Characterization Modeling to Support the Hanford 618-10 and 618-11 Burial Grounds Remediation Design Solution: Two Differing Approaches with Similar Results

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ABSTRACT

Two different approaches were applied to characterization modeling of the waste in the 618-10 and 618-11 burial grounds. The results were compared and it was found that the independent approaches validate each other. The 618-10 and 618-11 burial grounds, located on the Hanford site in Washington state, received primarily radioactive laboratory waste in the 1950's and 60's; however, disposal records from burial activities have since been destroyed. North Wind Inc. (NWI) is completing a technology demonstration project, funded by DOE Headquarters to develop methodology for remediation of the vertical pipe units and develop supporting documentation. Washington Closure Hanford (WCH) is developing a design solution for remediation of the 618-10 and 618-11 burial grounds, including the development of a characterization model and estimates of radioactivity and waste volumes present. Each company independently developed their characterization models and radionuclide inventories, using a different methodology; however, the results of each model revealed only a two to five percent difference, which is significant given the complexity of the waste matrices, the high dose rates of the waste when disposed, and relatively high levels of transuranic radionuclides projected.

INTRODUCTION

The United States Department of Energy (DOE) contracted North Wind Inc. (NWI) to develop documentation in support of the *In-Situ Transuranic (TRU) Waste Delineation and Waste Removal at Hanford 618-10 and 618-11 Burial Grounds* Project. As part of this project, NWI is developing a methodology for remediation of the vertical pipe units which are located in the burial grounds. Washington Closure Hanford (WCH), the site contractor, is developing a comprehensive design solution for remediation of the 618-10 and 618-11 burial grounds, including the development of a characterization model and estimates of radioactivity and waste volumes present.

Background

The 618-10 and 618-11 burial sites (618-10 & 11) were operated from 1954 to 1967. The 618-10 burial site operated from March 1954 to September 1963. The 618-11 burial site operated from March 1962 to December 31, 1967. Records indicate that in 1965, control of these sites was transferred from General Electric to Battelle Pacific Northwest Laboratory (PNL).

Documentation also indicates that characterization records of these two burial sites were destroyed.

Higher dose rate items were generally transported to 618-10 & 11 in bottom-opening shielded casks and placed in either vertical pipe units (VPUs) or caissons (618-11 only) by remotely opening a trap door in the bottom of the cask and allowing the waste to fall to the bottom of the VPU or caisson. VPUs are constructed from five 208-liter (55-gallon) drums, ends removed, welded end-to-end, and buried to create an empty shaft with both ends open (Figure 1). Remaining waste was disposed in trenches. Some high dose rate waste was disposed in trenches by either loading cardboard boxes of waste into shielded load luggers or centering small quantities of waste in a drum and pouring either concrete or a combination of concrete and lead around the waste, some of which was containerized liquid waste. This resulted in a significant dose rate reduction for personnel disposing of the waste when generated; however, could be of concern when the waste is exhumed.

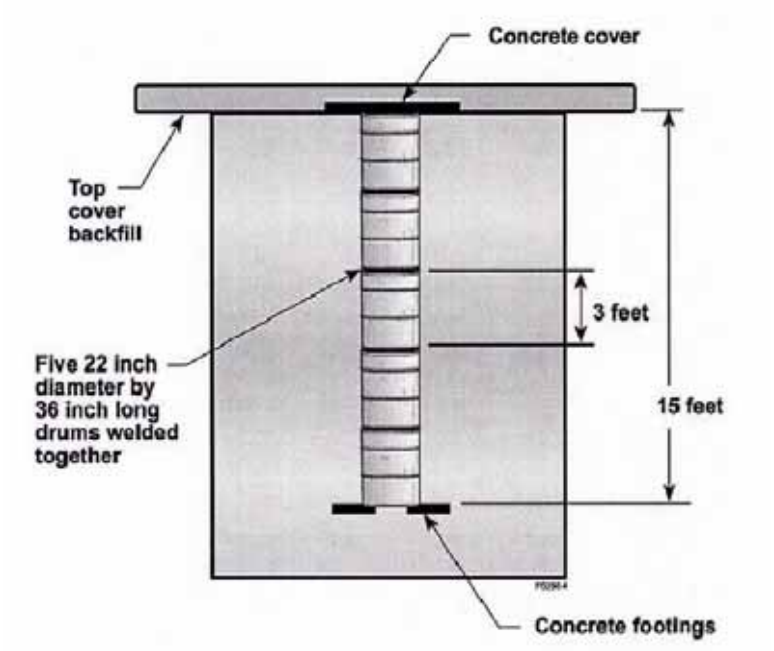

Fig. 1. Sketch of a Vertical Pipe Unit (VPU) used for disposal at the 618-10 and 618-11 Burial Grounds.

Waste Materials

The waste material was generated during laboratory examinations and studies, including analyses of fuel reactor samples, characterization of the chemical and physical properties of immobilized forms of plutonium, and analysis of ruptured reactor fuel. [1, 2, 3] These analyses, performed in glove boxes, fume hoods and hot cells, used a wide variety of electrochemical, spectrophotometric, and physical tests that generated primarily inorganic (e.g., aluminum- and iron-based metal, glass, ceramics, and asbestos) and organic debris (e.g., plastic, rubber, paper, cloth, wood) waste materials. Specific waste items may include wipes, towels, protective clothing, cardboard, metal cans, High Efficiency Particulate Air (HEPA) filters, stainless steel tubing, plastic pipe, lead (bricks and sheeting), polyethylene bottles, failed machinery, used lab

ware (beakers, pipettes, vials, and tubing), gloves, lab equipment (balances, drying ovens, heating mantles, pumps and reaction vessels), thermometers, concrete, soil, plumbing fixtures, and tools (screw drivers, wrenches, and shears). [3] Some drums disposed in trenches contain oil. Also included are sample residues from fuel pellets, ruptured fuel elements, ceramics and grouted plutonium in cans. [3]

The radiological inventory includes uranium oxides, fission products, and plutonium. In most cases, plutonium will be found with various fission products, but in some of the generating facilities, separation of various isotopes took place, creating isolated streams of plutonium, promethium, cesium, curium, strontium, and americium. [3]

NORTH WIND APPROACH

As part of NWI's technology development, it was necessary to establish a firm understanding of the waste material that would be encountered in the burial grounds. A survey of hundreds of historical documents was performed as well as anecdotal evidence from workers who were familiar with the processes during that time period. Although no direct characterization materials were available, a body of evidence existed that could be used to assemble reasonable scenarios from which radiological characteristics could be derived. The evidence included intermittent disposal records, operational radiological limits, fuel composition data, laboratory practices, and survey records. This body of evidence was paired with ORIGEN2 data and MicroShield to determine reasonable expectations for present-day radiological field conditions in the burial grounds.

In the few years that 618-10 & 11 were in operation, waste disposal practices evolved, becoming more conservative and protective of workers. A worst case scenario was built based on the waste in 618-10, the older of the two burial grounds. From disposal records, it was determined that Building 327 was the facility that contributed to the majority of waste in the 618-10 burial ground VPUs. The characterization model developed by North Wind assumed that the waste originated in the 327 Building.

Methodology and Analysis

The 327 building performed destructive and non-destructive analyses on fuel elements from the production reactors in Hanford's 100 Area. The fuel elements were sectioned and dissolved, but were not separated or specific isotopes concentrated. The isotopic distribution from waste originating in the 327 building should reflect the isotopic distribution in the fuel that was being examined. The ORIGEN program, a standard code developed by Oak Ridge National Laboratory, was used to determine an approximate isotopic distribution and an estimate for the volume of TRU waste from Building 327 after 40 years of decay.

Shipping data does not provide adequate information to directly determine the volume or activity of waste disposed at 618-10. Many records have been lost or discarded. However, a fair amount of data exists that can help build a foundation for reasonable estimates. Data that was collected to develop an estimate includes:

- Fuel composition data

- Fuel element burn-up
- Time from reactor to disposal
- Activity limits on waste
- Survey records of exposure to personnel
- Survey records of exposure from waste
- Waste volumes and weights allowed in packages
- General waste management practices

Additional data from 300 Area activities was pursued. This data includes records from the radioactive liquid waste system (RLWS), special nuclear material (SNM) inventory, safeguards and securities, and other recorded information related to waste generation in the 327 building. However, according to a number of sources interviewed, with access to this type of information, the records no longer exist, cannot be found, or are still classified.

Data from 100 Area fuel ruptures was compiled and analyzed to determine the average isotopic content of the material examined in the 327 building. Forty-two records from 1948 to 1964 of 873 ruptured elements were examined and the data averaged. At that time, uranium fuel was used in the reactors. As described in various historical documents, the reactor power steadily increased from 1948 through 1964. The data from the records showed that the power increased from less than 400 Megawatt days (MWD) of burn-up in the reactor to about 1,300 MWD of burn-up (Figure 2) [4]. The power selected for the ORIGEN run based on this information was 1,000 MWD of burn-up.

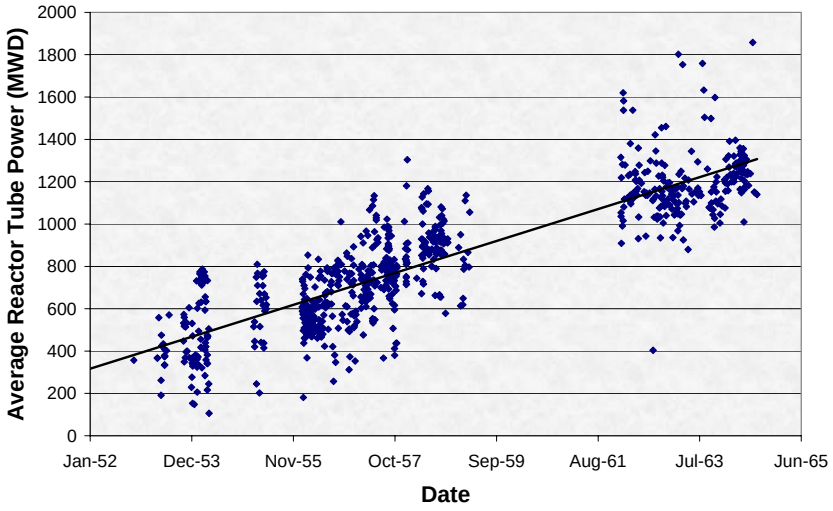

Fig. 2. Reactor power data from fuel rupture reports during the period of interest [4]

According to fuel examination reports, the fuel elements were received in the lab as soon as a few days after being discharged from the reactor. [5] Often, examination took place immediately, but wet storage was also available for holding fuel elements until examination could be performed. After examination, fuel was placed in storage and could remain there for more than a year before being sent to reprocessing. It is not clear how much time expired between when a fuel element was discharged from the reactor and its contaminated waste

disposed. A one-year duration was chosen as a conservative estimate, allowing time for considerable decay of short-lived isotopes.

An ORIGEN run was performed based on an initial mass of 1 kg uranium exposed to 1,000 MWD of burn-up in a reactor and decayed to one year and forty years. The one-year decay data is used for the activity at disposal, the forty-year decay data is used for the activity now, and the TRU isotopes from the forty-year decay data is used to estimate the TRU concentration now.

In order to use this data, the amount of the waste in a VPU must be determined. Monthly waste disposal records from 1960 to 1962 provide quantities of milk pails and juice cans disposed to the VPUs. [6] This information was averaged to provide an approximate disposal rate of 249 liters (8.8 ft^3) of intermediate- and high-level waste per month. Based on these records, 96% of the waste volume was intermediate-level and 4% was high-level. To determine the number of VPUs that were filled each month, the number of months that the burial ground was open was compared to the number of VPUs. The total length of time that the 618-10 burial ground VPUs were used was from January 1955 to September 1963, which is equivalent to 105 months. There are 94 VPUs in the burial ground, which means that approximately one VPU was filled each month or about 249 liters (8.8 ft^3) of waste was disposed to each VPU. The VPUs made from 208-liter (55-gal) drums have a total capacity of 1039 liters (36.7 ft^3), which made them roughly 24% full of waste and 76% full of backfill materials such as soil and concrete.

A final step to determine the TRU concentration is to include the total estimated mass of waste in the final waste package. It is assumed that the VPUs will be retrieved using a patent-pending over-coring method with a 109-cm (43-inch) diameter, 8.5-meter (28-foot) steel casing filled with 7.3 meters (24 feet) of soil and waste. The entire retrieved and enclosed VPU waste form would weigh about 15,400 kg (34,000 lbs).

In summary, the following assumptions were used to generate the estimated TRU concentrations below:

- Fuel segmented in 327 laboratory was uranium fuel before being exposed in the reactor
- Fuel burn-up was 1,000 MWD
- Fuel decayed for 1 year before disposal
- Fuel decayed 40 years between disposal and remediation
- 1 VPU was filled per month
- 249 liters (8.8 ft^3) of waste was disposed into a VPU (~24% full)
- 96% of the waste volume in a VPU was intermediate-level waste, 4% high-level
- Final waste package weighs 15,400 kg (34,000 lbs)

The activity of the waste is also necessary to determine the TRU concentration. A range of assumptions based on process knowledge were used to establish what a reasonable activity might have been.

North Wind performed case studies using the standard ORIGEN code and varying input parameters in order to develop a reasonable range of TRU concentration estimates for VPUs in 618-10. The case studies were based on information found in historical documents ranging from

procedures to survey records. The decision point for TRU materials is a concentration of 3.7 kBq/gm (100 nCi/gm) of TRU isotopes with a half-life greater than 20 years.

The first case considered was a "best-case scenario". Waste handling and disposal records indicate that "low-level waste" was defined as waste less than 10 Ci/ft^3, intermediate-level waste was from 1.31 GBq/L (10 Ci/ft^3) to 1307 Gbq/L (1000 Ci/ft^3), and "high-level waste" was greater than 1307 Gbq/L (1000 Ci/ft^3). Intermediate-level and "high-level" waste was disposed into VPUs; "low-level" waste was disposed into trenches. Under these conditions, given the previously-stated assumptions, the best-case scenario would be that all 96% of the waste that was intermediate-level was 1.31 GBq/L (10 Ci/ft^3) and the high-level portion was 1307 Gbq/L (1000 Ci/ft^3). Using this ratio and these activities, the inventory in one VPU at disposal would have been about 1.56E+04 GBq (421 Ci). The ORIGEN data shows that the activity will have decreased by a factor of about 28 over the intervening 40 years. It also indicates that the TRU isotopes consist of about 1.2 percent of the total remaining curies, resulting in a final TRU isotope inventory of about 6.29 GBq (0.17 Ci). Factoring in the total mass of the waste package, the final TRU concentration becomes 0.407 kBq/gm (11 nCi/gm). The third case is similar to this case, but uses a worst-case scenario projection at the upper bounds of the definitions of intermediate- and high-level waste, 1176 and 3920 GBq/L (900 and 3000 Ci/ft^3) respectively, resulting in a final TRU concentration of 8.51 kBq/gm (230 nCi/gm).

Case study #2 is based on documentation that states that there was less than 1 kg of fuel material contamination on the waste in each milk pail disposed to a VPU. As an example, 500 gm was used as a reasonable mid-range value. This mass of fuel material contamination results in 2.6E+03 (70 Ci) of initial activity per milk pail. Based on average disposal rates, a VPU would initially have about 2500 Ci, which decays and scales to 2.44 kBq/gm (66 nCi/gm).

Case study #4 used an operational limit set for waste handling and disposal. As the VPUs filled with waste, the radiation exposure was measured. After the addition of waste, some soil was added to the VPU to provide shielding and prevent spread of contamination. If the dose measured at the top of the VPU exceeded 0.15 Sv/hr (15 Rem/hr), the VPU would be considered "full" of waste and was backfilled with soil, gravel, and/or concrete. It is assumed that as waste was disposed to the VPU, an equivalent volume of soil was added, filling the VPU roughly halfway. A point source was chosen to simplify the dose model, which makes this case conservative. Using available MicroShield data from a cesium source, it was determined that a 0.15 Sv/hr (15 Rem/hr) dose at 2.29 m (7.5 ft) would result from a source of 8.81E+03 GBq (238 Ci) of cesium. Due to the available MicroShield data, we assume that cesium is the only isotope that is measured to determine the dose; this assumption is also conservative. Scaling up to the rest of the isotopes, 394 TBq (1.07E+04 Ci) would have been in the VPU. This decays and scales to about 10.6 kBq/gm (285 nCi/gm) for the final waste form. A summary of the results from the case studies is provided in Table I.

Table I. Case Studies for TRU Estimation of 618-10 VPUs

Case #	Key Assumptions	Estimated TRU Conc. kBq/gm (nCi/gm)	Comments
1	• Intermediate-level waste activity = 13 GBq/L (10 Ci/ft^3) • High-level waste activity = 1300 GBq/L (1000 Ci/ft^3)	0.407 (11)	Best-case scenario, lower bound on both intermediate- and high-level definitions
2	• 500 grams of fuel contaminated the waste in each milk pail (<1 kg in waste package)	2.44 (66)	Estimate of possible waste contamination
3	• Intermediate-level waste activity = 1200 GBq/L (900 Ci/ft^3) • High-level waste activity = 4000 GBq/L (3000 Ci/ft^3)	8.51 (230)	Worst-case scenario, upper bound on both levels
4	• Limit on VPU activity before backfilling = 0.15 Sv/hr (15 Rem/hr) @ top of VPU • Assume a point source halfway down VPU	10.6 (285)	Does not take into account distributed source, shielding from dirt/concrete layers, and dose from non-cesium isotopes

In light of these results, North Wind determined that there was insufficient evidence to guarantee that the waste in 618-10 is TRU or non-TRU. The case studies reveal that there are possibilities that the waste could be non-TRU or TRU. In-situ characterization of the waste was proposed to make an accurate determination of the TRU waste inventory. Characterization additionally compliments these case studies in identifying the specific isotopes and activities and providing confirmation of what is a best case and what is realistic. The case studies presented above are built upon assumptions garnered from historical documents. Some assumptions were conservative, such as allowing for a decay of 1 year before disposing the waste. The data is close enough to the decision point that further field characterization is necessary. Currently, characterization activities at 618-10&11 are in the planning stages.

WASHINGTON CLOSURE HANFORD APPROACH

Of the three distinct waste disposal configurations located in the 618-10 and 618-11 burial grounds, trenches, caissons, and VPUs, characterization modeling of VPUs is the focus primarily because the current plan is to remediate the trenches and caissons in a piece-by-piece removal operation, which would allow for sampling and analysis of the waste. The VPUs on the other hand, are planned for over-casing and removal as a unit with no plan for sampling prior to removal. A VPU, as described earlier, consists of five 208-liter (55-gallon) drums tack welded together and buried vertically in the burial ground. The primary container disposed to the VPU was a "milk pail", a 6.2-liter (1.7-gallon) cylinder. The waste was placed into the milk pail and a gelatin was poured in to retain the waste until it was dropped out of a bottom-loaded cask into the VPU. The maximum weight of a milk pail, per procedure, was 15,000 grams (33 pounds).

Radionuclide Distribution Determination

Where North Wind relied primarily on historical documents, publicly available and generated to support characterization generated some 20 years subsequent to burial grounds closure (including about 600 radiological surveys), WCH was able to access records located in Hanford

archives. An extensive document search of archived records was conducted, which yielded over 100 boxes of potentially applicable records. From these boxes, over 3200 radiological survey records, five years of semi-annual container disposal summaries, and 18 months of monthly disposal reports by container type and radioactivity concentration. Additionally, a limited number of facility operating procedures were retrieved, including the two buildings generating over 85 percent of the waste (325 and 327 buildings) that contained lists of chemicals used. This information was invaluable in determining which EPA waste codes may be present.

Of the over 3200 radiological surveys retrieved, over 85% were from the 325 and 327 buildings including virtually all of the high dose rate surveys relative to 618-10 and 618-11 waste disposal. These two primary facilities, the 325 Radiochemistry Building and the 327 Radiometallurgy Building, performed separations experiments and metallurgical analysis of spent fuel.

Since the mission of these buildings was primarily spent fuel sample analysis, application of the ORIGEN2 computer code is appropriate for single-pass reactor radionuclide distribution identification. For the purposes of 618-10&11 radionuclide distribution, an ORIGEN2 analysis from Hanford's C reactor was used. Input parameters into the program assumed one metric ton of fuel and fuel discharge mass from C Reactor's operating history. C Reactor was selected because it was the only single-pass reactor in continual operation during the time when the burial grounds were accepting waste (1954 to 1967). Based upon this information, the resulting radionuclide distribution was identified as representative for 618-10 and 618-11 burial grounds waste.

Table II. 618-10 and 618-11 ORIGEN2-Based Radionuclide Distribution (One year Decay)

Cs-137	Sr-90	Eu-154	Eu-155	Pu-241	Pu-238	Pu-239	Pu-240	Pu-242	Am-241
5.1E-01	4.8E-01	1.6E0-3	2.9E-03	4.5E-03	2.2E-06	6.1E-03	5.8E-04	3.5E-09	2.7E-04

Application of the Radionuclide Distribution

Using the mass of a milk pail and the radionuclide distribution identified above, the total amount of radioactivity in a milk pail was calculated, assuming TRU concentrations of 3.7, 7.4, 11.2, 14.9, 18.6, and 37.2 kBq per gram (100, 200, 300, 400, 500, and 1000 nanocuries per gram).

Once the amount of radioactivity per radionuclide was calculated and decayed one year to remove short-lived radionuclides, the dose rate associated with each TRU concentration were calculated using the ®MicroShield computer program.

The results of the MicroShield calculations were compared to the average dose rate associated with actual waste disposals of milk pails into the VPUs. Although the radionuclide concentration associated with 18.6 kBq/gm (500 ηCi/gm) could have been applied, a more conservative radionuclide concentration associated with 37.2 kBq/gm (1000 ηCi/gm) was

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selected. This radionuclide concentration was then decayed 47 years for 618-10 and 42 years for 618-11 to 2006. These periods of decay are associated with the year when most of the waste was generated per burial ground. The results were applied to subsequent calculations.

Table III. Milk Pail Dose Rate Comparison – Modeled vs. Average Milk Pail dose rates from Radiological Surveys

	Modeled Dose Rate	618-10 Average Radiological Survey Dose Rate	618-11 Average Radiological Survey Dose Rate
Dose Rate (Sv/hr [Rem/hr])	0.366 (36.6)	0.179 (17.9)	0.062 (6.2)

Once the amount of radioactivity was calculated per milk pail, the amount of radioactivity per VPU was calculated. In review of radiological surveys and radiological control procedures in use when the waste was generated, when dose rates at grade level reached approximately 0.15 Sv (15 Rem/hr) backfill material (site soil, gravel, or concrete) was added to reduce dose rates to below 0.05 Sv (5 Rem/hr). Using these dose rate values, an amount of fill material and milk pails per VPU was calculated. It was determined that each VPU contained a maximum of 40 milk pails (ten per VPU drum), assuming that, per historical documents, the top VPU drum was topped off with concrete to minimize dose rates at grade level. Given the amount of fill material and radioactivity calculated above, each VPU was modeled to have a TRU concentration of 9.3 kBq/gm (343 ηCi/gm).

The next step in the process was to determine the TRU concentration in the proposed remediation configuration (over-casing the VPU in a 107-cm (42-in) or 122-cm (48-in) diameter pipe, grouting below and above, and preparation for disposal). The rationale for averaging the radioactivity across the over-cased mass is that the VPUs have been in the ground for between 49 and 52 years and are likely deteriorated to the point where there is a potential for some radionuclide migration laterally, as well as vertically and that the surrounding soil, especially in the lower areas of the VPU are radioactively contaminated. When averaging the amount of VPU radioactivity over this over-cased mass the resulting TRU concentrations are:

Table IV. Dose rates and TRU Concentrations for Over-cased VPU Configurations

	107 cm (42 in) Over-cased VPU	122 cm (48 in) Over-cased VPU
TRU Concentration kBq/gm (ηCi/gm)	2.2 (63)	1.3 (36.6)
618-10 2006 Dose Rate mSv/hr (mRem/hr)	1.32 (132)	0.45 (45)
618-11 2006 Dose Rate mSv/hr (mRem/hr)	1.51 (151)	0.51 (51)

To remain below high radiation area controls during removal and subsequent handling, a 48-inch VPU over-casing will be selected.

Characterization Model Verification

Prior to retrieval, the 618-10 VPUs will be surveyed by installing cone penetrometers and detection tubes at a distance of approximately four inches from the VPU and performing radiological surveys, 90 degrees apart, in one foot increments along the length of each VPU. The results of this survey will provide an average gamma dose rate, gamma radionuclides and their activities, and plutonium concentration. The radionuclides identified and their concentrations will be compared to the ORIGEN2 and MicroShield calculations to validate the model and to determine of corrections are required. Subsequently, the measured Cs-137 activity and dose rates will be used for dose rate to curie conversion for waste remediated and the radionuclide distribution will be used to identify transuranic and other hard to detect radionuclides for waste removed from trenches and caissons.

CONCLUSION

Both of the methods employed to model radiological activity utilized different assumptions and source records while using the same basic conditions in the burial ground. The results garnered through both methods lie in the same range and result in the same conclusion. This agreement provides validation for these methods and supports action to seek verification using field instrumentation around the actual waste forms. The key value used for comparison between the two methods is the concentration of transuranic radionuclides within a 107-cm (42-in) over-cased VPU. The North Wind method produced a result of 1.78 kBq/gm (66 nCi/gm) and the WCH method produced a result of 2.2 kBq/gm (63 nCi/gm), a variance of approximately five percent.

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