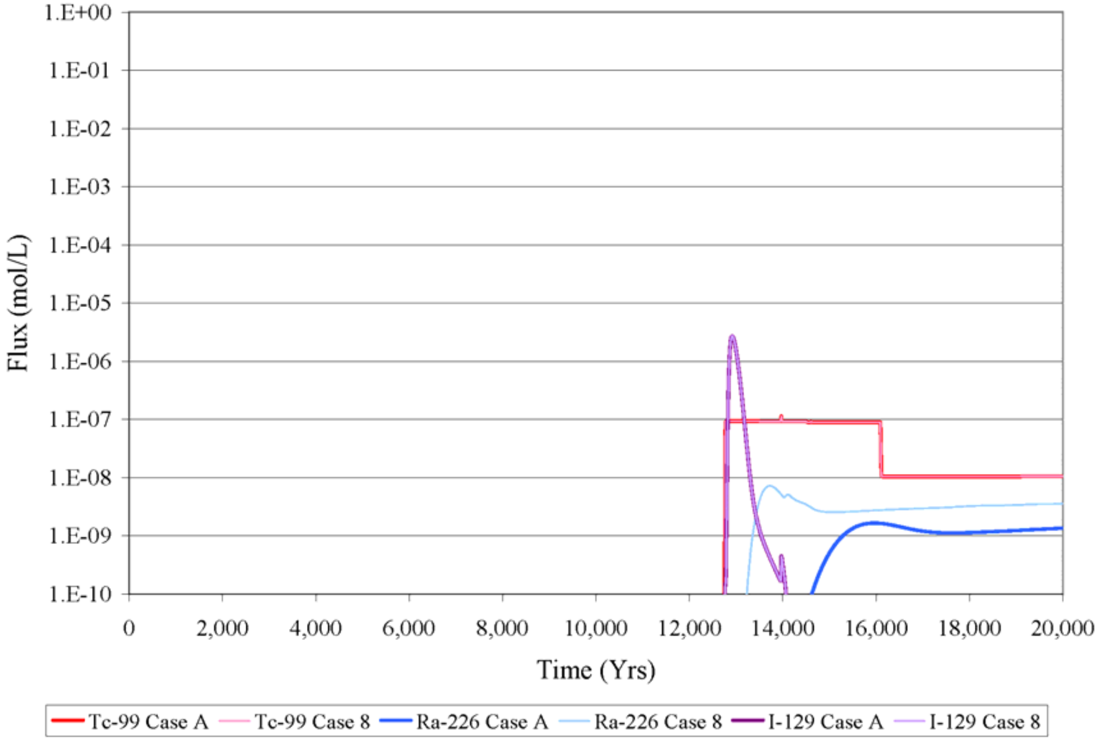


Figure 3.5-3: Slow-Moving Radionuclide Fluxes Tank 21 (Vadose Zone)

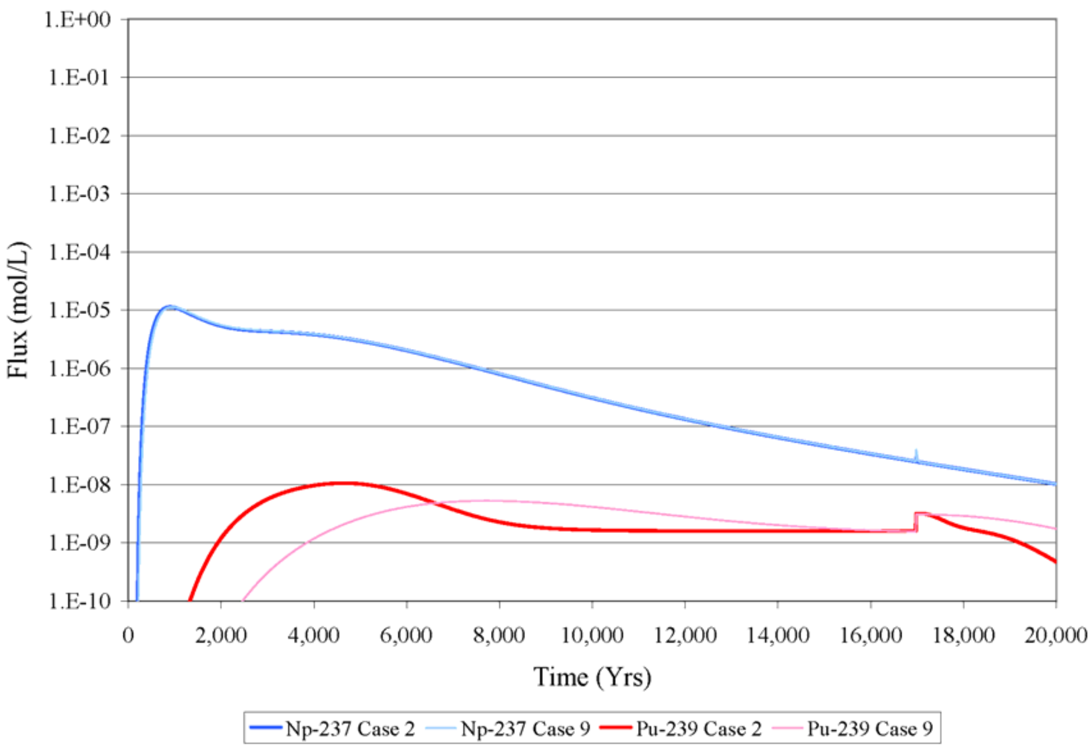
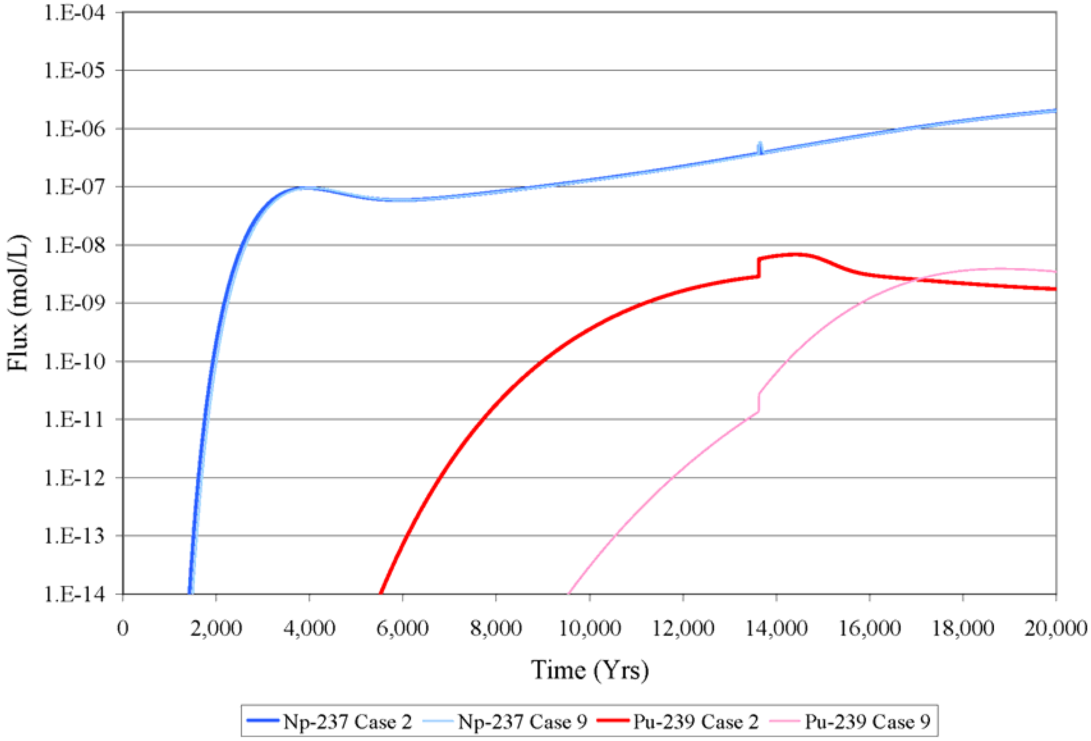


Figure 3.5-4: Slow-Moving Radionuclide Fluxes Tank 36 (Vadose Zone)



4.0 CONCLUSIONS

The closure cap and the vadose zone have less of an impact on radionuclide fluxes, relative to the liners, the CZ, and the waste tank grout. The importance of each barrier on radionuclide transport is element specific for the CZ, the waste tank grout and the vadose zone, whereas the liner and closure cap are inclined to have a similar affect for all radionuclides (Table 4.0-1). Liner failure has the largest impact on the timing of peak flux for the different radionuclides. The earlier a waste tank liner fails, the earlier the peak release for that radionuclide. Depending on the time of early failure, the peak flux can occur earlier by thousands of years. The change in the magnitude of the peaks varies by waste tank type and radionuclide. The liner is an effective barrier to radionuclide migration because it is designed to prevent flow and mass transport out of the waste tanks. Failure of the liner allows mass built up behind the liner to be rapidly flushed from the bottom of the waste tanks. Secondary effects of liner failure include accelerated hydraulic degradation of the grout, which influences the timing of solubility changes in the CZ and K_d transitions in the cementitious materials and vadose zone. In this way, the timing of liner failure strongly controls peak flux.

The CZ, which mostly impacts peak Tc-99 and Pu-239 releases (and Np-237 in later years), acts to delay and decrease the Tc-99 peak fluxes by several orders of magnitude, however it has little to no impact on the transport of I-129 and Ra-226. The CZ also dampens the Pu-239 peak flux by approximately one order of magnitude; however, it does not affect the timing of the flux peak. The CZ effectively dampens the flux of Tc-99 and Pu-239 out of the waste tanks because (1) these radionuclides are strongly controlled by solubility, and (2) their aqueous concentration in the CZ remains at or close to the solubility limit. If their aqueous concentrations were less, the CZ would be less effective at limiting the release of these radionuclides.

The integrity of the waste tank grout plays an important role in delaying the releases of I-129, Ra-226, Np-237, and Pu-239; although the peak magnitude is not significantly different. The integrity of the waste tank grout indirectly affects the Tc-99 releases, in that degraded waste tank grout has the ability to impart its reducing capacity onto the CZ, which causes the CZ chemical transitions to occur later. The impact of the grout on Type II tanks (with intact or initially degraded liner) is more difficult to discern because the radionuclide releases are overprinted by the inventory coming from the sand pads and annulus. More specifically, the large fluctuations in the hydraulic conductivity through the grout can greatly change the flow fields through the waste tank system, including redirecting flow through the annulus, which acts as a sink/source of inventory prior to liner failures.

Although an independent barrier analysis of the annulus grout was not done, it is apparent from the interpretation of the time histories presented that the timing of annulus grout transition (Type II tanks) greatly influences the timing of Tc-99 peaks. The annulus transition triggers a large decrease in Tc-99 sorption onto the annulus grout (from a K_d value of 5,000 mL/g to 0.8 mL/g). This transition combined with a significant inventory in the annulus (some initiated in the sand pads) produces significant releases prior to liner failure.

The closure cap plays an important role in that it limits flow into and through the tanks; however the impact of the closure cap is negated by the presence of an intact steel liner. The effectiveness of this barrier can, potentially, be extended by delaying the placement of the closure cap until the degradation of the cementitious materials has significantly advanced, thereby reducing infiltration when the system is more vulnerable to the release of contaminants. The impact of the faster flow in the first few thousand years from removal of this barrier results in greater Np-237 and Pu-239 releases by as much as two orders of magnitude. The vadose zone dampens radionuclide releases especially for those radionuclides with higher soil K_d values (e.g., plutonium and radium, as well as the parents of radium and neptunium); however, this barrier plays a lesser role in controlling peak releases.

In summary, the timing of liner failure is the most important barrier to the timing of the peak fluxes for all radionuclides. The integrity of the CZ also has a significant impact on the timing and magnitudes of Tc-99 and Pu-239 fluxes (and Np-237 in later years) due to relatively high concentrations and low solubility limit controls. The tank grout integrity effects the timing (although not necessarily the magnitude) of the peak fluxes releases of I-129, Ra-226, Np-237, and Pu-239, but not Tc-99. The closure cap is only important during the first few thousand years, but is less significant as the barrier degrades. Radionuclides with higher K_d values (such as Pu-239) are dampened (i.e., release is slowed) by the vadose zone. In general, the later the peak flux, the lower the magnitude of the peak flux, regardless of the dominant controlling barrier or the released contaminant.

Table 4.0-1: Importance of Barrier

Barrier	N (Nominal)	P (Partially Degraded)	F (Fully Degraded)
Closure Cap	Infiltration profile per Case A (HTF PA Table 4.2-23). Important during the first few thousand years. Small delay and peak reduction in radionuclide releases to the saturated zone. Prevents earlier transition times from contributing to releases from the annulus, sand pads, waste tank grout, and CZ.	N/A	Infiltration constant at 16.45 inches per year. [WSRC-STI-2007-00184] Less sorptive fast-moving radionuclides show a moderate impact. Pu-239 and Np-237 show about an order of magnitude increase in flux when closure cap absent, primarily due to earlier releases.
Tank Grout and Cementitious Materials (K_d controlled)	Hydraulic properties (e.g., failure date) and chemical properties unchanged per Case A. Plays an important role in delaying changes in transition times by maintaining reducing capacity. Intact liner and absence of fast flow paths contribute to effectiveness.	N/A	High flow throughout grout causes grout to impart reducing capacity onto CZ. Chemical transitions are a function of the "failed" flow fields. Degradation causes large fluctuations in the hydraulic conductivity through the grout, redirecting flow through the waste tank system affecting I-129, Ra-226, Np-237, and Pu-239 releases.
CZ (Solubility controlled)	Initial solubility limits in the CZ associated with Case A. Solubility controls in the CZ are effective in limiting the peak release timing and magnitude of solubility dependent radionuclides.	N/A	Solubility controls are removed for Tc-99 and Pu-239, and set to facilitate faster releases for the remaining radionuclides Failure of liner and grout degradation contributes to changing transition times which contributes to releases from the CZ. Tc-99 and Pu-239 concentrations are controlled by the solubility constraints.
Waste Tank Liner [b]	Liner prevents release of radionuclides and accelerated hydraulic degradation of the grout. Timing of liner failure influences the timing of solubility changes in the CZ and K_d transitions in the cementitious materials and vadose zone	Early liner failure (based on grouted CO_2 diffusion coefficient of 1E-04 cm^2/s). Delayed impact to fully degraded condition.	No liner at time = 0 yrs. Initial liner failure causes peak flux for Tc-99 and I-129 to occur earlier and be higher in magnitude. Less effect on slow moving and solubility dependent radionuclides.
Vadose Zone (K_d controlled)	Native soil K_d values are set equal to Case A values. Dampens radionuclide releases for those radionuclides that sorb to soil; however, this barrier plays a lesser role in controlling peak releases.	N/A	Native soil K_d values are as defined in Table 1.1-5. No appreciable impact on radionuclides with little to no affinity for sorbing to soil (Tc-99, I-129, and Np-237) Affects the release of radionuclides with higher soil K_d; values (Ra-226 and Pu-239).

5.0 REFERENCES

SRR-CWDA-2010-00128, *Performance Assessment for the H-Area Tank Farm at the Savannah River Site*, Rev. 0, Savannah River Site, Aiken, SC, March 14, 2011.

WSRC-STI-2007-00184, Phifer, M. A., *FTF Closure Cap Concept and Infiltration Estimates,* Savannah River Site, Aiken, SC, Rev. 2, October 15, 2007.