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Review of Analytes of Concern & Sample Methods for Closure of DOE High Level Waste Storage Tanks

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Abstract - *Sampling residual waste after tank cleaning and analysis for analytes of concern to support closure and cleaning targets of large underground tanks used for storage of legacy high level radioactive waste (HLW) at Department of Energy (DOE) sites has been underway since about 1995. The DOE Tanks Focus Area (TFA) has been working with DOE tank sites to develop new sampling methods for assessment of residual waste inventories. This paper discusses regulatory analytes of concern, sampling plans, and sampling methods that support closure and cleaning target activities for large storage tanks at the Hanford Site, the Savannah River Site (SRS), the Idaho National Engineering & Environmental Laboratory (INEEL), and the West Valley Demonstration Project (WVDP).*

I. INTRODUCTION

Sampling residual waste after tank cleaning and analysis for analytes of concern to support closure of large underground tanks used for storage of legacy HLW at DOE sites has been underway since about 1995. The DOE sites that have large HLW tanks ranging from 1.1 to 4.9 x 10^6 liters (0.3 – 1.3 million gallons) in capacity are the Hanford Site, SRS, INEEL[a], and WVDP[b]. The DOE Tanks Focus Area (TFA) has been working with the tank sites to develop new sampling methods for assessment of residual waste inventories and holding workshops to discuss tank closure issues. The analytes of concern and some of the sampling methods developed recently are discussed below.

II. REGULARTORY ANALYTES OF CONCERN FOR CLOSURE OF HLW TANKS

Two inherent assumptions in the closure of large underground HLW tanks is that from a cost benefit standpoint it is not practical to remove all the waste and that the remaining residual waste inventory must be characterized to support the closure performance assessments (PA). Regulatory radionuclide analytes of concern for closure of HLW tanks are given in DOE Order 435.1-1 and the Code of Federal Regulations 10 CFR 61.55. The radionuclides fall into four categories:

1. Long-lived: ^{14}C, ^{59}Ni, ^{94}Nb, ^{99}Tc, ^{129}I, ^{241}Pu, and ^{242}Cm.
2. Short-lived: ^{3}H, ^{60}Co, ^{63}Ni, ^{90}Sr, and ^{137}Cs.
3. Transuranic (TRU) radionuclides with half lives > 5 years: Pu, Np, Am, and Cu isotopes.
4. Others of interest: ^{79}Se, ^{126}Sn, and ^{237}Np.

Because most legacy HLW is also classified as mixed hazardous waste, some analytes of concern defined by the Resource Conservation and Recovery Act (RCRA) in 40 CFR 261.24 may have to be characterized. There are eight elements (i.e., Ag, As, Ba, Cd, Cr, Hg, Pb, and Se) and 32 organic compounds, which are defined as toxicity characteristic (TC). In addition, several hundred organic and inorganic compounds are defined as listed hazardous constituents in Appendix VIII of 40 CFR 261. Depending on process history, analytical data, and negotiations between the DOE and regulatory agencies, the RCRA analytes of concern will vary from tank to tank and site to site.

The PA for immobilizing and leaving residual waste in place will have to show compliance of potential future contaminant releases with EPA Drinking Water Standards per 40 CFR 141/142/143. The standards list maximum concentration limits for 168 radionuclides, elements (i.e., Sb, As, Ba, Be, Cd, Cr, Cu, Pb, Hg, Se, and Tl), and anions (i.e., CN^-, F^-, NO_3^-, and NO_2^-). Based on the above DOE, RCRA, and EPA regulatory requirements, the analytes of concern are summarized in Table I, which indicates a high degree of commonality among the sites.

[a] Since 1996, the INEEL tanks are no longer use for storage of HLW.

[b] Under the WVDP Act of 1980, DOE is responsible for treating and disposing of the radioactive waste in the HLW tanks, but the land and facilities of the WVDP belong to the state of New York.

TABLE I

Analytes of Concern in DOE HLW Tank Closure Activities

Analyte Group	INEEL Analytes[1]	SRS Analytes[2]	Hanford Analytes[3]
Gamma Emitters	^{60}Co, ^{94}Nb, ^{137}Cs, $^{154/155}$Eu	^{60}Co, ^{95}Zr, ^{137}Cs, $^{154/155}$Eu, ^{241}Am	^{60}Co, ^{137}Cs, ^{241}Am
Beta Emitters	^{3}H, ^{90}Sr, ^{99}Tc, ^{129}I	^{3}H, ^{90}Sr, ^{99}Tc, ^{79}Se	^{90}Sr, ^{99}Tc
Uranium & TRU Isotopes	$^{234/235/236/238}$U, ^{237}Np, $^{238/239/240/241}$Pu, ^{241}Am, ^{244}Cm	$^{238/239/241}$Pu, ^{241}Am, ^{244}Cm	$^{239/240}$Pu, ^{241}Am
TC Elements	Ag, As, Ba, Cd, Cr, Hg, Pb, Se	Ag, As, Ba, Cd, Cr, Hg, Pb, Se	Ag, As, Ba, Cd, Cr, Pb
Other Elements	Al, Be, Ca, Co, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Sb, Tl, V, Zn	Al, B, Ca, Ce, Co, Cu, Fe, K, La, Li, Mg, Mn, Mo, Na, Ni, P, Si, Sn, Sr, Ti, V, Zn, Zr	Al, Bi, Ca, Ce, Co, Cu, K, La, Mg, Mn, Mo, Na, Nd, Ni, P, S, Si, Sm, Sr, Ti, Tl, U, V, Zn, Zr
Listed Organic Compounds	24 organic compounds listed in closure plan[4]	Analytes not specified	Analytes not specified
Anions	Cl^-, F^-, PO_4^{-3}, NO_3^-, SO_4^{-2}	Cl^-, F^-, CO_3^{-2}, $C_2O_4^{-2}$, NO_3^-, NO_2^-, SO_4^{-2}	Cl^-, F^-, PO_4^{-3}, NO_3^-, NO_2^-, SO_4^{-2}

II. SAMPLE PLANS AND METHODS FOR RESIDUAL WASTE IN HLW TANKS

The large underground HLW storage tanks at DOE facilities with storage capacities ranging from 1.1 to 4.9 x 10^6 liters have floor diameters between 15 - 26 meters and wall heights between 6 - 10 meters. The floor and wall of the larger tanks have about 530 and 820 m^2 of surface area, respectively. Sampling tools for residual waste solids typically take a 1.3 - 5.0 cm diameter sample, which would sample an area of 1.2 - 20 x 10^{-4} m^2. The cross-sectional area of the sample would represent 0.16 - 4.0 part per million of the area of the floor or wall per sample. The DOE tank sites have not developed or adopted a consistent set of guidelines, which govern the number of samples required to achieve representative sampling in the large HLW tanks. In general, tank sampling will be highly constrained by costs, available sampling tools and deployment devices, allowed worker exposure limits, lack of access into the tank, and impediments due to internal tank structure. Consequently, the sampling plans are guided more by these constraints and DOE negotiations with their regulatory agencies rather than the number of samples that would be required based on statistical models. To date, methods to estimate residual waste volume rely on visual inspections, videotape, still photographs, and dimensions of known reference points based on as-built drawings. Examples of number of samples planned or taken, sampling tools used or planned supporting tank closure activities are given below.

II.A. Tanks WM-182 and WM-183 at the INEEL

To support a scheduled closure, beginning in 2003, of two 1.1 x 10^6 liter underground storage tanks at INEEL, acquisition of five residual waste heel samples from each tank is planned[1] after tank washing and heel retrieval campaigns. A light duty utility arm (LDUA) and sampling end effector (EE) developed under TFA sponsorship are planned for use in the heel sampling campaign. Four

LDUAs, with similar specifications, were fabricated for use at the Hanford Site, INEEL, and the Oak Ridge National Laboratory (ORNL) and a generic description of the robotic arms can be found in a DOE Innovative Technology Summary Report[5]. The INEEL LDUA has an articulated arm with seven degrees of freedom and an off-riser axis reach of 4 meters when fully extended. The arm is positioned under riser via a vertical position mast with a 14-meter reach. Risers with a diameter of 30.5 cm or greater are required for adequate LDUA clearance. The major components of the LDUA are shown in Figure 1 which are: a) the LDUA robotic system with the articulated arm down in the tank, b) a LDUA utility trailer, c) an end effector utility trailer, d) an operator control trailer, e) an auxiliary robotic end effector exchange system for above riser operations, and f) a sample transfer cart.

The sampling EE can sample up to 1200 cm^3 of liquid and soft sludge and contains a light, video camera, gamma radiation detector, two sample pumps, and a detachable sample chamber. Two views of the sampling EE and some of the components are shown in Figure 2. The housing, which seals the upper cylindrical compartment, is not shown to allow illustration of internal components. A detailed description of the first generation sampling EE can be found in a DOE Innovative Technology Summary Report[6]. The sampling EE shown in Figure 2 has been slightly modified to improve performance based on lessons learned in prior heel sampling campaigns at the INEEL.

To ensure random sampling within reach of the LDUA (i.e., up to a 4-meter radius circle), the tank floor area was divided up into 930 cm^2 grids and a random number table used to select five of these grids for sample collection. The sample plan[1] assumes that agitation of the heel during the decontamination activities will result in the solids being sufficiently mixed such that five samples will meet the data quality objectives for representative sampling. A manual sampler, currently under development, is also planned for deployment under riser during the tank washing campaign and it is planned to compare the analyte composition of the manual and LDUA sample methods. If the sample results can be shown to be comparable, it is anticipated that the manual under-riser sampling method can replace the LDUA off-riser random sampling method in future tank activities as a cost savings measure.

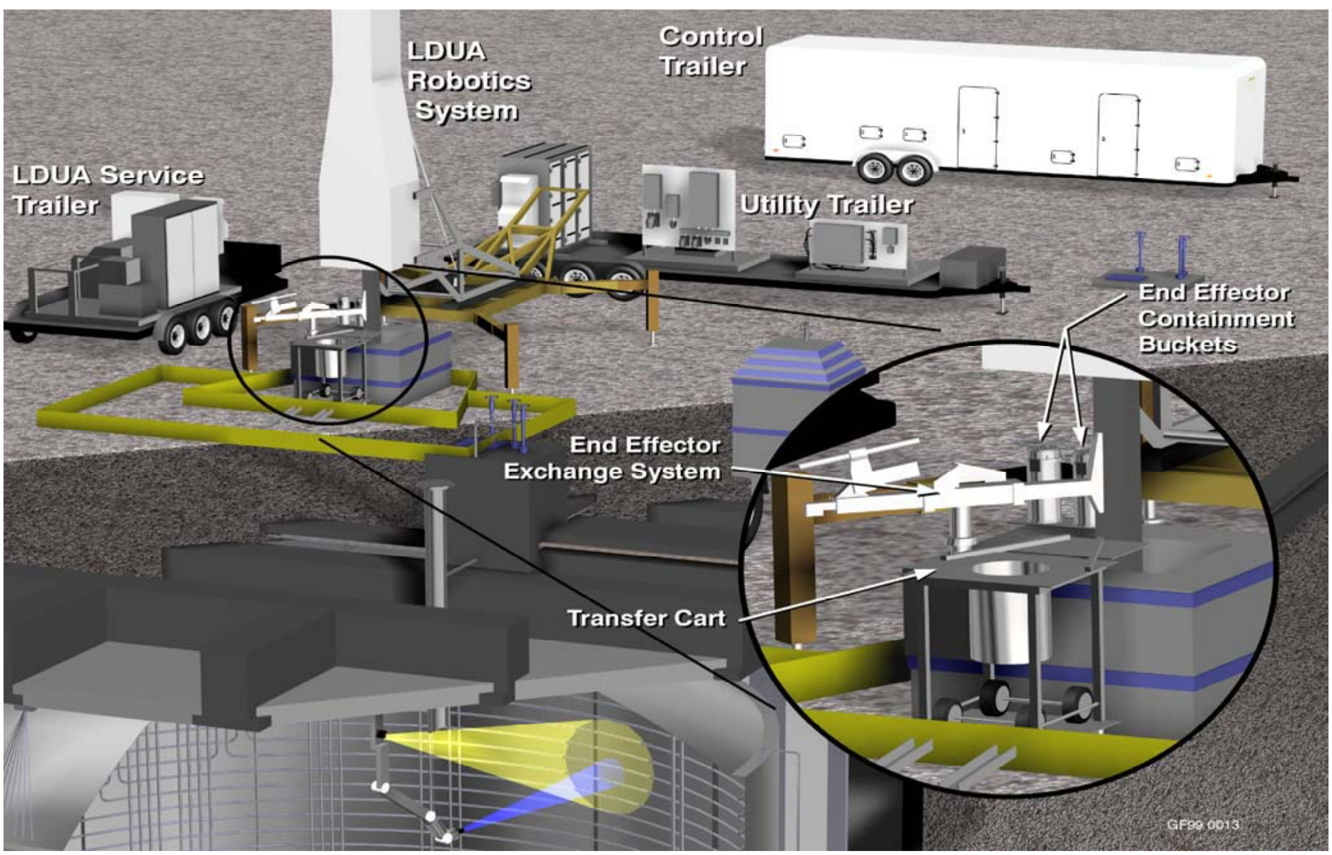

Figure 1. Major components of the LDUA for inspecting and sampling DOE underground storage tanks.

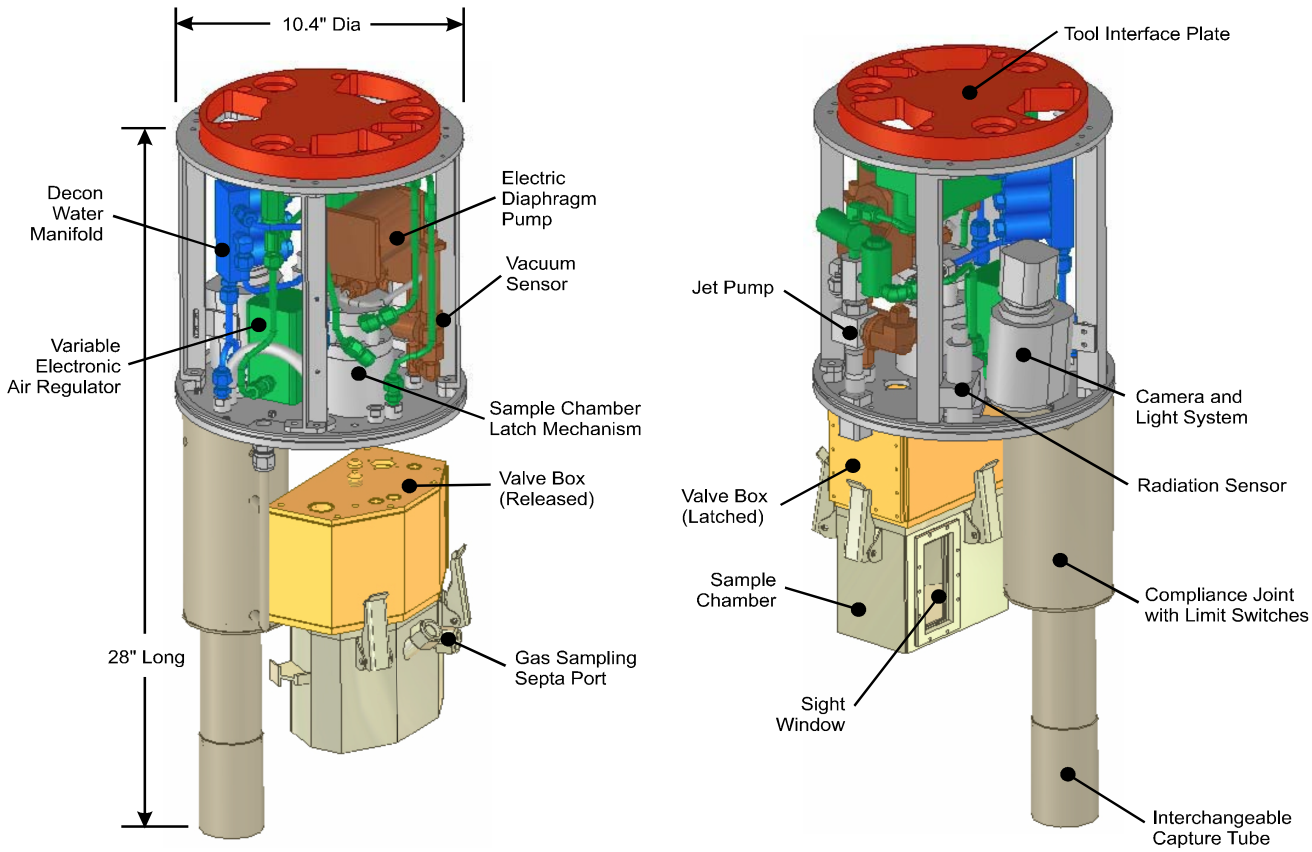

Figure 2. Two views of the INEEL LDUA sampling EE without the protective housing on the upper portion.

II.B. Tank 241-AX-104 at the Hanford Site

Under the TFA Hanford Tank Initiative, the plan[3] was to take up to 12 samples from the residual waste contained in a 3.8 x 10^6 liter underground Hanford storage tank 241-AX-104. A truck mounted LDUA with the same design features and support equipment as the skid mounted INEEL LDUA would have enabled partial sampling access to the floor, walls, and dome areas. A pneumatically actuated clam-shell sampler with a 50 cm^3 capacity, a quick release mechanism for change out of the sample chamber, light, and video camera was designed[7] for sampling dried residue. The Hanford LDUA sampling EE is shown in Figure 3.

The sample and analysis plan[3] indicated that after eight samples, increasing the number of samples would not significantly improve the percentage of the number of samples within a specified confidence limit (i.e, with regards to variability in the observed concentration of a given analyte).

Figure 3. Sampling EE for residual waste in Hanford tanks.

II. C. Tank 19 F at the SRS

Tank 19 F at the SRS is a 4.2 x 10^6 liter capacity tank scheduled for closure in 2003. The sampling plan[2] indicated up to five solid samples would be taken after the final cleaning campaign to assess the residual waste inventory. The waste samples were obtained with the manually deployed device developed by SRS called a vial snapper grab sampler shown in Figure 4. The sampling jaws are pneumatically actuated and the closing force is remotely controlled. Quick disconnect pins are used to remove the sample jaws with the sample and replace them with clean sample jaws. The detached jaws and sample are held together by placing them in a sample bottle. Sample jaws of different lengths, volume, and vertical or horizontal orientation can be used. Typical jaw sample volumes have been 50 - 150 cm^3. The sampler is lowered into the tank using a mast assembly of extension pipes. SRS has developed and deployed other manual sample designs for soft and hard sludges, slurries, liquids, and tank wall sampling. A detailed description of these manual sampling devices can be found in the proceedings of a recent American Nuclear Society Topical Meeting[8].

Figure 4. Pneumatically actuated vial snapper grab sampler.

II.D. Tank 8D-2 at the WVDP

The TFA has been supporting WVDP activities to characterize residual waste radionuclides in a 2.8 x 10^6 liter underground HLW tank that stored raffinate waste from commercial fuel reprocessing. The residual waste inventory is being characterized to determine if WVDP has met waste retrieval and cleaning targets agreed to with the U.S. Nuclear Regulatory Commission. Analytes of concern include most of the radionuclides listed in Table 1 and in particular the residual waste TRUs. Characterization activities have included deploying a burnishing sampler developed by the ORNL[9] via TFA support. The sampler shown in Figure 5 was used to collect over 50 solid samples[10] from the wall and support structure of tank 8D-2. The sampler was deployed using a remotely operated tool delivery system called a Mast, which provides off-riser access to a large area of the tank. The sampler takes a wall scrape sample from a metal surface using an air-driven flat-cut milling machine bit (1.25 cm in diameter and 0.076 cm deep). The shallow depth is designed to provide sampling without potential damage to the tank wall. A spring-loaded shroud (not shown in Figure 5) provides a sufficient seal between the wall and the sample head so that solids are transported via a vacuum through a hole below the bit into a collection chamber with a small HEPA filter.

After sampling, the burnishing sampler is retrieved from the tank via the Mast tool delivery system, the sample housing remotely removed, and transported to a laboratory. Each sample head is use only once. The typical sample size is about 0.5 g of solids. The samples obtained are analyzed to establish the radionuclide sample concentration per unit area and to make an estimate of the tank wall inventory via extrapolation to the total surface area in the region of the tank sampled. Lessons learned in hot deployment of the burnishing sampler are published elsewhere[10].

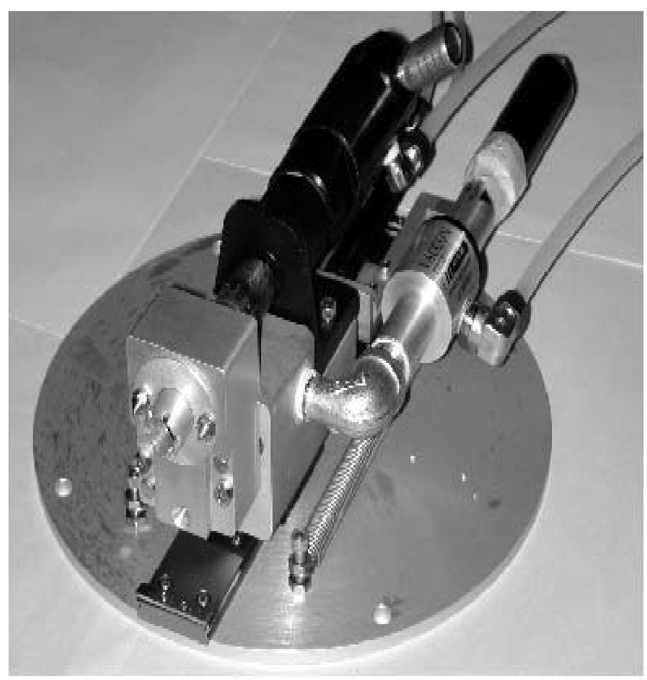

Figure 5. Wall burnishing sampler used in WVDP tank.

III. CONCLUSIONS

The regulatory radionuclides of concern with regards to cleaning tanks in preparation for tank closure are fairly consistent among the DOE sites. The inorganic and organic analytes of concern vary widely among the DOE sites as do the sample tools, sample volumes, and number of samples collected. This survey indicates a range of 5-50 samples were planned or taken to support tank closure and cleaning target goals. Over the last eight years a great deal of progress has been made in developing a wide variety of sampling tools for under-riser manual sampling and off-riser remote sampling via robotic deployment devices.

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