Savannah River Remediation
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INTEROFFICE MEMORANDUM

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PEAK DOSE SENSITIVITY TO CHANGES IN CHEMISTRY-DEPENDENT SOLUBILITY LIMITS FOR PLUTONIUM, TECHNETIUM, AND URANIUM

SUMMARY

The H-Tank Farm (HTF) and F-Tank Farm (FTF) GoldSim Models were used in deterministic mode to perform a sensitivity analysis designed to evaluate the degree of influence that changes in the chemical environment-dependent solubility limits would have on the peak doses resulting from releases from each Tank Farm, based on performance assessment (PA) modeling. The purpose of this memorandum is to present the results of this sensitivity analysis.

For FTF, Version 3.0 of the FTF GoldSim Model (used for the *Tanks 5 and 6 Special Analysis for the Performance Assessment for the F-Tank Farm at the Savannah River Site* [SRR-CWDA-2012-00106]) was modified. Similarly, for HTF, Version 4.0 of the HTF GoldSim Model (updated as discussed in the *Updates to the H-Area Tank Farm Stochastic Fate and Transport Model* [SRR-CWDA-2014-00060 Rev. 2]) was modified. Specifically, for the FTF and HTF analyses, the solubility limit values for key elements (Pu, Tc, and U) were varied to assess the relative importance the individual radionuclide's solubility has on radionuclide releases from tanks found in the FTF and HTF, and the calculated Member of the Public (MOP) peak-dose values were used to evaluate whether performance objectives have been met for the two tank farms. The simulations were completed out to 100,000 years and the parameter sensitivities were examined over 20,000-year and 100,000-year time spans.

The parameter sensitivity simulations were performed using the Base Case (Case A) parameters with all variables held constant except for plutonium, technetium, and uranium solubility values. In this study, plutonium, technetium, and uranium solubility values were varied, one element at a time. For each of the elements (plutonium, technetium, and uranium), results for a "conservative case" and "nominal case" were derived and compared to the deterministic Base Case dose results, which use the FTF PA solubility values for the FTF GoldSim Model and the HTF PA/Waste Release Model (WRM) solubility values for the HTF GoldSim Model.

Because the HTF PA solubilities represent updated values relative to the FTF PA values, the deterministic FTF PA Model was also run with all variables held constant except for plutonium, technetium, and uranium solubility values which were updated to use the HTF PA values. These results were also compared to the "conservative case" and the "nominal case" dose results.

Examination of the resulting MOP doses from the FTF Model and respective comparisons against the doses resulting from the "conservative case" and the "nominal case" solubility sensitivities, showed that for FTF the MOP dose curves, peak-dose magnitude, and peak-dose timing were very sensitive to the plutonium solubility data and insensitive to the technetium and uranium solubility data. Similarly, the results from the HTF Model showed that the MOP dose curves and associated peak-dose values were only slightly sensitive to the plutonium solubility data and insensitive to the technetium and uranium solubility data.

DISCUSSION

Plutonium Solubility Variability

To study the potential impact that plutonium solubility has on MOP groundwater pathway doses, the FTF and HTF GoldSim Models were run using their respective Base Case (Case A) parameters with all variables held constant except for the Pu solubility values associated with Reduced Region II, Oxidized Region II, and Oxidized Region III cementitious materials. Dose results from using four different sets of Pu solubility values for the FTF simulations and three of the four for the HTF simulations (see Table 1) were compared. The values denoted as FTF PA are the Pu solubilities used in the FTF PA (SRR-CWDA-2012-00106). The values denoted as HTF PA/WRM are the Pu solubilities determined in the current HTF Waste Release Model (WRM) (SRNL-STI-2012-00404) and used in the HTF PA (SRR-CWDA-2010-00128). Note that the FTF GoldSim Model simulations include an extra (fourth) simulation based on the HTF PA/WRM Pu, Tc, and U solubilities, which represent information from the most recent WRM, which was issued after the FTF PA. The two other sets of values are a "conservative case" and "nominal case" that allow for examination of the effect increased plutonium solubility would have on FTF and HTF dose results. The "conservative case" and "nominal case" are based upon a considered evaluation of preliminary waste release test data.

Table 1: Plutonium Solubility

	Reduced Region II	Oxidized Region II	Oxidized Region III
Solubility Data	mol/L	mol/L	mol/L
FTF PA[1]	4.1E-12	4.0E-14	5.7E-05
HTF PA/WRM[2]	3E-11	3E-11	3E-11
Conservative Case[2]	5E-09	1E-08	2E-08
Nominal Case[2]	1E-09	1E-09	1E-08

[1] Used only in the FTF simulations
[2] Used for both FTF and HTF simulations

FTF Solubility Sensitivity

As seen in Figure 1, for the FTF sensitivity analysis, although the FTF PA and HTF PA/WRM solubility value sets are different (see Table 1), the doses over 20,000 years show negligible differences, indicating that the dose contributions from plutonium radionuclides are masked by the releases of other more dominant radionuclides over this time span. Figure 1 also shows that for the "nominal case" and "conservative case" the greater plutonium solubility values (see Table 1) have an impact on the FTF dose, and plutonium (mainly in the form of Pu-239) is a clear contributor to dose. As seen in Figure 2, over a period of 100,000 years, the constant but low Pu solubilities (3.2E-11 mol/L) for the HTF PA/WRM

data (see Table 1), limit the release of plutonium radionuclides to a level where their dose contribution is obfuscated by the contributions of other species (mainly Nb-93m). When comparing the FTF PA data results with the "nominal case" and "conservative case" results, it can be seen that the Oxidized Region III solubility for the FTF PA data simulation generates the maximum peak dose over the 100,000 year period. Relative to the FTF PA data based results, the much higher Reduced Region II and Oxidized Region II solubilities associated with the "conservative case" data (see Table 1), generates an earlier and broader peak of less magnitude for the "conservative case" results.

HTF Solubility Sensitivity

As seen in Figure 3, in the HTF sensitivity analysis, even assuming greater Pu solubility values (i.e., using the Pu solubility values from Table 1) for the "nominal" and "conservative" cases, there is no significant impact on the HTF peak dose within 20,000 years. Over the course of 100,000 years, for the "conservative" and the "nominal" cases there is a discernible but small increase in the peak doses (Figure 4) of approximately 8% and 2%, respectively, at around 75,000 years. Over the 100,000 years, Ra-226 is the dominant dose contributor and the increased Pu-239 only slightly increases the dose.

Figure 1: FTF MOP Doses for Various Pu Solubility Values – 20K Years

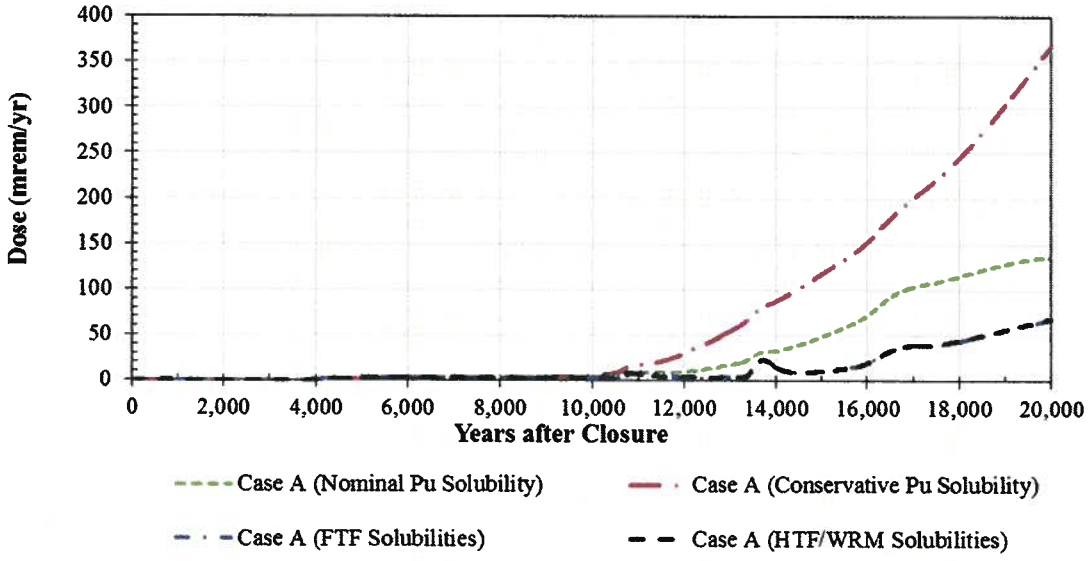


Figure 2: FTF MOP Doses for Various Pu Solubility Values – 100K Years

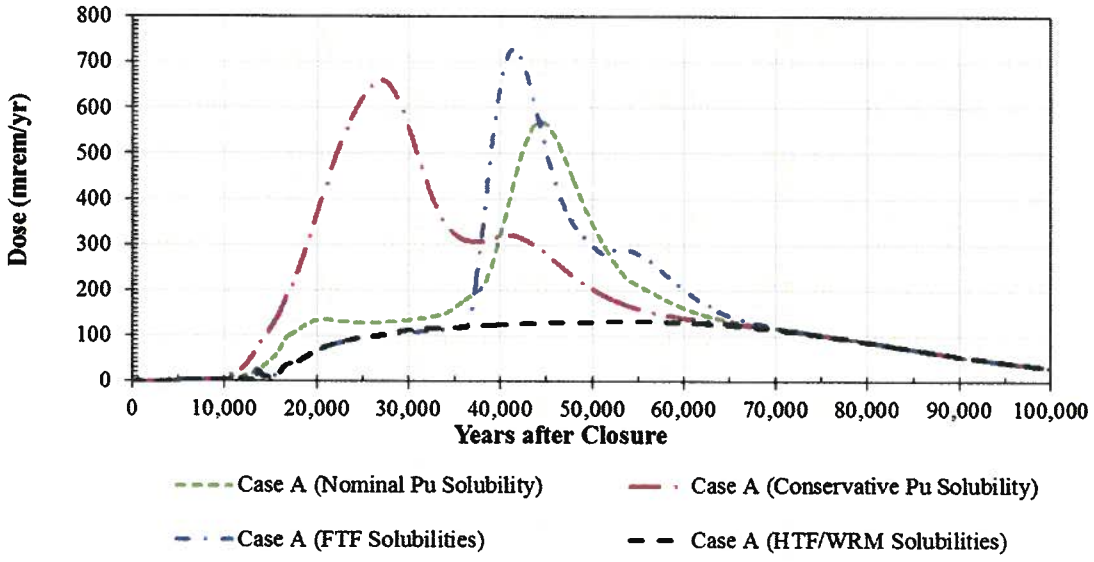


Figure 3: HTF MOP Doses for Various Pu Solubility Values – 20K Years

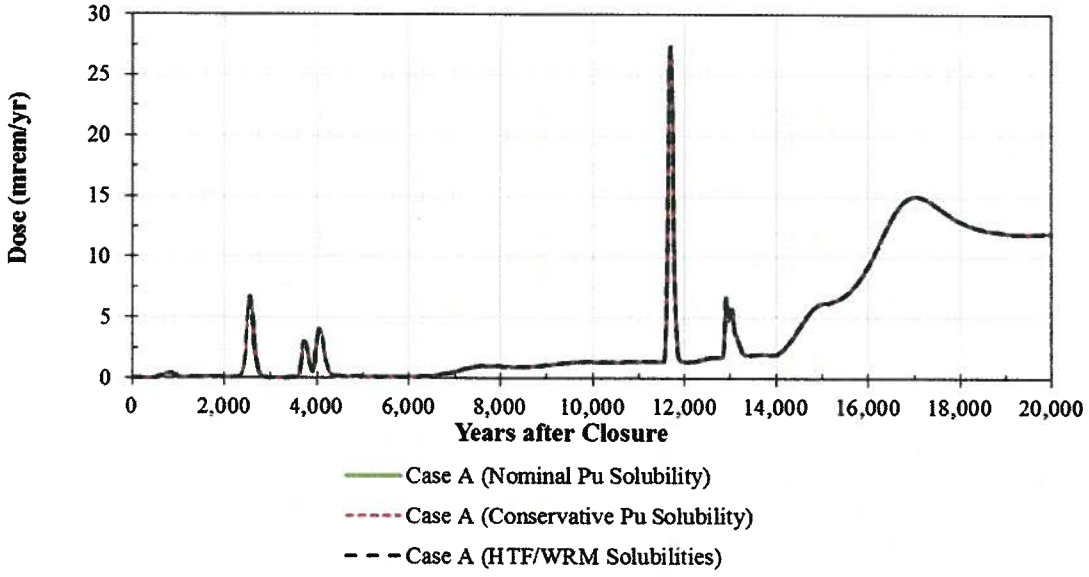
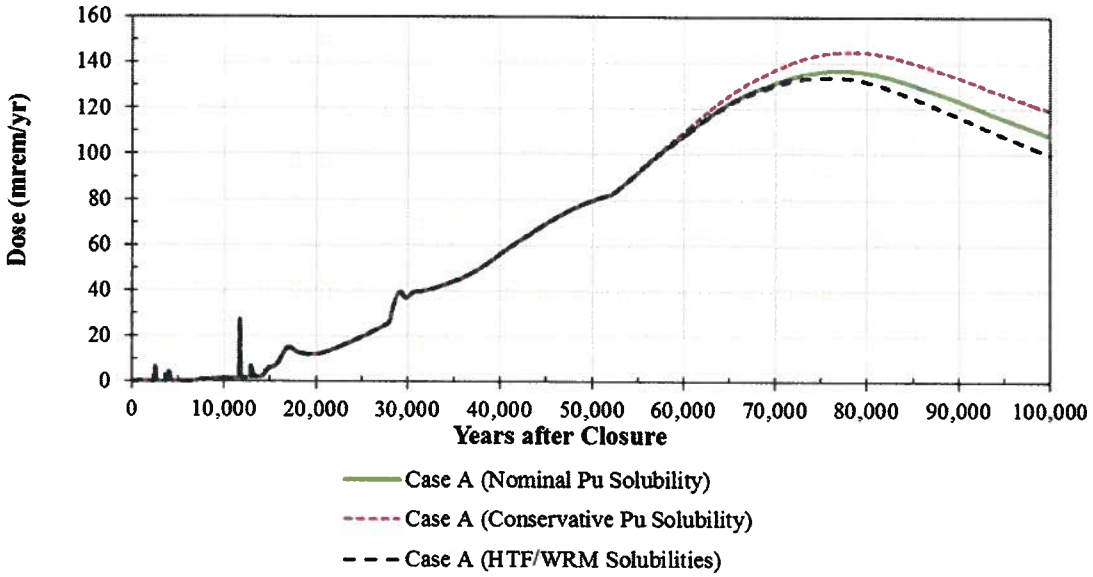


Figure 4: HTF MOP Doses for Various Pu Solubility Values – 100K Years



Technetium Solubility Variability

To study the potential impact that technetium solubility has on MOP groundwater pathway doses, the FTF and HTF GoldSim models were run using their respective Base Case (Case A) parameters with all variables held constant except for the Tc solubility values associated with Reduced Region II, Oxidized Region II, and Oxidized Region III cementitious materials. Dose results from using four different sets of Tc solubility values for the FTF simulations and three for the HTF simulations (see Table 2) were compared. The first set of values listed are the Tc solubilities used in the FTF PA (SRR-CWDA-2012-00106). The second set of values are the Tc solubilities determined in the current HTF Waste Release Model (WRM) (SRNL-STI-2012-00404) and used in the HTF PA (SRR-CWDA-2010-00128). The two other sets of values are a "conservative case" and "nominal case" that allow for examination of the effect increased Tc solubility would have on FTF and HTF dose results. The "conservative case" and "nominal case" are based upon a considered evaluation of preliminary waste release test data.

Table 2: Technetium Solubility

	Reduced Region II	Oxidized Region II	Oxidized Region III
Solubility Data	mol/L	mol/L	mol/L
FTF PA[1]	3.1E-11	3.00E-13	instantaneous
HTF PA/WRM[2]	1E-14	1E-13	2E-15
Conservative Case[2]	3E-09	2E-08	2E-08
Nominal Case[2]	6E-10	1E-08	1E-08

[1] Used only in the FTF simulations
[2] Used for both FTF and HTF simulations

FTF Solubility Sensitivity

As seen in Figure 5, although the FTF PA, HTF PA, "conservative case" and "nominal case" solubility value sets are quite different (see Table 2), the total dose breakthrough curves over 20,000 years show negligible differences, indicating that the peak-dose contributions from Tc-99 are masked by releases of other radionuclides (such as C-14, Pa-231, Np-237, Ra-226, and Nb-93m) which dominate the dose results over this time span. Over a period of 100,000 years, the dose contributions from radionuclides such as Nb-93m and Pu-239, dominate the total dose calculations for the FTF PA, the "conservative case" and the "nominal case" data (see Table 2); thereby obfuscating any changes in the dose results associated with different Tc-99 dose contributions (Figure 6). For the HTF PA/WRM data, which consists of constant but low plutonium solubilities (3.2E-11 mol/L), as shown in Table 1, the release of plutonium radionuclides is limited (see Figure 2) and for this set of data, Nb-93m is the dominant dose contributor over the 100,000 year time span.

HTF Solubility Sensitivity

As seen in Figure 7 and Figure 8, even assuming greater Tc solubility values (i.e., using the Tc solubility values from Table 2) there is no significant impact on the HTF peak doses within either the 20,000 year time span or the 100,000 year time span.

Figure 5: FTF Peak MOP Doses for Various Tc Solubility Values – 20K Years

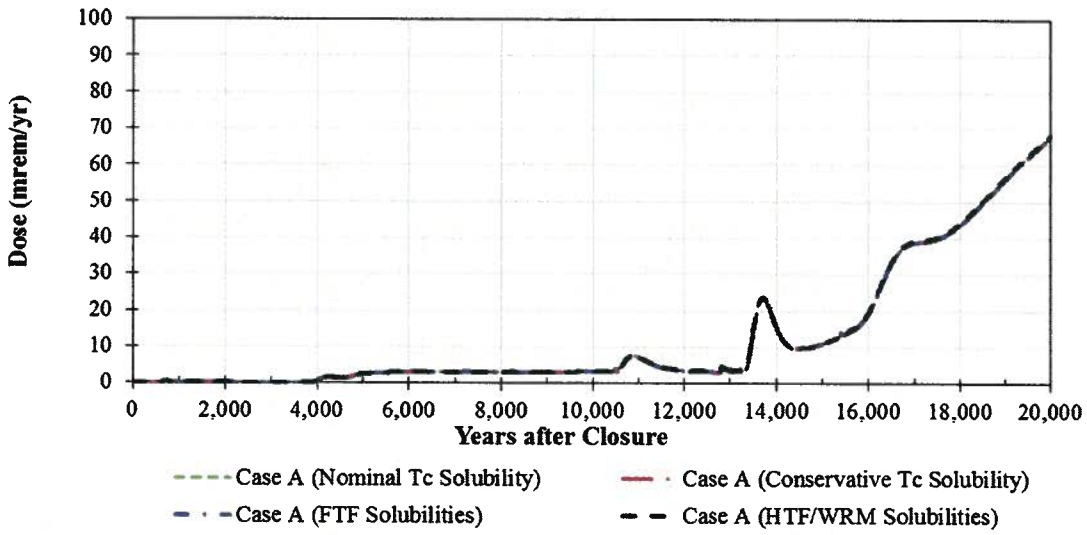


Figure 6: FTF Peak MOP Doses for Various Tc Solubility Values – 100K Years

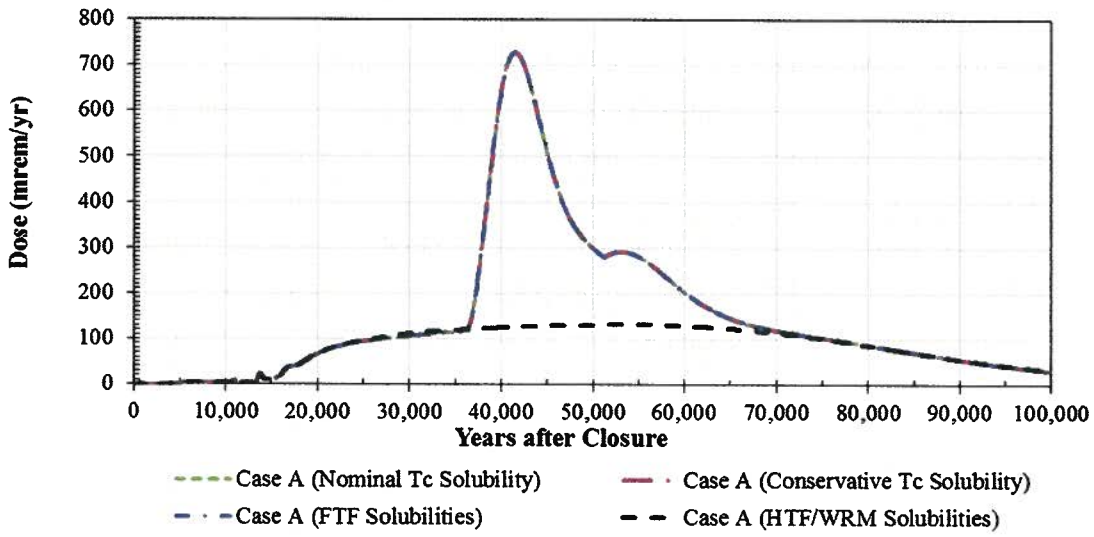


Figure 7: HTF Peak MOP Doses for Various Tc Solubility Values – 20K Years

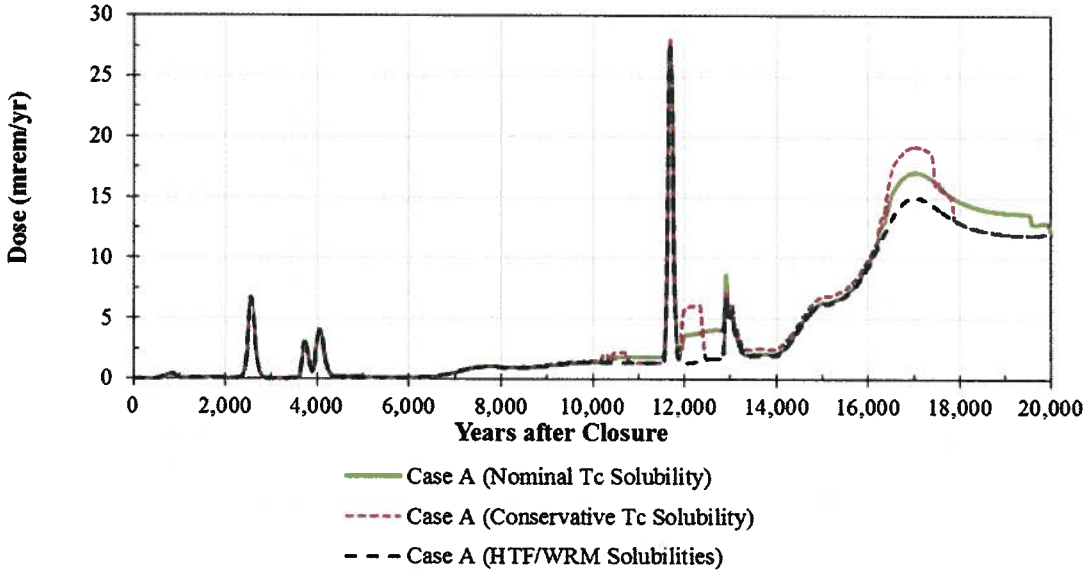
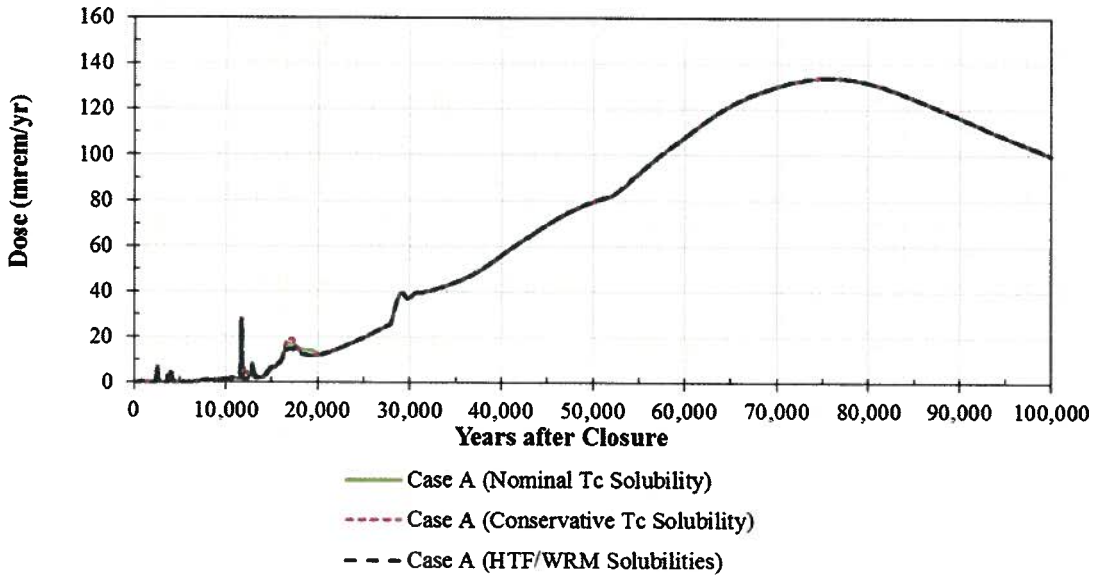


Figure 8: HTF Peak MOP Doses for Various Tc Solubility Values – 100K Years



Uranium Solubility Variability

To study the potential impact that uranium solubility has on MOP groundwater pathway doses, the FTF and HTF GoldSim models were run using their respective model Base Case (Case A) parameters with all variables held constant except for uranium solubility values. Dose results for simulations using the different sets of U solubility values (listed in Table 3) were compared. The first set of values are the U solubilities used in the FTF PA (SRR-CWDA-2012-00106) The second set of values are the U solubilities determined in the current HTF Waste Release Model (WRM) (SRNL-STI-2012-00404) and used in the HTF PA (SRR-CWDA-2010-00128). The two other sets of values are a "conservative case" and "nominal case" that allow for examination of the effect increased U solubility would have on FTF and HTF dose results. The "conservative case" and "nominal case" are based upon a considered evaluation of preliminary waste release test data.

Table 3: Uranium Solubility

	Reduced Region II	Oxidized Region II	Oxidized Region III
Solubility Data	mol/L	mol/L	mol/L
FTF PA[1]	1.7E-09	1.60E-11	3.40E-05
HTF PA/WRM[2]	5E-09	5E-05	4E-06
Conservative Case[2]	1E-03	5E-05	5E-04
Nominal Case[2]	2E-06	4E-06	1E-04

[1] Used only in the FTF simulations
[2] Used for both FTF and HTF simulations

FTF Solubility Sensitivity

As seen in Figure 9, although the FTF PA, HTF PA, "conservative case" and "nominal case" solubility value sets are quite different (see Table 3), the dose breakthrough curves over 20,000 years show only very slight differences, indicating that the peak-dose contributions from uranium radionuclides are masked by releases of other radionuclides (such as C-14, Pa-231, Np-237, Ra-226, and Nb-93m) over the time span. Over a period of 100,000 years, the dose contributions from radionuclides such as Nb-93m and Pu-239, dominate the total dose calculations for the FTF PA, "conservative case" and "nominal case" data (see Table 3); thereby obfuscating any changes that the release of uranium or uranium-ingrowth species such as Ra-226, have on dose contributions (Figure 10). For the HTF PA/WRM data, which consists of constant but low Pu solubilities (3.2E-11 Mol/L) as shown in Table 1, because of the limited release of Pu-239, Nb-93m dominates the dose contributions over the 100,000 year time span.

HTF Solubility Sensitivity

As seen in Figure 11 and Figure 12, regardless of the U solubility values simulated (see Table 3), there is no significant impact on the HTF peak dose within either the 20,000 year time span or the 100,000 year time span, respectively.

Figure 9: FTF MOP Doses for Various U Solubility Values – 10K Years

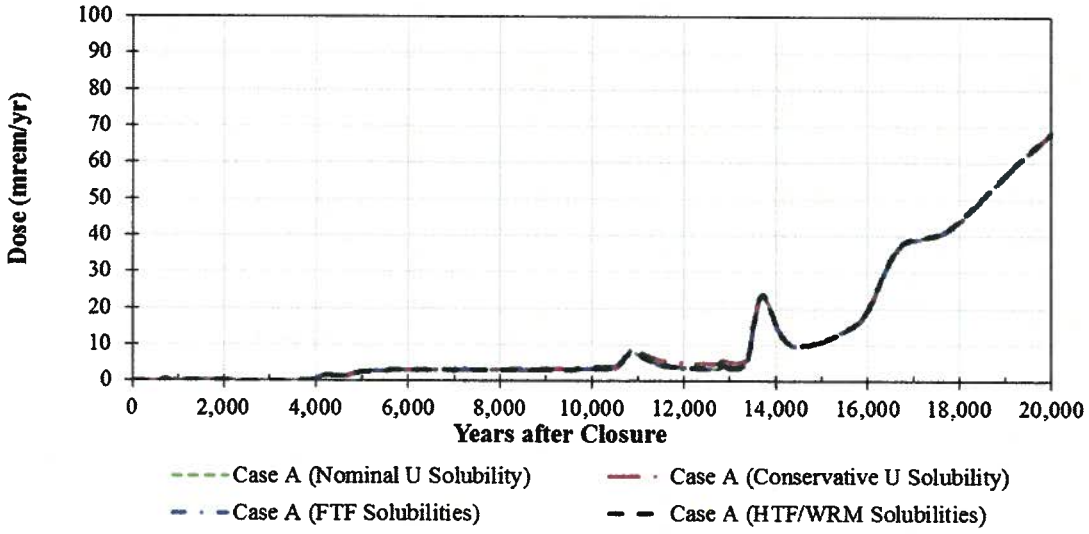


Figure 10: FTF MOP Doses for Various U Solubility Values – 100K Years

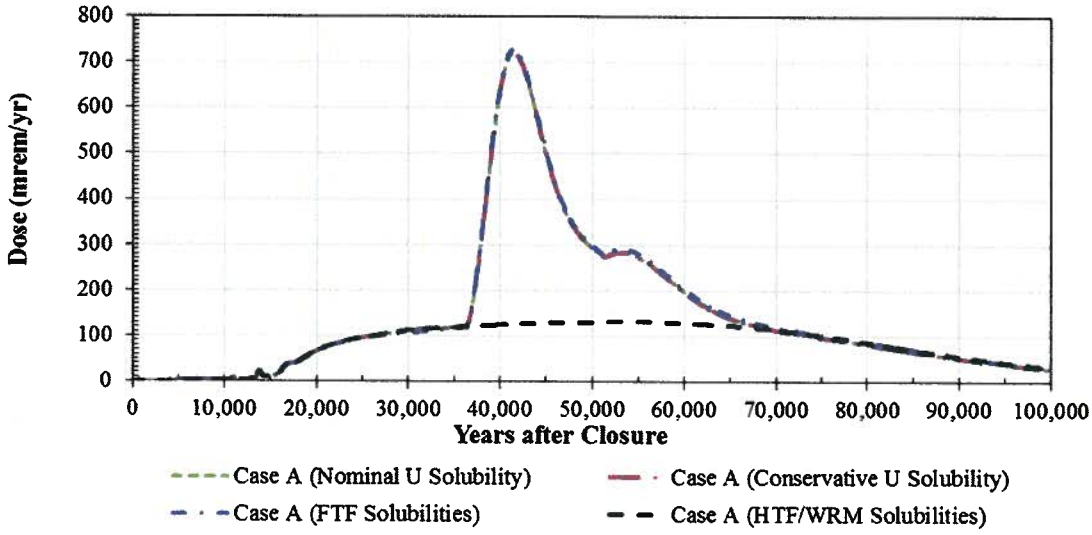


Figure 11: HTF MOP Doses for Various U Solubility Values – 20K Years

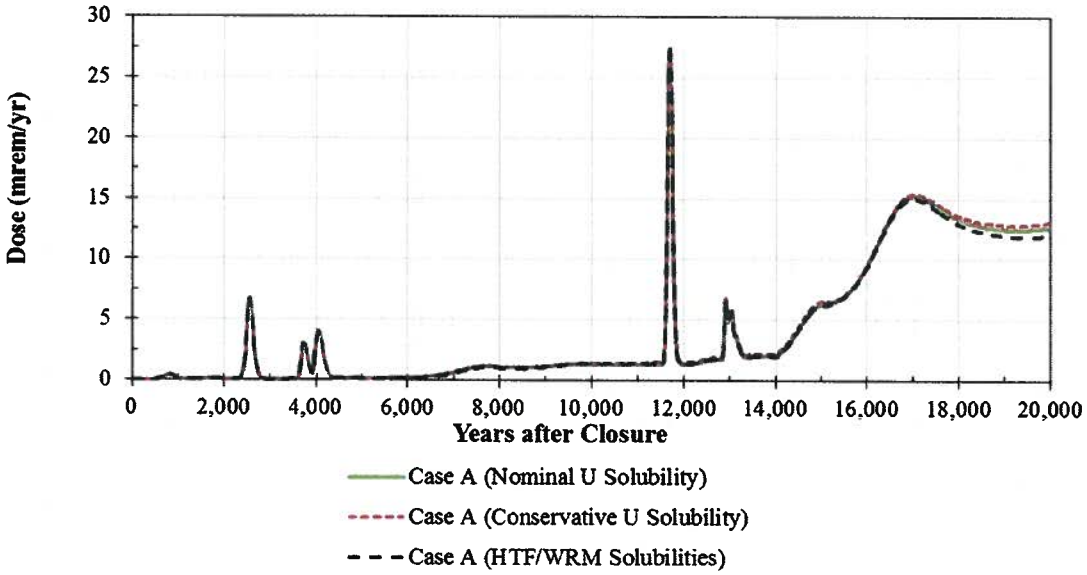
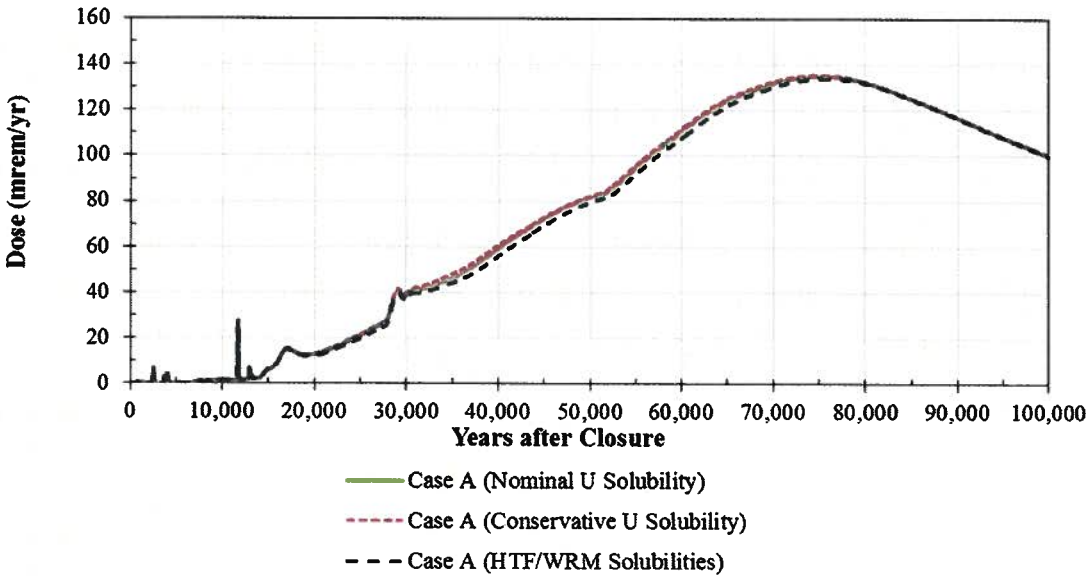


Figure 12: HTF MOP Doses for Various U Solubility Values – 100K Years



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