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Secondary Waste Management for Hanford Early Low Activity Waste Vitrification

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management
Contractor for the U.S. Department of Energy
Office of River Protection under Contract DE-AC27-99RL14047

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Secondary Waste Management for Hanford Early Low Activity Waste Vitrification

July 14, 2008

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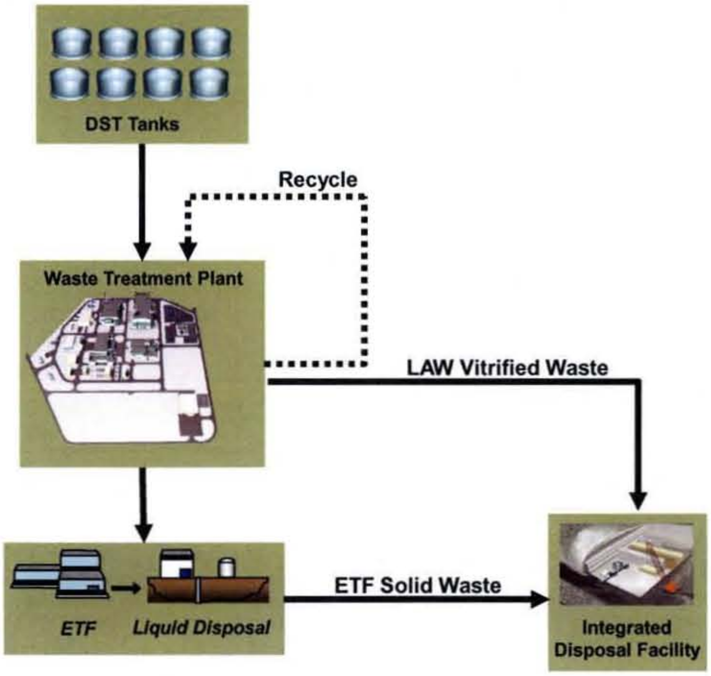
Introduction

More than 200 million liters (53 million gallons) of highly radioactive and hazardous waste is stored at the U.S. Department of Energy's Hanford Site in southeastern Washington State. The DOE's Hanford Site River Protection Project (RPP) mission includes tank waste retrieval, waste treatment, waste disposal, and tank farms closure activities. This mission will largely be accomplished by the construction and operation of three large treatment facilities at the Waste Treatment and Immobilization Plant (WTP).

- a Pretreatment (PT) facility intended to separate the tank waste into High Level Waste (HLW) and Low Activity Waste (LAW),
- a HLW vitrification facility intended to immobilize the HLW for disposal at a geologic repository in Yucca Mountain, and
- a LAW vitrification facility intended to immobilize the LAW for shallow land burial at Hanford's Integrated Disposal Facility (IDF).

The LAW facility is on target to be completed in 2014, five years prior to the completion of the rest of the WTP. In order to gain experience in the operation of the LAW vitrification facility, accelerate retrieval from single-shell tank (SST) farms, and hasten the completion of the LAW immobilization, it has been proposed to begin treatment of the low-activity waste five years before the conclusion of the WTP's construction. A challenge with this strategy is that the stream containing the LAW vitrification facility off-gas treatment condensates will not have the option of recycling back to pretreatment, and will instead be treated by the Hanford Effluent Treatment Facility (ETF). Here the off-gas condensates will be immobilized into a secondary waste form; ETF solid waste.

Figure. 1. Simplified diagram displaying the location of the WTP recycle stream.

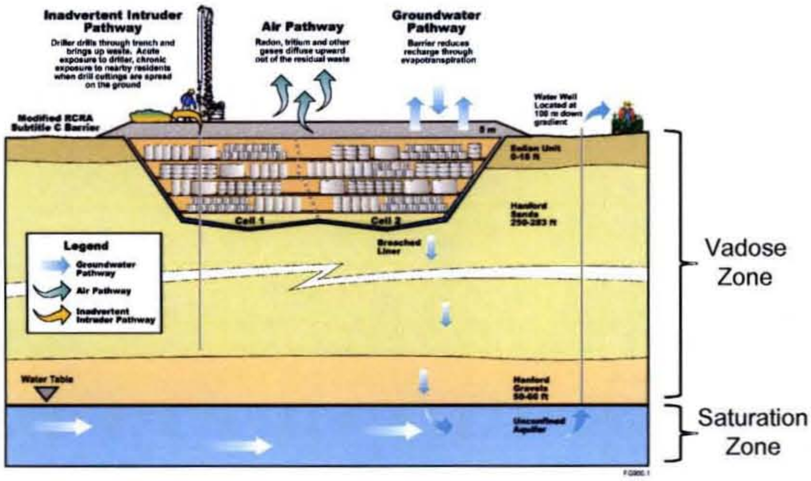


From a groundwater contamination perspective, ETF solid waste is a less stable waste form than vitrification glass and therefore results in an increased potential for releasing contaminants into the soil. The use of this early LAW system has been predicted to increase the groundwater contamination of several waste constituents. To address this issue a study was performed by CH2M Hill including the contaminants of concern (COCs) ^{99}Tc, ^{129}I, ^{238}U, NO_3, Cr, Hg, and ^{Total}U. Also, several approaches to reduce the environmental impact of early LAW were evaluated.

Methods of Analysis

Five variables were considered in the evaluation of groundwater contamination. Two of them, the vadose zone recharge rate and the vadose distribution coefficient, are dependant on the natural characteristics of the environment and are largely independent of decisions made in plant design. The vadose zone recharge rate describes the rate at which surface water moves through vadose zone into the saturation zone as shown in figure 2. The vadose distribution coefficient deals with the extent to which a component is withheld as a solid, within the vadose zone, before the remaining liquid phase portion is allowed to pass on to the saturation zone.

Figure. 2. Vadose Zone and Zone of Saturation at Hanford IDF.



Besides these two natural variables, three controllable variables that are affected by the overall waste treatment strategy were also studied by CH2M Hill. These variables contained different approaches to lower the groundwater impact of utilizing early LAW and were:

- the type of early LAW feed,
- the modification to the WTP LAW secondary waste stream, and
- the ETF solid waste form performance.

The first proposal is to change the feed content to the LAW vitrification facility used during the early LAW time period. By selecting a feed stream with lower ^{99}Tc content during the five year period when the LAW vitrification facility is unable to make use of a recycle stream, a lower quantity of ^{99}Tc will be captured in the ETF solid waste form during the plant's lifetime. A second approach is to simply recycle the entire secondary waste stream after concentration, to be stored in double shell tanks for storage until the completion of the WTP construction. An alternative to this second approach involves removing the ^{99}Tc from the secondary waste through an additional unit operation, such as ion exchange. Finally, a third approach entails improving the physical properties of the ETF solid waste form in order to slow its release of contaminants into the groundwater.

These five variables, two natural and three designed, were studied using a parametric analysis. Table one describes the range of each of the variables that was studied.

Table 1. Range of Variables

^{99}Tc Feed Approaches - (1,175 MT Na/yr for both WTP ILAW and 1-line STP)	**Change to WTP LAW Secondary Waste Stream**	**ETF Solid Waste Form Performance – Diffusion Coefficient, De (cm2/s)**	**Vadose Zone Recharge Rate (mm/yr)**	**Vadose Distribution Coefficients , Kd (ml/g)**
			1) High – 4.2	
		1) Low – 3.0 E-8	2) Natural – 0.9	1) Low – All zero
Reference ^{99}Tc – DST supernatant, DST salt cake, & high SST salt if needed	**1) None - No ^{99}Tc removal nor any recycle of WTP LAW secondary liquid waste streams**	**2) Mid – 5.0 E-9**	**3) Base – 0.5**	**2) Base – ^{129}I = 0.1 U = 0.2 Others = 0**
Mid ^{99}Tc – SST salt cake West (Sound tanks in U, S or SX farms) & DST feed tanks	2) Remove ^{99}Tc - Reduction of ^{99}Tc in Vitrification Secondary Waste Destined to ETF by at least a factor of 100	3) High –5.0 E-11	4) Low – 0.1	
Low ^{99}Tc – SST salt cake East or West & DST feed tank(s)	3) Recycle - Recycle of WTP ILAW secondary waste streams (Equivalent to No Early LAW)			
Estimated Range of Parameter				
5	>100	600	42	0 for ^{99}Tc

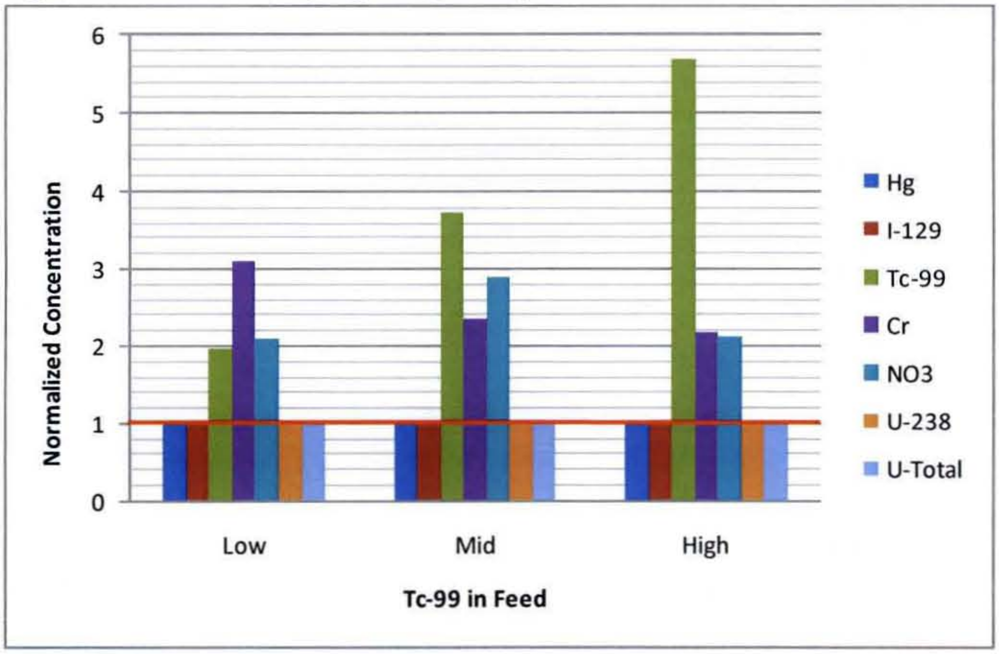
Using the variables from table 1, there is a possibility of 216 parametric combinations. However, in this study, only 18 cases were required to assess variable sensitivity and reach a variable technical solution. Charts were created depicting the resulting change in groundwater concentration when only one variable was changed while holding the others constant. Each case was normalized to a base case of no early LAW, to observe the relative effect on groundwater that utilizing early LAW would create. The effect of changing each variable was measured by comparing it to a reference case (displayed in bold in the table 1). The reference case is defined as using a high (easily accessible) ^{99}Tc feed, not employing any ^{99}Tc removal or recycle operations, using a mid performance ETF solid waste form, and assuming base case recharge rates and Kd coefficients.

To determine the various distributions of waste forms resulting from each variable selection, a system mass balance spread sheet was constructed. This balance allowed the effect of changing the system feed, and changing the action taken to the WTP LAW secondary waste stream, to be enumerated. This system mass balance essentially calculated the quantity of contaminants which would be present in: WTP vitrified glass, bulk vitrified glass, residual products from bulk vitrification, ETF solid waste, and other forms of solid waste (HEPA filters, ect.). For each of these various waste forms a groundwater contamination contribution factor, normalized to a unit mass of component waste, was used to determine the contribution of each waste form to groundwater contamination. This groundwater contamination was evaluated as the maximum groundwater concentration present in a 10,000 year window.

Results

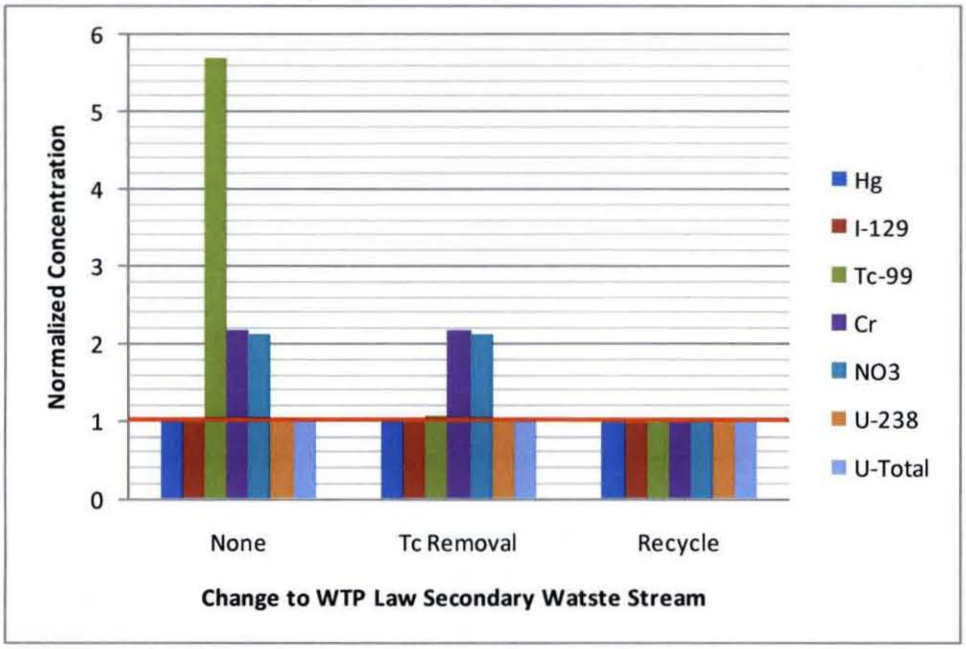
In figure 3, the normalized maximum groundwater concentration ratio in a 10,000 year window is shown for seven contaminants paired with three types of waste feed. As shown, the effect of different feeds is fairly small on the groundwater concentration for both Hg, ^{129}I, and uranium. However, the concentration ratio of ^{99}Tc increases approximately three fold between the low ^{99}Tc feed and the high ^{99}Tc feed. In this diagram the high ^{99}Tc feed is representative of the waste within the double shell tanks, and represents approximately a three fold increase in ^{99}Tc groundwater concentration ratio over the low ^{99}Tc feed. While changing to a low or mid ^{99}Tc feed lowers the groundwater concentration ratio of ^{99}Tc, it actually increases the concentration of Cr and NO_3 due to tank inventory differences between the selected feed groups. This is due to the categorization of feeds being based on their ^{99}Tc content, while being independent of nitrate and chromium.

Figure. 3. Effect of Varying the Quantity of ^{99}Tc in the Feed



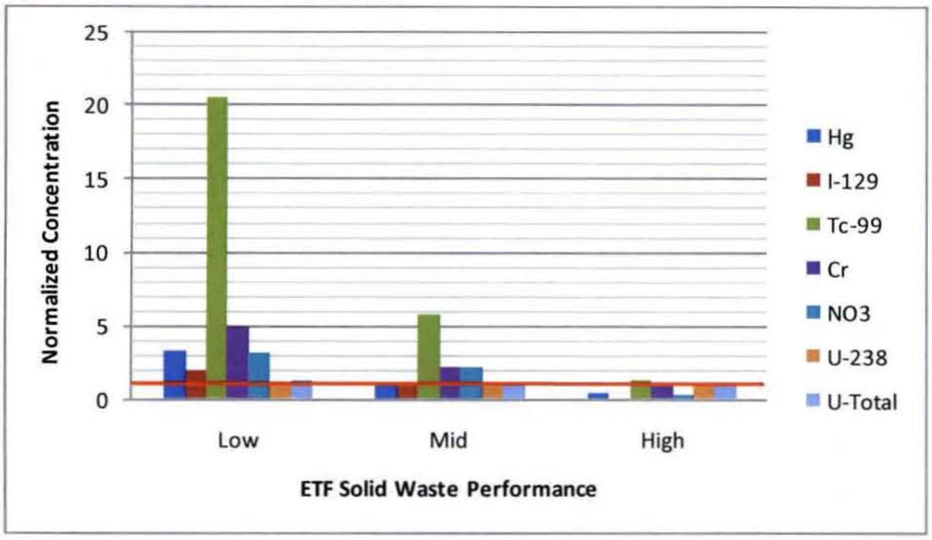
In figure 4, the effect on groundwater contamination attributed to three different methods of handling the IPS secondary waste stream is shown. The first method, None, represents all secondary waste ^{99}Tc going to the ETF. In the second method, Tc Removal, 99% of the ^{99}Tc within the secondary waste is removed. This 99% represents a reasonable ^{99}Tc removal estimate and could be accomplished through ion exchange. The final method, Recycle, represents all secondary waste being sent back to the tank farms after concentration. As shown, there is a negligible difference in groundwater concentration between ^{99}Tc removal and recycle for ^{99}Tc.

Figure. 4. Effect of Varying the Change to WTP LAW Secondary Waste Stream



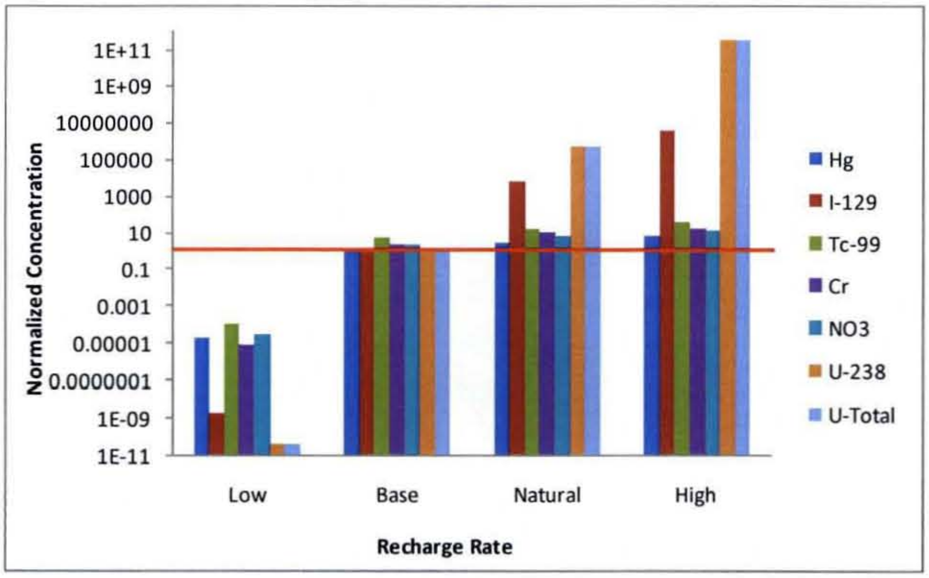
In figure 5, the effect of ETF solid waste performance on groundwater contamination is shown. As depicted, the concentration ratio of ^{99}Tc increases to over 20 times that of the baseline when low performance ETF solid waste is used. However, when a high performance solid waste form is used, the concentration ratio is only 1.22 with other contaminants at or below the baseline. The concentration ratios of other contaminants also decrease with increasing ETF solid waste performance.

Figure. 5. Effect of Varying Solid Waste Form Performance



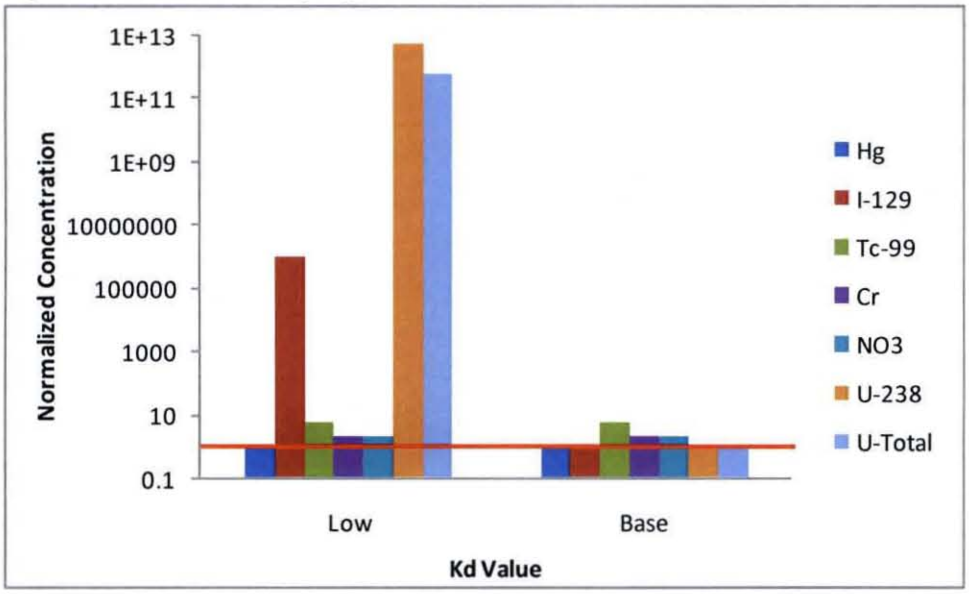
In figure 6, the groundwater contamination ratio dependence on the assumed recharge rate is shown. Take note that this chart is on a logarithmic scale, due to the large effect this variable has on all concentrations. As shown here, for high recharge rates, the concentration ratios of ^{129}I and uranium become much higher than the baseline. This large change results from bringing the peak concentration from way beyond 10,000 years (e.g., the peak groundwater concentrations occur at about 30,000 years in the future) into the 10,000 year window. In order to avoid high recharge rates increasing groundwater contamination, specialized barriers (caps) will be designed to go over the IDF and reduce the recharge rate through the process of evapotranspiration, as shown in figure 1. However, even with these measures taken, the recharge rate is still largely dependant on the natural variables of the Hanford site, as well as the performance of the caps over time.

Figure. 6. Effect of Varying the Assumed Vadose Zone Recharge Rate



In figure 7, the normalized maximum groundwater concentration ratio in a ten thousand year window is shown for seven contaminants paired with two different predicted values of Kd[*]. When base Kd values are chosen, ^{129}I is assumed to have a Kd value of 0.1, U is assumed to be 0.2, and all others 0. The base case represents the best possible estimate of actual Kd values. The low Kd option refers to all contaminants having a Kd of 0. This represents the "worst case scenario" and is not credible, but it is bounding. As shown, the selection of Kd values has a profound effect on estimated groundwater concentration. When a low Kd is used for Uranium, a resultant increase of over 11 orders of magnitude takes place. This shows that this system is extremely dependent on the values of Kd.

Figure 7. Effect of Varying the Vadose Distribution Coefficient



Conclusion

Of the seven COCs analyzed, only three (^{99}Tc, Cr, and NO3) were particularly sensitive to changes in the design variables. The groundwater concentrations of Uranium and ^{129}I were found to be mostly independent of the design variables, while being very dependent on the natural variables. Figure 8 summarizes the effect of utilizing the different options related with decreasing the groundwater contamination associated with early LAW.

In addition to the effectiveness of each option in reducing groundwater contamination, other factors to be included in their evaluation include the costs and inherent risks of each option. Using a medium or low ^{99}Tc feed would require transporting feed from SST's not close in

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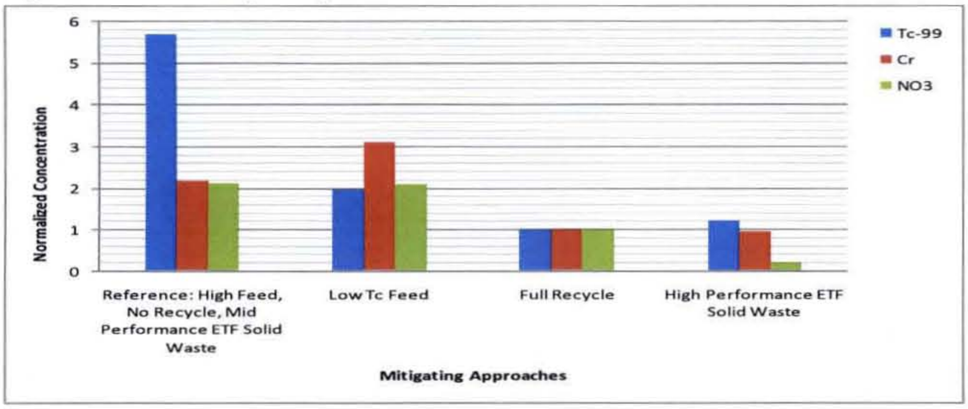
[*] These are comparative estimates based on rough analyses of impacts to groundwater. Detailed studies with extensive uncertainty analysis will have to be completed to satisfy regulatory requirements.

proximity to the LAW vitrification facility. Also retrieval of this SST waste at a rate needed to sustain LAW vitrification operations will be very difficult in the early stages of waste retrieval while new retrieval technologies are being tested and developed. Although these tanks would need to be emptied eventually, doing so during the five year early LAW time period would accelerate the needed funding into the near term. Recycling the secondary vitrification waste back to the tank farms would use up valuable double-shell tank (DST) space that could be used for the retrieval and closure of SSTs instead. Also, due to high halide concentrations in the secondary waste resulting from vitrification operations, new specialized equipment would be required to concentrate the waste before placing it back in the DSTs. Increasing the performance of the ETF solid waste is seen as a risk since the development work to achieve the needed levels of performance has not been completed.

Figure 8 – Summary Graph



Cost Range:	$400M-$450M	$35M-$135M	$35-$40M
Key Programmatic Risks:	•Retrieval rates of salt cake assumed leaker SSTs unlikely to meet LAW treatment feed demand based on experience	•Negative impact on DST space with Recycle (up to 0.5 Mgal/yr) •Potential corrosion issue due to high halide concentration	•Acceptability ETF solid waste as secondary waste form is uncertain •Performance would be increased regardless of early LAW

As shown in figure 8, it was found that while all approaches contributed to the reduction of groundwater contamination, using a full recycle and improving ETF solid waste form performance were found to be the most effective. Of these, improving ETF waste form performance was the most practical and cost effective approach, but other approaches could be integrated and implemented to further reduce uncertainty and long term risk, if necessary to meet the eventual disposal system performance standard, once established.

Ben Unterreiner is a summer intern working with CH2M Hill from Montana State University. He can be reached at bunterreiner@hotmail.com. Tom Crawford is an engineer working in technical integration and Fred Mann is a physicist working in the vadose zone program.

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Extra Images

The Hanford complex in Southeastern Washington.

The Integrated Disposal Facility (IDF) – used for permanent storage of vitrified LAW and ETF solid waste.

A Hanford single shell tank farm

Hanford's Effluent Treatment Facility (ETF) – Used for the treatment of dilute waste streams.