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U.S. Department of Energy Office of Environmental Management

The EM Center for Sustainable Groundwater and Soil Solutions at the Savannah River National Laboratory

Technical Evaluation of Soil Remediation Alternatives at the Building 812 Operable Unit, Lawrence Livermore National Laboratory Site 300

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### Technical Evaluation of Soil Remediation Alternatives at the Building 812 Operable Unit, Lawrence Livermore National Laboratory Site 300

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Appendix A: Technical Assistance Request

## **Executive Summary**

The Department of Energy Livermore Site Office requested a technical review of remedial alternatives proposed for the Building 812 Operable Unit, Site 300 at the Lawrence Livermore National Laboratory. The team visited the site and reviewed the alternatives proposed for soil remediation in the draft RI/FS and made the following observations and recommendations.

Based on the current information available for the site, the team did not identify a single technology that would be cost effective and/or ecologically sound to remediate DU contamination at Building 812 to current remedial goals. Soil washing is not a viable alternative and should not be considered at the site unless final remediation levels can be negotiated to significantly higher levels. This recommendation is based on the results of soil washing treatability studies at Fernald and Ashtabula that suggest that the technology would only be effective to address final remediation levels higher than 50 pCi/g.

The technical review team identified four areas of technical uncertainty that should be resolved before the final selection of a preferred remedial strategy is made. Areas of significant technical uncertainty that should be addressed include:

- Better delineation of the spatial distribution of surface contamination and the vertical distribution of subsurface contamination in the area of the firing table and associated alluvial deposits
- Chemical and physical characterization of residual depleted uranium (DU) at the site.
- Determination of actual contaminant concentrations in air particulates to support risk modeling.
- More realistic estimation of cost for remedial alternatives, including soil washing, that were derived primarily from vendor estimates.

Instead of conducting the planned soil washing treatability study, the team recommends that the site consider a new phased approach that combines additional characterization approaches and technologies to address the technical uncertainty in the remedial decision making. The site should redo the risk calculations as the future use scenario has changed for the site. As a result, the existing model is based on very conservative assumptions that result in calculation of unreasonably low cleanup goals. Specifically, the review team proposes that LLNL consider:

- Revising the industrial worker scenario to a reasonable maximum exposure (RME) for a site worker that performs a weekly walk down of the area for two hours for 25 years (or an alternative RME if the exposure scenario changes).
- Revising the ESSI of 2 mg U per kg soil for the deer mouse to account for less than 0.05 of the total ingested uranium being adsorbed by the gut.

- Revising bioaccumulation factors (BAFs) for vegetation and invertebrates that are based on 100 mg of soluble uranium per kg of soil, as the uranium concentration in the slope soil does not average 100 mg/kg and it is not all in a soluble form.
- Measuring actual contaminant concentrations in air particulates at the site and using the actual values to support risk calculations.

The team recommends that the site continue a phased approach during remediation. The activities should focus on elimination of the principal threats to groundwater by excavating (1) source material from the firing table and alluvial deposits, and (2) soil hotspots from the surrounding slopes with concentrations of U-235 and U-238 that pose unacceptable risk. This phased approach allows the remediation path to be driven by the results of each phase. This reduces the possibility of costly "surprises", such as failure of soil treatment, and reduces the impact of remediation on endangered habitat. Treatment of the excavated material with physical separation equipment may result in a decreased volume of soil for disposal if the DU is concentrated in the fine-grained fraction, which can then be disposed of in an offsite facility at a considerable cost savings. Based on existing data and a decision to implement the recommended phased approach, the cost of characterization, excavation and physical treatment of the contaminated materials is roughly estimated to be one third to one fourth of the cost of the current baseline treatment. This is an estimated cost; the actual cost of the project will be sensitive to actual soil/sediment volumes that can be refined with the results from characterization studies.

The technical team encourages the site to promote a more holistic approach during remediation of contaminated sediments at Site 300. It is true that the presence of low levels of residual DU on the steep slopes may stress the ecosystem, as it is not possible to block the exposure of resident biota. It is clear that remediation of the primary source areas will reduce potential effects to humans. However, the site should consider that excavation of the slopes will profoundly disrupt the ecosystem and it may take decades to recover. The site should consider very invasive remedial approach only in areas primarily impacted by airborne contamination. In fact, ecosystem recovery may occur more quickly if the very low levels of residual contamination are left in place and monitored.

## 1.0 Introduction

The Department of Energy, Livermore Site Office (LSO), is responsible for remediation at the Lawrence Livermore National Laboratory (LLNL) Site 300 experimental test facility. A draft Remedial Investigation/Feasibility Study (RI/FS) was prepared for the Building 812 Operable Unit (OU) under the terms of the Site 300 Federal Facility Agreement between DOE, the U.S. Environmental Protection Agency (EPA), California Department of Toxic Substances Control (DTSC), and the California Regional Water Quality Control Board. The purpose of the RI/FS is to identify the nature and extent of contamination, to evaluate impact to human and ecological receptors, and to evaluate remedial alternatives in accordance with CERCLA guidance.

A major component of the RI/FS forms the basis for evaluating and selecting technologies to remediate contaminated media at the Building 812 OU. The proposed soil remedial alternatives identified in the draft RI/FS include:

- 1. Excavate shallow subsurface soil at the Building 812 Firing Table (up to 6 feet deep) and all rippable surface and subsurface soil with depleted uranium and metal concentrations exceeding the PRG or background (if PRG is below background) and dispose offsite at a permitted landfill.
- 2. Excavate and treat shallow subsurface soil at the Building 812 Firing Table (up to 6 feet deep) and all rippable surface and subsurface soil with depleted uranium and metal concentrations exceeding the PRG or background (if PRG is below background) using soil washing; replace treated soil.
- 3. Excavate and solidify, stabilize, and consolidate shallow subsurface soil at the Building 812 Firing Table (up to 6 feet deep) and all rippable surface and subsurface soil with depleted uranium and metal concentrations exceeding the PRG or background (if PRG is below background).
- 4. Excavate shallow subsurface soil at the Building 812 Firing Table (up to 6 feet deep) and all rippable surface and subsurface soil with depleted uranium and metal concentrations exceeding the PRG or background (if PRG is below background) and dispose at an onsite constructed landfill.

The EPA and DTSC comments on the draft RI/FS indicated that their preferred alternative for the site was soil washing followed by the on-site replacement of the treated soil. They requested additional research and a field-scale pilot test be conducted to evaluate the potential effectiveness of the soil washing technology at the site. Because soil washing appears to currently be the regulator's preferred alternative, DOE concurred that conducting a treatability study would reduce uncertainties associated with this technology and allow for a better understanding of: (1) the limitations of the soil washing technology for site-specific conditions (e.g., soil clay content, solubility of site contaminants, attainment of cleanup goals), (2) cost implications, and (3) the possible need for coupling soil washing with a second technology. Due to the significant level of technical uncertainty associated with the implementation of soil washing at Site 300, the LSO requested assistance from DOE's Office of Environmental Management to assist with the analysis of remedial alternatives.

The technical review team was provided with a detailed charter that contained the following specific tasks:

- Review the draft Building 812 RI/FS with emphasis on the soil remediation options presented in Section 3 and Appendix E-2.
- Review case studies and lessons learned from other soil washing projects to assess feasibility of soil washing at the Building 812 Firing Table. The site was particularly interested in degree of success with uranium contaminated soil at other sites.
- Determine other feasible technologies/options available for remediation of contaminated soil at Building 812 Firing Table.
- Provide a briefing and report to DOE and LLNL that summarizes the team's evaluation and makes recommendations on options for soil remediation at the firing table.

A selected group of technical experts attended a workshop at the Lawrence Livermore National Laboratory from July 27 through 29, 2009. During the first day of the workshop, both contractor and DOE site personnel briefed the technical team, and took them on a tour of Site 300 and Building 812 OU. On subsequent days, the team reviewed baseline data and reports, were provided additional technical and regulatory information from site personnel, evaluated work plans, developed a general site conceptual model, determined critical issues and uncertainties, and developed specific recommendations for additional characterization and remediation activities at the site. This report documents the findings and recommendations of the independent technical review team.

## 2.0 Background

### 2.1 General Site Conceptual Model

The Building 812 OU was an explosives testing facility where charges and monitoring instruments were set on a firing table and detonated adjacent to an observation building, which contained additional monitoring devices. The firing table is a one to three foot thick gravel bed underlain by native soils. Most of the tests involved a depleted uranium core that was surrounded by a shell of high explosives or directional shots from an artillery gun. After a sufficient number of tests compacted the gravel, and it no longer had the capacity to absorb shock, the gravel and associated debris were disposed at onsite landfills or at NTS (after 1989). There is evidence that between detonations, the firing table was graded, and gravel was occasionally pushed off the side of the firing table platform into the adjacent canyon.

Given this generalized conceptual model, the nature of the experiments and the results of chemical analysis of ground water, it is reasonable to assume that residual undetonated high explosive materials are not present at the site, however, fragments of depleted uranium and oxidized depleted uranium particles are expected and, in fact, have been found in the firing table gravels, surrounding bedrock exposures, and widely dispersed as dust fallout on the surrounding slopes. The distribution of contamination at the site is expected to be heterogeneous with most of the larger fragments and particles of residual contaminants localized in the vicinity of the firing table, firing table sediments, and aggregate/sediments (Qal) that were pushed into the adjacent canyon. However, field personnel presented anecdotal evidence that indicates that large pieces of shrapnel and debris have been found at considerable distances from the firing table. The sediments/gravels in the prime source areas reach a maximum thickness of about 10 feet, the approximate depth to bedrock in this area. Additional contamination areas are found on the surrounding slopes, as the distribution of fallout particles is controlled by the blast geometry, surrounding topography, wind direction and particle size. The level of contamination on the adjacent slopes is expected to be lower in concentration, as it is likely to be composed primarily of very fine-grained materials that are widely dispersed with some larger pieces of DU. Preliminary data support this conceptual model, as greater heterogeneity is observed for measured U-238 concentrations near the firing table, relative to areas on the slopes on the opposite side of the canyon.

This distribution pattern implies that a random grab sampling pattern used to characterize the site will not reflect the true nature of the extent of contamination within the soil. For example, the presence of a small depleted uranium particle in a sample will bias the analytical result to a high level that does not represent the extent of contamination in the surrounding soil. Conversely, a sample lacking a uranium particle does not imply that the surrounding soil has no contamination.

### 2.2 Other Sites with Explosive DU Releases

The advent of DU use in munitions deployed in war zones provides other sites that may be analogous to the Building 812 Firing Table area for the nature of DU particles released to soils in explosions. In general, soils in Kuwait and Kosovo that were shelled with DU munitions contain small particles of DU. Török et al. (2004) found that uraniumrich particles in contaminated Kosovo soils had an average diameter of 0.8 um, though 15% were agglomerated with soil particles and had an average diameter of 0.9 to 1.9 um. This is similar to results reported by Danesi et al. (2003). Likewise, Lind et al. (2005) found that >50% of the DU particles in soil they studied from Kosovo were less than 1.5 um in diameter. Salbu et al. (2005) characterized DU particles from battle sites in Kuwait and found a slightly larger particle size range. Swipes of DU penetrator holes in vehicles and sands beneath DU penetrators had DU particles that ranged from 2 to 64 um with a median of 13 um.

These studies suggest that much of the DU contaminated soil at Building 812 is probably contained in small um-sized particles, though this doesn't preclude the presence of larger particles. DU fused to pieces of metal or even large pieces of oxidized DU may exist, but the soil concentration data suggest particles larger than 0.5 cm in diameter are rare. This conclusion is reached by considering that if all the U-238 in a one kg soil sample had an activity of 100 pCi/g concentrated in one spherical particle of the mineral schoepite (a relatively low density uranium mineral), the diameter of that particle would be approximately 0.9 cm.

In general, the mineralogy of Kosovo and Kuwait DU particles is mixed oxides dominated by the U(IV) oxidation state (Török et al., 2004; Salbu et al., 2005; Lind et al., 2009). However, Lind et al. (2009) do report finding DU in a variety of forms including DU metal, a DU-Fe alloy, and a DU carbide. The leachability of DU from soils varies, but Lind et al. (2009) found that in simulated gastric fluids (pH=1 HCl) DU particles from battle site soils leached significantly slower than DU from a fire in a DU storage facility. After 148 hours the maximum DU leached was 90% from the fire sample and about 86% from the Kosovo sample and 75% from the Kuwait sample. In contrast, only 24% of the DU in the battle site sands was leached after 2 hours compared to 84% of the DU from the fire samples. This suggests the form of DU is important to the dose received in the ingestion pathway.

Oliver et al. (2008a and 2008b) studied DU particles in soils from British firing ranges that had used DU munitions since the 1980s. They used a sequential extraction procedure that categorized DU as exchangeable (easily leached), reducible (associated with Fe and Mn oxides, oxidizable (associated with organic matter), and residual (associated with silicates). They found that approximately 50% of the DU was associated with organic matter. Whether it is present as U(IV) minerals stabilized by the reducing conditions afforded by the organic matter or whether it is chemically bound to the organic matter is unclear. Oliver et al. (2008b) looked at biouptake by earthworms and plants and found that DU at the firing ranges was more bioavailable than natural uranium. They calculated concentration ratios (CR), defined as the ratio of the concentration of uranium (mg/kg) in the biota to the concentration in the soil. In two samples where CRs were calculated for earthworms, they were 0.67 and 0.33. This is

despite the fact that the earthworms were likely primarily feeding on the organic matter in which DU was concentrated. The CRs in two plant samples were 0.017 and 0.018. The earthworm data are important because their CRs are less than 1 even though they are feeding on the very material in which the DU is concentrated.

One caution in comparing these analogues to the DU at the Building 812 Firing Table is the age of the DU. The Kosovo samples were analyzed within 1-2 years after the DU was deposited in the soil. The Kuwait samples of Salbu et al. (2005) were taken approximately 13 years after they were deposited. The samples of Oliver et al. (2008a and 2008b) are the oldest – the DU may have been in the soils for about 25 years. This may explain why a large fraction of DU in the older British firing range samples is in the residual silicate fraction, a median of 18% with a range of 3-74%. This is much higher than would be expected in the younger Kosovo and Kuwait samples based on their DU mineralogy. Furthermore, the oxidizable fraction is inversely correlated to the residual fraction (Figure 1), suggesting that soils with higher residual fractions may be more oxidized.

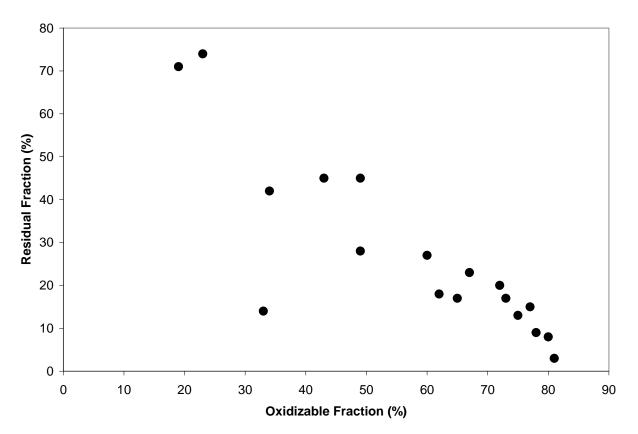


Figure 1: Residual fraction of DU versus oxidizable fraction from the BCR extraction of DU-contaminated soils at British firing ranges by Oliver et al. (2008a)

Weathering of  $UO_2$  in oxidizing soils is known to follow a path toward lower solubility. The mineralogical sequence goes from  $UO_2$  to less soluble U(VI) hydroxides such as schoepite and ends with relatively insoluble silicates such as uranophane (Schindler et al., 2009). The youngest DU at the Building 812 site is about the same age as the DU Oliver et al. (2008a) studied. Thus, it might be expected that as least as much DU is in the silicate fraction as Oliver (2008a) found in the British firing range soils. The soil conditions at the Building 812 are conducive to forming uranophane considering the reaction:

 $2UO_2 + O_2 + Ca^{+2} + 2SiO_{2(a)} + 7H_2O = 2H^+ + Uranophane$ 

Pore waters of calcareous soils, which exist at Site 300, often have elevated Ca<sup>+2</sup>, SiO<sub>2</sub>, and OH<sup>-</sup> all of which promote the formation of uranophane or other uranium silicates. If a significant fraction of the DU in the Building 812 soils is in a silicate phase, it lowers the DU leachability and bioavailability substantially. Figure 2 shows solubility curves for hydroxides UO<sub>2</sub>(OH)<sub>2</sub> and schoepite compared to uranophane and soddyite (another U-silicate) calculated in equilibrium with calcite and quartz. At a pH of 7.5, typical of calcareous soil pore fluids, and assuming chemical equilibrium, the solubility of schoepite is approximately 790 pCi/L whereas the solubility of uranophane is approximately 0.2 pCi/L. Equilibrium with quartz ensures a relatively low silica concentration; at higher dissolved silica concentrations, the solubility of the silicate minerals would be even lower.

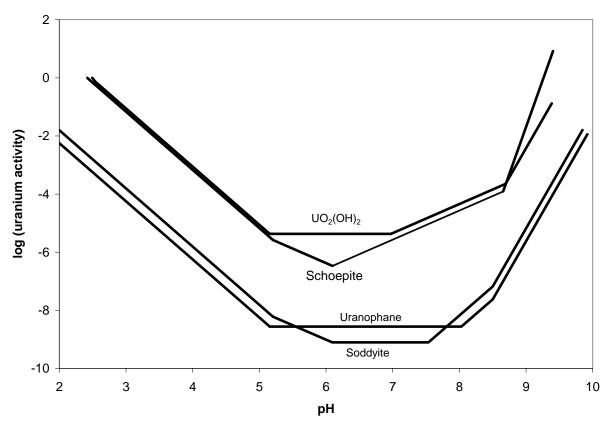


Figure 2: Solubility curves for four uranium minerals calculated with The Geochemist's Workbench® (Bethke, 2005) assuming equilibrium with calcite and quartz (note that uranium activity refers to chemical activity rather than radioactivity).

There is also an adsorption component for the DU in soil. Understanding the distribution of DU between uranium minerals and a sorption fraction is important to developing the proper remedial action, and this is the basis for recommending additional characterization studies. The form of uranium is important to bioavailability and also to any attempt to separate the DU from soil. For example, if a substantial fraction of the DU is in a silicate form, removing it by chemical extraction (e.g., soil washing) will be difficult.

### 2.2 Previous Soil Washing Experience for Uranium (Fernald/Mound)

Soil washing may be a cost-effective remedial objective if the uranium forms are soluble at ambient temperature, of similar size, and of uniform distribution within the material. These conditions were not met at the Fernald and Mound sites, and are seldom met at any site. At Fernald, uranium was present as oxide, phosphate, and silicate phases, and adsorbed onto iron oxides (Buck et al., 1994). The variation in distribution, solubility and particle size precluded the use a single washing reagent to remove the needed mass of uranium from the soil. For example, carbonate extractions removed approximately 60 to 85 percent of the uranium at circum-neutral pH, while citric acid/citrate removed 68 to 99 percent at pH 3 to 4 (Francis et al., 1993). However, it is not cost effective to maintain a low pH in Fernald soils, as approximately 50 percent of the soil particles are carbonate grains.

The two Fernald soils used in these studies contained 387 and 470 mg U per kg of soil (Francis et al., 1993). Since the carbonate extraction is the only viable cost-effective approach for Fernald soils, the removal of only 60 percent of the uranium from the waste incinerator soil (470 mg/kg) resulted in a residual U concentration of 187 mg/kg, which was far above the final remediation level of 82 mg/kg. Additionally, the bench-scale studies used soil that was processed and homogenized prior to testing. The extensive pre-processing of the soil facilitates reagent contact with the grains, and this would not occur in a large-scale operation because the time needed to process the soil and achieve the homogenization would add a significant cost to the treatment. The technical challenges exposed by the bench-scale tests indicated soil washing was not a cost-effective solution at the Fernald site.

Other soil washing pilot studies have shown similar results. Treatment of Ashtabula site soils containing uranium dust with a 0.2M sodium carbonate/sodium bicarbonate solution (Kulpa and Hughes, 2001) achieved 85% uranium removal, but the clean soils retained a uranium concentration of 15-18 pCi/g. Likewise, a pilot demonstration of soil washing at the 300-FF-1 operable unit (Hanford) cleaned soils to the point that the sandy fraction had a uranium concentration of 28.5 pCi/g (Mann, 1999). Dermont et al. (2008) lists 37 field applications of soil washing of various metals and most were not able to achieve 90% removal. Choy et al. (2006) achieved 50-60% removal of DU from firing range soils using either citric acid or sodium bicarbonate and was able to achieve up to 80% removal if a hydrogen peroxide step was added. However, this was in a soil containing less than 10% silt and clay. These studies suggest that soil washing at Building 812 is unlikely to be viable because of the very low clean-up goals and the high silt and clay content of the soils.

# 3.0 Determination of Technical and Cost Uncertainty

### 3.1 Technical Uncertainty associated with nature and extent of contamination

There are two significant areas of technical uncertainty associated with the Building 812 OU; (1) the spatial distribution of contamination, and (2) the physical and chemical form of the contaminant. To fully understand the risk posed by the contaminated soils and to plan actions to minimize unacceptable risks, these uncertainties must be identified and clarified during the remedial planning process. Additional field characterization is required to reduce the level of uncertainty to an acceptable level.

The nature of contaminant release suggests a very heterogeneous spatial distribution of the contaminants. The spatial distribution of contaminant is critical to understanding where the main sources to groundwater are and whether there are any hot spots of contamination. The point source data collected provides a good basis for constraining an additional survey of contaminant distribution, but is not sufficient for planning the remediation strategy.

The contaminant form determines its solubility, its susceptibility to removal by physical or chemical means, and its bioavailability. Studies of DU from various battle sites suggest the forms associated with pyrophoric releases can be variants of the mineral shoepite, other oxides of both U(IV) and U(VI) valence states, alloys with iron, carbides, or other phases. Threat to groundwater from the DU in soils depends on what phases are present. Likewise, bioavailability depends on the phases present. For example, Lind et al. (2009) found there was a significant difference in bioavailability between DU from exploded ordinance in Kosovo and DU released in a storage facility fire because of the different forms of DU present. Thus, analyses of the chemical forms of DU present in the contaminated soils is important to understanding the risk the DU poses, as well as the best way to mitigate unacceptable risk.

The physical form of the contaminants is also important, as it affects mobility and potential impact to the various exposure pathways. Discrete particles of DU phases are likely to respond differently to wind and surface water mobilization than particles that are agglomerates of DU and soil particles. Particle size is also critical to mobilization of DU by wind and surface water. Thus, analyses of soil samples for chemical form and particle size distribution are important to understanding the risk the contamination poses and how best to mitigate any unacceptable risk.

### 3.2 Cost Uncertainty

A large suite of remedial technologies were evaluated and screened for applicability given the specific contaminants of concern present and sediment properties found at the site. Four specific technical approaches were identified as potentially viable, and these technologies were retained if the cost of implementation was considered to be low or medium. Since cost is a major driver for the selection of the preferred alternative, it is

imperative that the cost estimates be as robust and accurate as is possible. Based on efforts completed at Fernald, the technical team reviewed and made an effort to better estimate costs of implementation of the remedial alternatives, especially for the less mature technologies such as soil washing.

The four alternatives that were considered are listed below. Alternative 1 for soil remediation is no action, and this is not a viable path forward due to the risk posed to human and ecological receptors by the contaminants in the soil. The detailed cost analysis for Alternatives 2 through 5 (Appendix F) is broken into four components (Tables F.7 through F.10):

Component A – Post-excavation soil verification sampling and analysis

Component B – Risk and hazard management

Component C – Soil excavation

Component D – (Specific to each alternative)

The costs for Components A, B and C are the same for Alternatives 2 through 5, and they are based on previous LLNL project experience (Bldg 850 site). Component D for each of the alternatives is:

- Alternative 2 Shipping and offsite disposal
- Alternative 3 Soil treatment using soil washing
- Alternative 4 Soil treatment using solidification and consolidation

Alternative 5 – Onsite disposal in an engineered landfill

Shipping and offsite disposal costs (Component D, Alternative 2) are straightforward and well constrained by vendor quotes. Soil treatment using solidification and consolidation (Component D, Alternative 4) is presently being performed at the Building 850 site, and this experience is used as the basis for the component cost. Onsite disposal in an engineered landfill (Component D, Alternative 5) is a mature remedy that has been selected and executed at multiple DOE sites (e.g., Fernald, Oak Ridge, Savannah River), and the design and construction costs are well documented and robust. Therefore, the principal uncertainties with the cost analysis in Appendix F are the volume of soil that must be excavated and the cost to perform soil washing.

Soil volumes were estimated for the shallow deposits mantling the slopes (52,740 yd<sup>3</sup>) and the contaminated soil beneath the firing table that constitutes a source of uranium to ground water (4,440 yd<sup>3</sup>). For the shallow deposits, the area perimeter was extended beyond the soil samples with U-238 above 3.1 pCi/g and LLNL used Earth Vision modeling software to account for slope geometry. This area (712,000 ft<sup>2</sup>) was multiplied by an assumed soil depth of 2 ft to arrive at the soil volume of 52,740 yd<sup>3</sup> for the slope deposits. Borings within the firing table area (12,000 ft<sup>2</sup>) indicated a depth of about 10

ft, and these values were used to calculate a volume of 4,440 yd<sup>3</sup> for the deep sediment bed.

The mantle of soil on the slopes is generally no deeper than 1 foot, and in many places there are rock outcrops without soil. A depth of 2 feet was used to obtain a conservative estimate of the soil volume, but additional depth profiling in the slope mantle, firing table, adjacent soil pile and Qal deposits could reduce the uncertainty in the estimate of the remedial soil volume.

Additionally, depth profiling and a revised estimate of the soil volume could substantially reduce the remedial costs. For example, if the soil volume is reduced by 50 percent, the excavation, transportation, and disposal costs would be reduced by 50 percent (Alt 2 from \$44.0 M to \$22.3 M; Alt 3 from \$16.0 M to \$8.4 M; Alt 4 from \$18.8 M to \$10.1 M; Alt 5 from \$8.5 M to \$5.2 M). However, it is also realized that a detailed excavation design for the slopes is unlikely to be executed with precision under the difficult field conditions, and this generally leads to an increase in soil volume. This problem emphasizes the logic behind taking another look at either physical separation to reduce the soil volume and/or a risk scenario that allows the slope soil to remain in place.

Vendor quotes for soil washing contain insufficient detail for a reliable cost estimate because the uranium solid phases and uranium partitioning onto soil particles have not been identified and evaluated to provide the needed data for an accurate assessment of the mechanical and chemical steps required to execute a successful soil washing remedy. Extensive characterization and leaching studies were carried out from 1992 through 1994 to evaluate soil washing at the Fernald site (Kneff et al., 1992; Lee and Marsh, 1992; Kneff et al., 1993; Schilk et al., 1993; Cunnane et al., 1993; Francis et al., 1993; Lee et al., 1993; DOE, 1993; Bertsch and Hunter, 1994; Buck et al., 1994; Mason et al., 1994; Turney et al., 1994). Results from these investigations identified four potential protocols for washing Fernald soil, and the costs associated with the protocols (Douthat et al., 1995a; 1995b) exceed the soil washing costs in Table F.8. A summary of the Fernald costs and comparison to Table F.8 costs is provided in Table 1.

In Table 1, the fixed capital investment (FCI) for the Fernald protocols is about 3 to 5 times greater than the estimate for Building 812; there is no start-up cost estimated for Building 812; and operational costs for Building 812 are at the lower end of the costs associated with the Fernald protocols. It is evident that there is a significant difference in the FCI for the Fernald protocols, and this is partly due to the larger facility that would have been constructed for the Fernald work. Assuming a reduction of 50% for the FCI of Fernald protocols, the cheapest protocol (heap leaching) is \$17M, relative to the \$12.8 M estimated for Building 812.

However, the costs are likely to exceed \$17 M by a substantial amount, as no single leaching protocol is successful in removing all the uranium phases present in the soil (see section on technical uncertainties). Presently, without testing results to support the soil-washing estimate of \$12.8 M for Building 812, the cost estimate for soil washing remains very uncertain and unreliable.

Table 2 summarizes the potential to reduce the cost of Component D (Table F-10 in RI/FS) by decreasing the volume of soil to remediate (66,000 to 10,000 cubic yards, primarily firing table and Qal). Note that a soil volume reduction will produce a similar scaled savings for Component C (Table F-10). Table 2 also shows that the RI/FS estimate for soil washing is likely to be too low, given that the uncertainties in the size, distribution and chemical form of the uranium particles will probably require multiple reagents as part of a soil-washing remedy. It is also noted that the cost for the physical separation facility, under Alternative 3, could be applied to Alternatives 2, 4 and 5 to reduce the volume of soil (10,000 cubic yards) that required treatment or disposal. If characterization studies indicated that the uranium contamination could be mechanically isolated in the small size fraction, and this size fraction was less than 5,000 cubic yards, the reduction in soil volume could pay for the construction and operation of the separation facility. A characterization study is needed to validate the feasibility of reducing the soil volume and a cost analysis is required to evaluate if building the facility to reduce soil volume offsets the cost of disposal.

Protocol	Fixed Capital Investment	Start-Up Costs	6-month Operational Cost*
Fernald – Aqueous	\$23,600,000	\$2,400,000	\$10,200,000
Biphasic Extraction Fernald –	\$30,700,000	\$3,100,000	\$6,700,000
Carbonate/Bicarbonate Extraction			
Fernald – Heap	\$18,700,00	\$1,900,000	\$5,700,00
Leaching with Carbonate/Bicarbonate			
Extraction			
Fernald – Tiron Extraction	\$30,500,000	\$3,100,000	\$9,900,000
Building 812 – Protocol	\$6,800,000	Not available	\$6,000,000
Unknown			

### Table 1. Soil Washing Costs for the Fernald Site and Building 812 Site

\*Estimated duration for soil washing at Building 812 site.

### Table 2: Cost Breakdown for Alternatives Off-site Disposal—Alternative 2, Component D Shipping costs

Table F.7, Draft Bldg 812 RI/FS LLNL Site 300 66,000 cubic yards 4125 truck trips ~ \$14.6 M

Unit train estimate (Fernald experience) 5000 tons per train, 100,000 tons (66,000 cubic yards) 20 unit trains @ \$500,000/train ~ \$10 M

Volume reduction estimate Process only firing table and Qal 10,000 cubic yards 625 truck trips ~ \$2.2 M Disposal fee: \$15.5 M lowered to \$2.4 M

#### Soil Washing—Alternative 3, Component D

Physical Separation

Soil handling and separation facility (no estimate in draft RI/FS) Controls dust via HEPA system Eliminates use of water for dust control FCI ~ \$2.15M (Fernald; ORNL-6882) Operational cost ~ \$19/ton (Fernald; ORNL-6882)

Extraction Process

Reagents unknown at this time (large uncertainty in vendor quotes) Table F.8, Draft RI/FS LLNL Site 300 FCI ~ \$6.8 M 6-month operating cost~\$6.0M

Aqueous biphasic extraction (Fernald; ORNL-6882) FCI ~ \$23.6 M Start-up cost ~ \$2.4 M 6-month operating cost ~ \$10.2 M

Carbonate/bicarbonate extraction (Fernald; ORNL-6882) FCI ~ \$30.7 M Start-up cost ~ \$3.1 M 6-month operating cost ~ \$6.7 M

Heap leaching w/ carbonate/bicarbonate extraction (Fernald; ORNL-6882) FCI ~ \$18.7 M

Tiron extraction (Fernald; ORNL-6882) FCI ~ \$30.5 M Start-up cost ~ \$3.1 M 6-month operating cost ~ \$9.9 M

### Solidification and Consolidation –

Alternative 4, Component D Volume reduction estimate

Process only firing table and Qal 10,000 cubic yards Consolidate & place in CAMU: \$14.5 M lowered to \$2.2 M

### Onsite Disposal – Alternative 5,

### Component D

Volume reduction estimate Process only firing table and Qal 10,000 cubic yards Design, construct, excavate and place: \$4.26 M lowered to \$0.65 M.

### 3.3 Risk Uncertainty

### 3.3.1 Human Health

Uranium is the most widespread contaminant, and other metal contaminants are collocated with uranium principally in the firing table and Qal sediments, but high metal concentrations also occur in the hill slope sediments west and south of Building 812. It is assumed that most contamination will be removed by excavating the firing table and contaminated Qal sediments, which leaves the soil mantling the surrounding slopes to serve as a risk source for the inhalation, ingestion and dermal exposure pathways. Using the very conservative assumptions summarized below, this soil mantle would also be excavated and removed from the site. However, it is recommended that a reasonable exposure model be used to allow the soil mantle to remain after hotspots and the primary sources are removed.

EPA Region IX preliminary remediation goals (PRGs) for an industrial site were used to set proposed soil clean-up levels. The receptor is an industrial worker that works 8 hr/day, 250 day/yr for 25 years. Using the industrial worker scenario and default EPA Region IX exposure values (oral reference dose (RfD<sub>o</sub>) for uranium of 2.0E-4 mg/kg-d, inhalation rate of 20 m<sup>3</sup>/day, soil dust inhaled at 0.76 ug/m<sup>3</sup>, soil ingestion rate of 100 mg/day, exposed skin surface area of 3300 cm<sup>2</sup>/day, and a soil adherence factor of 0.2 mg/cm<sup>2</sup>) the soil PRG for uranium toxicity (non-carcinogen) is 200 mg/kg. The non-carcinogen value of 200 mg U/kg soil corresponds to the maximum allowable hazard quotient (HQ) of one. The EPA Region IX PRG table does not provide a reference dose for inhalation (www.epa.gov/region09/superfund/prg), and the HQ is the sum of the ingestion and dermal exposure paths. The dominant contribution to HQ is the ingestion pathway.

EPA's PRG table at the above-cited web page lacks information for radioisotopes, yet the RI/FS reports that the uranium isotope carcinogens U-235 and U-238 have PRGs of 0.42 pCi/g and 1.8 pCi/g for an incremental lifetime cancer risk (ILCR) of 1E-6. This value is obtained if one evaluates the inhalation, ingestion and external radiation pathways (U-235+D and U-238+D slope factors:  $CSF_i = 1.01E-8 \& 9.35E-9; CSF_o= 1.63E-10 \& 2.10E-10; CSF_x = 5.43E-7 \& 1.14E-7. http://rais.ornl.gov)$ , as there is no dermal pathway evaluated for radioisotopes.

The background activity for U-238 (3.1 pCi/g) exceeds the PRG and U-235 background (0.074 pCi/g) is less than the PRG. Therefore, the proposed clean up levels for U-238 and U-235 are 3.1 pCi/g and 0.42 pCi/g, respectively (that is, whichever is greater). Note that USEPA defines the ILCR as the allowable risk in excess of background, and proposed clean-up levels of 4.9 pCi/g (3.1 + 1.8) and 0.49 pCi/g (0.42 + 0.074) for U-238 and U-235 would maintain an ILCR of 1E-6

above background. Because the total uranium concentration associated with the proposed clean-up activities is about 9.3 mg/kg, there is no concern with the residual contamination exceeding the soil PRG for uranium toxicity (200 mg/kg).

However, the above exposure scenario for the industrial worker is not a reasonable exposure scenario for the Building 812 Operable Unit, and much higher residual soil concentrations are possible if a reasonable scenario is used to calculate the ILCR. EPA guidance in the PRG Users Manual (see EPA link above) allows reasonable risk scenarios to be used in lieu of the EPA default scenario.

Based on the termination of explosive testing at the Building 812 site and general historic use, it is far more reasonable to assume that an industrial worker will inspect the site once a week for a period of two hours. This equates to 52 two-hour inspections a year for 25 years. Using this reasonable maximum exposure (RME) scenario and the EPA Region IX exposure values noted above, the U-238 and U-235 activities that correspond to an ILCR of 1E-6 are 16 and 5.5 pCi/g, respectively (Table 3; detailed calculations in Attachment I). As the ILCR is measured as risk above background, the new proposed soil clean-up targets would be 19 (16 + 3.1) and 5.6 (5.5 + 0.07) pCi/g for U-238 and U-235. If the risk above background can be negotiated to 1E-5, the proposed clean-up levels rise to 163 (160+3.1) and 55 (55+0.07) pCi/g for U-238 and U-235. This later scenario would allow all the soil to remain on the slopes.

# Table 3. Incremental Lifetime Cancer Risk and Hazard Quotient for the<br/>Reasonable Maximum Exposure of an Industrial Worker at Building<br/>812

Contaminant	pCi/g	mg/kg	ILCR^	HQ
U-235	5.5	NA	1.0E-6	NA
U-238	16	NA	1.0E-6	NA
Total U*	NA	1000	NA	1.0
U-235	55	NA	1.0E-5	NA
U-238	160	NA	1.0E-5	NA

\*There is no reference dose for inhalation reported in the EPA Region IX PRG tables, and the HQ is based on dermal and ingestion pathways, with ingestion being the primary factor in the HQ result.

^The primary driver for risk is the external radiation pathway.

### 3.3.2 Ecological

The most sensitive ecological receptor is the deer mouse, which has a calculated ESSI value of 2.0 mg U/kg soil. The ESSI value is calculated using the values in Table 2-20 (toxicity reference value; TRV), Table 2-21 (bioaccumulation factor; BAF) and Table D-3-1 (dietary fraction, DF; body weight, BW; and dry matter intake, DMI) of the draft RI/FS. Background values for surface and subsurface

soil are 9.4 and 5.8 mg/kg, respectively (Table 2-22 of draft RI/FS). Therefore, background values for uranium are the proposed clean-up levels for this ecological receptor.

Several conservative assumptions with the current ESSI calculation should be evaluated to assess the potential for increasing the ESSI and uranium clean-up levels. For example, the TRV of 0.1 mg U per kg body weight per day is based on studies that use uranyl acetate dihydrate (Paternain et al., 1989; Domingo et al., 1989a, Domingo et al., 1989b and Llobet et al., 1991), which is very soluble (77 g of  $UO_2(C_2H_2O_2)_2.2H_2O$  (43 g of U) per liter of cold water; CRC Handbook of Chemistry and Physics). Therefore, 100 percent of the uranium is available to the gut for absorption. Appendix D-2 of the draft RI/FS states that a study by Thorne (2003) indicates that

"...absorption of uranium compounds is almost always less than 0.05 and can be less than 0.01. Once it does enter the blood stream, uranium is either taken up by the tissues or excreted in the urine. In long-term chronic exposures, about 50% of the uranium entering the blood stream will have been lost by urinary excretion within 24 hours."

This is a reasonable exposure scenario, as uranium compounds exist today in the soil because they are not as soluble as uranyl acetate dihydrate. If they were, the rain would have dissolved and transported most of the uranium out of the soil.

In a similar manner, the BAFs for vegetation and invertebrates are based on 100 mg of soluble uranium per kg of soil (Sheppard and Evenden, 1992). This is not a reasonable exposure scenario for the vegetation and invertebrates that will be consumed by the deer mice in the Building 812 area. The BAFs for vegetation (0.039) and invertebrates (0.37) are likely to be much lower for flora and fauna in the Building 812 area because the uranium concentration in the slope soil does not average 100 mg/kg and it is not all in a soluble form (see discussion under Section 3.1).

Therefore, a correction should be considered for the food intake equation to account for less soluble forms of uranium. It is recommended that a gut absorption coefficient of 0.05 be adjusted by a multiplier of 0.5, and this would account for less soluble forms of uranium and a 50 percent reduction of the uranium via excretion in urine in the first 24 hours (i.e., a correction factor of 0.025 should be made to the uranium intake to account for only the soluble uranium forms that are taken up by tissue). The ESSI equation (p. 35 of draft RI/FS) would then take the following form:

ESSI  $(mg_U/kg_{soil}) = TRV/[0.025^{*}(BAFv^{*}DFv+BAFi^{*}DFi+BAFs^{*}DFs)^{*}DMI/BW]$ 

Using the correction factor of 0.025 results in an ESSI for the deer mouse of 79 mg U per kg of soil, relative to a value of 2 without the factor (see Attachment I).

The uranium soil concentration of 79 mg/kg exceeds the concentration that corresponds to the human health ILCR of 1E-6, using the RME model discussed above (16 pCi/g of U-238 is about 48 mg U/kg soil). Therefore, if reasonable models are constructed for human health and ecological receptors, demonstrating protectiveness for humans will result in compliance for ecological receptors as well.

# 4.0 Recommendations

### 4.1 Characterization

Investigations at the Building 812 operable unit are at the point in the remedial process where data collection for soil contamination is being used to locate contamination and estimate its extent, support excavation design, and to provide for closure activities. Although a significant investment has been made to characterize the site, the technical team believes that there remains a significant level of uncertainty about the amount, location and physical state of contaminated soils. Additional characterization efforts can only limit this uncertainty, not remove it. Regulatory acceptance of data collection technologies and approach should only be an issue with closure or verification data collection but the site should seek and include regulatory input throughout the data collection process.

The team makes the following recommendations for additional work to characterize the nature and extent of contamination:

- Systematic sampling on a grid to provide full coverage of the contaminated soils in the Building 812 area with an HPGe system, if a modified risk analysis shows present uranium levels are unacceptable (Section 4.2). This area corresponds to the area delineated in Figure 2-14 of the draft RI/FS.
- There remains a significant level of uncertainty at the site with the nature and extent of vertical contamination in the firing table and adjacent Quaternary alluvium at the site. Soil volumes to be remediated are dependent on the depth of chunks and fragments of uranium oxide that penetrated the subsurface. Additionally, the distribution of contamination with depth in the adjacent Qal is unknown. Since most of the preferred alternatives include removal of this material, screening data would be most appropriate in the area. The technical team recommends vertical screening with a Nal detector. This should be implemented as a screening tool; total counts should be monitored to determine the depth of DU contamination.

# 4.1.1 Systematic Surface Scan using *in situ* Gamma Spectrometry (HPGe)

High Purity Germanium (HPGe) spectral gamma detectors are widely accepted as the standard for lab based gamma spectroscopic analysis. *In situ* gamma spectroscopy uses the same basic instrumentation as gamma spectroscopy in the laboratory but the instrument is deployed from a platform or tripod in the field. The technique can be used to estimate activity concentrations directly in exposed soil surfaces at detection levels appropriate for the B812 Firing Table facility. Field of view is an important consideration for these systems, and can be controlled by the height of the instrument off the ground and/or the use of collimation (shielding).

HPGe systems have been used to support environmental decision making and closure within the DOE system. At the Fernald Site, extensive use of in situ HPGe was used to demonstrate Th-232, Ra-226 and U-238 activities met final remediation levels prior to collecting final certification samples. The HPGe system was used at the Mound site to screen for a variety of radionuclides. HPGe instrumentation and services are available commercially from a variety of vendors including ORTEC Instruments and Canberra. The vendor will determine the most appropriate field of view for the instrument and the best way to achieve this either by instrument height above the ground or smaller field of views. In order to meet the detection levels required at B812, count times on the order of 5-10 minutes per sampling location may be required to obtain the appropriate sensitivity. The roughness of the surface may also require smaller field of views. In addition, data collection will be complicated by the presence of steep topography in the area. Technicians will need to roped and suspended during data collection activities on the steep slopes. Similar studies on steep walls, albeit on a smaller scale, were successfully completed at both the Fernald and Miamisburg Closure Projects.

The survey design and DQOs should be documented in a detailed sampling and analysis plan that explicitly describes QA/QC protocols, which will include hot spot criteria and final remediation levels. In some cases, based on existing information and confirmatory surface scans, the conclusion may be that soil will meet cleanup criteria and no further remediation will be necessary. In such cases, additional characterization data actually would constitute the verification or final status survey, in which case sample numbers and locations would be driven by verification needs. The initial survey should be designed to insure DQOs will be sufficient to support site closure.

### 4.1.2 Chemical and Physical Characterization of DU

The goals of physical characterization of DU-bearing particles are to assess the probability of their entering the different exposure pathways and to obtain information to assist removal and size separation. The primary information necessary is the particle size distribution, whether the particles are discrete DU particles or agglomerated with soil particles, and the integrity of coarse particles. Magnetic properties may also be useful to determine whether large particles can be detected in soil by electromagnetic methods.

Particle size distribution may be done in two steps. Standard sieving methods can be used in the first step and total uranium analyzed in each size fraction. For

the silt to clay fraction, scanning electron microscopy (SEM) can be used to measure particle sizes (e.g. Danesi et al., 2003; Török et al., 2004). Observations can also be made on whether particles tend to be discrete DU particles or agglomerates with soil. Integrity of coarse particles can be evaluated with simple crushing tests that simulate the size separation process.

The goal of chemical characterization is to understand how available the DU is to dissolution by surface water and infiltration, as well as how bioavailable it is. The primary information necessary is the dominant phases DU occurs in, their leachability, and their bioavailability. In addition, it will be important to establish whether concentrations of the other metals of interest are correlated to uranium concentrations.

SEM with energy dispersive x-ray analysis (EDS or EDAX) can be used to identify the elements present in DU-bearing particles. Semi-quantitative element ratios can help identify specific phases.

Sequential extractions and wet chemical bioavailability tests can be used to assess the overall leachability in soil and bioavailability. Choy et al. (2006) used the sequential extraction procedure of Tessier (1973) to understand general leachability. Lind et al. (2009) used leaching with simulated gastric fluids to assess bioavailability. General understanding of oxidation state can be gained through oxidizing and reducing leaches.

Quantitative chemical analyses and oxidation states of uranium in DU-bearing particles can also be determined by x-ray microbeam techniques such as micro-XRF and XANES (e.g. Lind et al.; 2009).

A sufficient concentration of DU-bearing particles must exist for SEM or x-ray microbeam techniques to be effective. In soils with insufficient concentrations, DU-bearing particles can be concentrated with density separations, magnetic separation, or other common mineral separation techniques.

### 4.2 Modification of Risk Analysis

Presently, the soil clean-up levels are being driven by very conservative assumptions that are not reasonable exposure scenarios for human or ecological receptors at the Building 812 Operable Unit (see Section 3.3). A modified risk analysis, with reasonable exposure scenarios, should be evaluated by the LLNL staff and proposed to the regulators as part of the overall remedial strategy. This analysis would require additional characterization data on the form and solubility of uranium phases to properly evaluate ecological receptors, but one need not perform all the in situ HPGe measurements prior to the risk analysis. If the regulators concur with a modified risk analysis and the analysis indicates that much higher levels of uranium can remain in the slope soil, the HPGe measurements required for the slope soil may be scaled back or eliminated, and significant cost savings will be realized with the removal of less soil in the ensuing remedial actions.

### 4.3 Phased Technical Approach

The team believes that no single soil remedial alternative proposed in the RI/FS is likely to meet all of the remediation goals and respect sensitivity to endangered species and habitat destruction. Removing all of the slope soil with U-235 and U-238 concentrations above background will have a long-term negative impact on endangered species habitat. Furthermore, it is unlikely that treatment of the soil by soil washing will reduce uranium concentrations to background levels.

The team favors a phased approach to remediation that eliminates the principal threats to groundwater and removes soil hotspots with concentrations of U-235 and U-238 that pose unacceptable risk. A phased approach allows the remediation path to be driven by the results of each phase. This reduces the possibility of costly "surprises" such as failure of soil treatment and reduces the impact of remediation on endangered habitat.

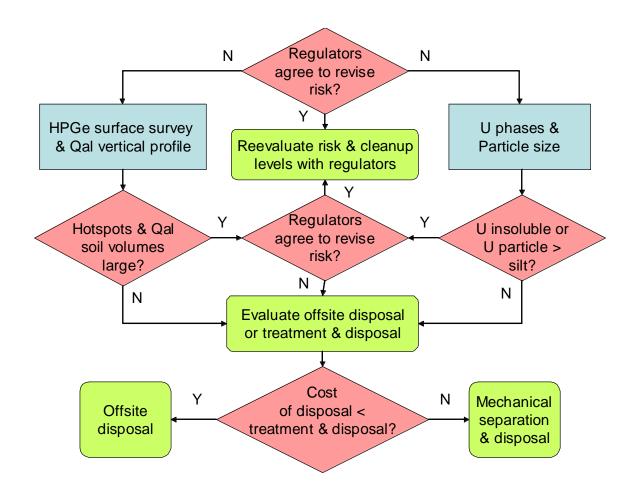
In general, a revised risk scenario should be negotiated with the regulators to determine if cleanup levels can be increased (Figure 3). If not, additional characterization activities should be implemented to evaluate the preferred remedial approach. Further characterization of the surface and subsurface distribution will refine the volume of soil requiring excavation. If the identified hotspots and Qal soil volumes are low, it may be cost effective to dispose of the soil offsite, whereas large volumes would imply very high cost and a need to reevaluate the risk and clean-up goals with the regulators. The form and solubility of the uranium will also play a role in the remedial decisions. If the uranium is soluble and comprised of fine particles, it is most cost effective to evaluate treatment and disposal at an offsite facility. When coarse, insoluble forms of uranium are present, the risk and clean-up levels should be renegotiated with the regulators. If either of the characterization paths indicates an unacceptable cost to dispose or treat the soil, then cleanup levels should be renegotiated with the regulators, if possible.

The general philosophy of Figure 3 is incorporated in the recommended phases described below:

### Phase I: Additional Site Characterization & Modified Risk Analysis

- Screening level characterization of the gross vertical distribution of contamination in the alluvium and firing table area to refine soil volumes.
- Characterization of the physical size and chemical form of DU in soils.

• Use of the physical and chemical characterization data to perform a modified risk analysis to determine the appropriate soil clean-up levels.



. Figure 3: Decision Tree for Characterization/Remediation Activities

### Phase II: HPGe Measurements & Removal of Contamination Hotspots

If the modified risk analysis indicates present uranium concentrations on slope soil are protective of human and ecological receptors, HPGe measurements may be unnecessary. However, if the clean-up levels are less than the measured uranium levels or the regulators require additional slope characterization data, HPGe measurements of the slopes should be performed to determine the spatial distribution of DU. This would consist of an entire site survey using HPGe detectors. Verification point sampling and a number of depth profiles will be done to construct contaminant concentration versus depth curves. This was described in detail in the previous section.

It is assumed that the area around the firing table itself and a portion of the QAL are the principal threats to groundwater. These and any hotspots identified by the HPGe measurements will be removed. The excavated soils will be size separated with the partition size determined by the Phase I characterization data. The contaminated fraction will be shipped off-site. The uncontaminated fraction will be re-used or disposed on-site.

### Phase III: Verification Sampling

Verification sampling and HPGe surveys of the soil excavation areas will be done to ensure excavation effectiveness.

### Phase IV: Final Risk Assessment

Human health, ecological risk and threat to groundwater will be re-assessed using the modified risk assumptions and post-remediation conditions attributed to soil removal actions.

### References

Bertsch, P.M and D.B. Hunter, 1994, Determination of Uranium Speciation in Contaminated Soils from the DOE Fernald Environmental Management Project Site by Micro-X-ray Absorption Spectroscopy, Savannah River Ecology Laboratory, Final Report submitted to FEMP.

Bethke, C.M., 2005, The Geochemist's Workbench® (geochemical modeling software), Release 6.0, University of Illinois.

Buck, E.C., N.R. Brown, N.L. Dietz, and J.C. Cunnane, 1994, Supporting Soil Remediation at Fernald by Electron Beam Methods, Argonne National Laboratory, Submitted to Waste Management 1994, Tucson, AZ.

Choy, C.C., G.P. Korfiatis, and X. Meng, 2006, Removal of depleted uranium from contaminated soils. Journal of Hazardous Materials 136, 53-60.

Cunnane, J.C., V.R. Gill, S.Y. Lee, D.E. Morris, M.D. Nickelson, D.L. Perry, and V.C. Tidwell, 1993, Uranium Soils Integrated Demonstration: Soil Characterization Project Report, FEMP/SUB-058, Final Report submitted to FEMP.

Danesi, P.R., A. Markowicz, E. Chinea-Cano, W. Burkart, B. Salbu, D. Donohue, F. Ruedenauer, M. Hedberg, S. Vogt, P. Zahradnik, and A. Ciurapinski, 2003, Depleted uranium particles in selected Kosovo samples. Journal of Environmental Radioactivity 64, 143-154.

Dermont, G., M. Bergeron, G. Mercier, M. Richer-Laflèche, 2008, Soil washing for metal removal: A review of physical/chemical technologies and field applications. Journal of Hazardous Materials 152, 1-31.

DOE Soil Decon Task Group, 1993, Removal of Uranium from Uranium-Contaminated Soils Phase I: Bench-Scale Testing, ORNL-6762, Oak Ridge National Laboratory.

Douthat, D.M., R.N. Stewart, and A.Q. Armstrong, 1995a, Fixed Capital Investments for the Uranium Soils Integrated Demonstration Soil Treatment Technologies, ORNL/TM-13004, Oak Ridge National Laboratory.ORNL-6882

Douthat, D.M., R.N. Stewart, and A.Q. Armstrong, 1995b, Operating and Life-Cycle Costs for Uranium-Contaminated Soil Treatment Technologies, ORNL-6882, Oak Ridge National Laboratory.

Domingo, J.L., Paternain, J.L., Llobet, J.M., Corbella, J., 1989a. The developmental toxicity of uranium in mice. Toxicology 55, 143–152.

Domingo, J.L., Ortega, A., Paternain, J.L., Corbella, J., 1989b. Evaluation of the perinatal and postnatal effects of uranium in mice upon oral administration. Archives of environmental Health 44 (6), 395–398.

Francis, C.W., A.J Mattus, L.L. Farr, M.P. Elless, and S.Y. Lee, 1993, Selective Leaching of Uranium from Uranium-Contaminated Soils: Progress Report 1, ORNL/TM-12177, Oak Ridge National Laboratory.

Kneff, D.W., G. Subbaraman, and R. J. Tuttle, 1992, Homogeneity Evaluation of Fernald Soils Prepared for Treatability Studies, ETEC/GEN-ZR-0018, Energy Technology Engineering Center, Rockwell International, with ORNL. Kneff, D.W., B.M. Oliver and G. Subbaraman, 1993, Updated Homogeneity Evaluation of Fernald Soils Prepared for Treatability Studies, ETEC/GEN-ZR-0018 Addendum, Energy Technology Engineering Center, Rockwell International, with ORNL.

Lee, S.Y., M. Elless, and F Hoffman, 1993, Solubility Measurement of Uranium in Uranium-Contaminated Soils, ORNL/TM-12401, Oak Ridge National Laboratory.

Lee, S.Y. and J.D. Marsh, 1992, Characterization of Uranium-Contaminated Soils from DOE Fernald Environmental Management Project Site: Results of Phase I Characterization, ORNL/Tm-11980, Oak Ridge National Laboratory.

Lind, O.C., B. Salbu, L. Skipperud, K. Janssens, J. Jaroszewicz, and W. De Nolf, 2009, Solid state speciation and potential bioavailability of depleted uranium particles from Kosovo and Kuwait. Journal of Environmental Radioactivity 100, 301-307.

Kulpa, J.P. and J.E. Hughes, 2001, Deployment of chemical extraction soil treatment on uranium contaminated soil. Proceedings of the Waste Management 2001 Conference, Tuscon AZ.

Llobet, J.M., Sirvent, J.J., Ortega, A., Domingo, J.L., 1991. Influence of chronic exposure to uranium on male reproduction in mice. Fundamental and Applied Toxicology: Official Journal of the Society of Toxicology 16 (4), 821–829.

Mann, M.J., 1999, Full-scale and pilot-scale soil washing. Journal of Hazardous Materials 66, 119-136.

Mason , C.F.V., W.R.J.R. Turney, , D.A. York, and C.J. Chisholm-Brause, 1994, Heap Leach Studies on the Removal of Uranium from Soil, II, LA-UR-94-3689, Los Alamos National Laboratory.

Oliver, I.W., M.C. Graham, A.B. MacKenzie, R.M. Ellam, and J.G. Farmer, 2008a, Distribution and partitioning of depleted uranium (DU) in soils at weapons test ranges – Investigations combining the BCR extraction scheme and isotopic analysis. Chemospere 72, 932-939.

Oliver, I.W., M.C. Graham, A.B. MacKenzie, R.M. Ellam, and J.G. Farmer, 2008b, Depleted uranium mobility across a weapons testing site: Isotopic investigation of porewater, earthworms, and soils. Environmental Science & Technology 42, 9158-9164.

Paternain, J.L., Domingo, J.L., Ortega, A., Llobet, J.M., 1989. The effects of uranium on reproduction, gestation, and postnatal survival in mice. Ecotoxicology and Environmental Safety 17 (3), 291–296.

Salbu, B., K. Janssens, O.C. Lind, K. Proost, L. Gijsels, and P.R. Danesi, 2005, Oxidation states of uranium in depleted uranium particles from Kuwait. Journal of Environmental Radioactivity 78, 125-135.

Schilk A.J., R.W. Perkins, K.H. Abel, and R.L. Brodzinski, 1993, Surface and Subsurface Characterization of Uranium Contamination at the Fernald Environmental Management Site, PNL-8617/UC-606, Pacific Northwest Laboratory.

Sheppard, S.C. and Evenden, W.G., 1992. Bioavailability indices for uranium: effect of concentration in eleven soils. Archives of Environmental Contamination and Toxicology 23, 117–124.

Török, S., J. Osán, L. Vincze, S. Kurunczi, G. Tamborini, and M. Betti, 2004, Characterization and speciation of depleted uranium in individual soil particles using microanalytical methods. Spectrochimica Acta Part B 59, 689-699.

Tessier, A., P.G.C. Campbell, and M. Bisson, 1979, Sequential extraction procedure for the speciation of particulate trace metals. Analytical Chemistry 51, 844.851.

Turney, W.R.J.R., D.A. York, C.F.V. Mason, C.J. Chisholm-Brause, D.C. Dander, P.A. Longmire, D.E. Morris, R.K. Strait, and J.S. Brewer, 1994, Heap Leach Studies on the Removal of Uranium from Soil, LA-UR-94-1361, Los Alamos National Laboratory.

### Appendix A: Technical Assistance Request

# Technical Assistance Request for DOE Independent Technical Review Team

### Evaluation of Technical Options for Soil Remediation at the Building 812 Firing Table, Lawrence Livermore National Laboratory Site 300

The Department of Energy, Livermore Site Office (LSO), is conducting remediation at the Lawrence Livermore National Laboratory (LLNL) Site 300 experimental test facility under CERCLA. A draft Remedial Investigation/Feasibility Study (RI/FS) has been prepared for the Building 812 Operable Unit (OU) in accordance with the terms of the Site 300 Federal Facility Agreement between DOE, the U.S. Environmental Protection Agency (EPA), and California Department of Toxic Substances Control (DTSC), and the California Regional Water Quality Control Board.

The RI/FS forms the basis for evaluating and selecting alternative technologies for remediation of contaminants at the Building 812 OU. The purpose of the RI/FS is to identify the nature and extent of contamination in the Building 812 OU and evaluate impacts to human and ecological receptors that could be exposed to contaminated environmental media and to develop and evaluate alternatives for remediation action in accordance with CERCLA.

The remedial alternatives in the draft RI/FS include:

- 5. No Action.
- 6. Excavate shallow subsurface soil at the Building 813 Firing Table up to 6 feet and all rippable surface soil with depleted uranium and metal concentrations exceeding background and dispose offsite at a permitted landfill.
- 7. Excavate and treat shallow subsurface soil at the Building 812 Firing Table up to 6 feet and all rippable surface soil with depleted uranium and metal concentrations exceeding background using soil washing; replace treated soil.
- 8. Excavate and solidify, stabilize, and consolidate shallow subsurface soil at the Building 812 Firing Table up to 6 feet and all rippable surface soil with depleted uranium and metal concentrations exceeding background.
- Excavate shallow subsurface soil at the Building 812 Firing Table up to 6 feet and all rippable surface soil with depleted uranium and metal concentrations exceeding background and dispose at an onsite constructed landfill.

The EPA and DTSC comments on the draft RI/FS requested additional research and that a field-scale pilot test be conducted to evaluate the potential effectiveness of the soil washing technology, particularly in fine-grained soil. Because soil washing appears to currently be the regulator's preferred alternative, DOE concurred that conducting a treatability study would reduce uncertainties associated with this technology and allow for a better understanding of: (1) the limitations of the soil washing technology for site-specific conditions (e.g., soil clay content, site contaminants), (2) cost implications, and (3) the possible need for coupling soil washing with a second technology. DOE/LLNL is currently developing work scope, cost, and a schedule for a soil washing bench scale and field scale treatability study.

The tasking for the DOE Independent Technical Review Team is to:

- Review the draft Building 812 RI/FS with emphasis on the soil remediation options presented in Section 3 and Appendix E-2.
- Review case studies and lessons learned from other soil washing projects to assess feasibility of soil washing at the Building 812 Firing Table.
  Particularly interested in degree of success with uranium contaminated soil.
- Determine other feasible technologies/options available for remediation of contaminated soil at Building 812 Firing Table.
- Provide a briefing and report to DOE and LLNL that summarizes the team's evaluation and makes recommendations on options for soil remediation at the firing table.

The Independent Technical Review Team should plan for a two-day visit to the Livermore Site Office. During the site visit, DOE/LSO and LLNL will present information on Site 300 and Building 812 remediation activities, and will take the team out to Site 300.