Applying a MARSSIM-Based Approach to the Release of Soil and Slag Stockpiles

K.E. Taylor and K.M. Kasper
Envirocare of Utah, LLC
17 College St., Suite D, Greenville, SC 29601
U.S.A

ABSTRACT

The methods outlined in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) were used to characterize and release large volumes of stockpiled materials.[1] As MARSSIM applies directly to the release of land areas and structures, the survey and sampling approach had to be modified to address the release of material stockpiles. MARRSIM-based principles such as survey unit classifications, statistically-derived sampling approaches, and scanning, sampling, and survey approaches were used. Methodologies described in Draft NUREG-1767, “Radiological Surveys for Controlling Release of Solid Materials” were also used.

INTRODUCTION

The Whittaker Site, a waste and slag storage area regulated under a U.S. Nuclear Regulatory Commission (NRC) source material license, is located in a rural area of northwestern Pennsylvania. Site metal production processes resulted in a thorium- and uranium-bearing slag waste byproduct that was disposed of within the boundaries of the site property. The first step in decommissioning the site was to completely excavate the section of the site that contained the highest volume of radioactive materials. As part of the excavation process, more than 15,000 tons of the slag was separated from the soil in which it was buried and then segregated according to its radioactivity. Slag with radioactivity over the site-specific derived concentration guideline level (DCGL) is required to be shipped offsite for disposal while slag with uranium and thorium concentrations less than the DCGLs can stay on site.

To remove the bulk of the slag material from collocated soil, an automated screener was used. The screener separated soil and the smallest pieces of slag and other debris (< 2.54 cm) from larger pieces of slag and debris. This split the buried material basically into a slag pile and a soil pile. The large volume of soil (about 4,500 cubic meters), which contained average radioactivity concentrations less than the DCGL, was staged onsite for later use as backfill in excavated areas. A MARSSIM-based sampling plan was developed to characterize the soil pile to provide a high level of confidence so that the soil could safely be used as clean backfill.

The slag removed from the soil had to be further segregated according to its level of radioactivity with respect to the DCGL. To do this, a combination of techniques was used. Slag was monitored for gross gamma radiation as it exited the soil/slag screener. Radiation models were used to predict instrument response for slag at the DCGL. Material was then separated into two piles: one for slag reading greater than the DCGL and one for slag reading less than the DCGL.

A MARSSIM-based sampling plan was developed to characterize the slag pile containing slag that was less than the DCGL to provide a high level of confidence so that the slag could be placed back on the site as site stabilization material.

APPLYING THE SITE RELEASE CRITERIA

The NRC has approved the site DCGLs for Th-232+D, Ra-226+D, and U-238 as provided in Table I in Becquerel per gram (Bq/g) and Pico curies per gram (pCi/g). U-238 and Ra-226+D have separate DCGLs because only a portion of the U-238 in the slag materials is in equilibrium with Ra-226 and its decay daughters. The DCGLs were developed based on an industrial exposure scenario.

The Th-232+D DCGL applies to Th-232 in equilibrium with its decay daughters. Th-232 in the slag at the Whittaker site is in equilibrium with its decay daughters since it takes about 30 years for Th-232 to reach secular equilibrium and the age of the slag is at least 30 years. This condition is also supported by laboratory data.

The Ra-226+D DCGL applies to Ra-226 in equilibrium with its decay daughters. Data indicate that the level of equilibrium between U-238 and Ra-226 in site slag materials is not predictable. However, because Ra-226 and its decay daughters provide most of the dose impact of the U-238 decay chain, the radionuclide is evaluated separately from its parent U-238.

The U-238 DCGL applies to U-238 and its initial short-lived daughters only (no long-lived decay daughters present). The DCGL is equal to the 0.05 weight percent source material concentration and the peak dose of the U-238 at this level is well below 0.25 millisevert per year (mSv/yr) [25 millirem per year (mrem/yr)]. Analysis of slag samples indicates that U-238 is present, on average, at 25% of the Th-232 concentration and never exceeds the Th-232 concentration. Therefore, the U-238 concentration is conservatively assumed to equal the Th-232 concentration in sample analysis and DCGL comparisons.

Table I. Whittaker Site DCGLs

	Th-232+D	**Ra-226+D**	**U-238**
DCGL in Bq/g (pCi/g)	0.26 (7.0)	0.36 (9.7)	6.2 (166.5)
Peak Dose mSv/yr (mrem/yr)	0.249 (24.9)	0.249 (24.9)	0.063 (6.30)

SOIL PILE CLASSIFICATION

As soil was screened from the slag and debris, the soil was periodically collected directly into large metal bins that held about 1.4 m^3 of material. These full containers were surveyed as large-volume samples using sodium-iodide gamma field survey instruments. The gamma count rate or exposure rate data was correlated to soil activity in Bq/g based on *in-situ* gamma spectroscopy measurements. If the large-volume surveys indicated that the soil's residual radionuclide concentrations were less than the DCGLs, it was placed in the clean soil sock pile. Soil that was suspected of having radionuclide concentrations greater than the DCGLs was placed in a separate pile.

Based on the initial large-volume sample screening of more than 450 samples, there was a high probability that the soil in the clean stock pile had radionuclide concentrations less than the DCGLs. MARSSIM defines a Class 2 area as an area having a potential for radioactive contamination or known contamination, but not expected to exceed the DCGL. Therefore, the clean soil pile is classified as a Class 2 area. MARSSIM protocols recommend that a Class 2 land area survey receive a minimum of a 10% walk-over survey and a statistically derived number of discrete samples.

SOIL SURVEY DESIGN

The MARSSIM and Draft NUREG-1761 approaches for the release of materials rely on the ability to survey the material with some sort of scanning method. These methods may include a walkover survey or a scanning using a conveyor system. However, because of the volume of soil stockpiles at the Whittaker site, a scanning approach is not appropriate. However, through July 2005, more than 470 large-volume samples, or more than 660 m^3 were surveyed (scanned) using gamma survey instruments, making up about 15% of the total volume of the stockpile. These surveys were assumed to meet the intent of the minimum 10% scan proposed by MARSSIM for Class 2 survey units. Therefore, an approach consisting of only discrete sampling was developed to add to the large-volume sample screening data.

However, the conventional two-dimensional sampling approach presented in MARSSIM is not appropriate for the three-dimensional soil pile. A three-dimensional sampling approach may often include sampling in multiple layers down to a reasonable depth and “releasing’ each layer. Because the soil pile was more than 6 meters high in some places and the pile height was not constant, a multi-layer approach would result in an unreasonably high number of samples. Therefore, a volumetric release approach was developed to as if the soil was spread out over the ground surface in a single 15 cm layer.

MARSSIM limits the size of a Class 2 outside survey unit to 10,000 m^2. Assuming that the thickness of the surface soil layer addressed by current MARSSIM protocols is 15 cm, the resulting volume of soil released in a single Class 2 survey unit can be up to 1,500 m^3. As such, the soil would be characterized in survey unit volumes that were limited to the volume of soil not greater than 1,500 m^3. Because the soil pile was estimated to contain about 4,500 m^3 of soil, the pile was divided into three survey unit volumes.

According to MARSSIM, if the radionuclides of concern are also part of background, the Wilcoxon Rank Sum (WRS) non-parametric statistical comparison test is to be used. The null hypothesis (H_o) that the WRS test is designed to prove or disprove is: the residual radioactivity concentrations of the contaminants of concern in the survey unit exceed the reference area concentrations by more than the DCGLs. For this test, the number of required samples for the combined reference area and survey unit (N) is determined following the protocols in MARSSIM.

For this sampling effort, it is determined that 11 samples are required for the background data set and 11 samples are required from each 1,500 m^3 soil pile survey unit for the WRS test to be statistically valid with a 95% confidence. The calculation of N is described in the following section.

Calculation of N for Soil Samples

The number of required samples for the combined reference area and survey unit is determined using the method provided in Section 5.5.2.2 if the contaminant is also present in background.

$$N = \frac{(Z_{1-\alpha} + Z_{1-\beta})^2}{3(P_r - 0.5)^2} \qquad \text{(Eq. 1)}$$

Where:

- N = Number of combined samples in the survey unit and the reference area (rounded up to the nearest integer value)
- Z = Z-statistic based on selected α and β error rates forum in Table 5.2 in MARSSIM [1].
- α = Acceptable Type I (false positive) error rate.
- β = Acceptable Type II (false negative) error rate.
- P_r = Probability of a random measurement from the survey unit exceeding a random measurement from the reference area by less than the DCGL; P_r, found in Table 5.1 of MARSSIM [1], is based on the relative shift Δ/σ.
  – Δ is the shift which is typically defined as the DCGL minus the lower bound of the gray region (LBGR).
  – σ is the estimated standard deviation of data from the survey unit.

Samples collected from the soil pile suggest that the standard deviation (σ) for Th-232+D and Ra-226+D are 0.0548 and 0.0192 Bq/g respectively and the U-238 σ is assumed equal to the Th-232+D σ. With multiple contaminants, the standard deviations are normalized using the unity rule as follows [2]:

$$\sigma = \sqrt{\left(\frac{0.0548}{0.26}\right)^2 + \left(\frac{0.0192}{0.36}\right)^2 + \left(\frac{0.0548}{6.2}\right)^2} = 0.218 \qquad \text{(Eq. 2)}$$

Furthermore, for multiple contaminants, the DCGL is normalized to 1.0.

Therefore, with a DCGL normalized to 1.0, if the LBGR is estimated as one-half of the DCGL (or 0.5), this means that Δ is 0.5. With σ equal to 0.219, the relative shift Δ/σ then equals 2.28. Looking up these values in Table 5.1 of MARSSIM, P_r is then 0.946.[1]

Typically, 5% is an acceptable error rate for both Type I and Type II errors. Therefore, α and β are both equal to 0.05 and the Z-statistics from Table 5.2 of MARSSIM is 1.645.[1]

Substituting P_r and the Z-statistics into the above equation gives:

$$\frac{(1.645+1.645)^2}{3(0.946-0.5)^2} = 19 \text{ (rounded up to the nearest integer)}$$

Then, according to MARSSIM protocols, the value for N is increased by 20%.

$$N = 19 \text{ x } 1.2 = 22 \qquad \text{(Eq. 3)}$$

Therefore, the number for samples combined in the survey unit and reference area is 22, or 11 samples in each survey unit and 11 samples in each background reference area.

Soil Sample Collection and Analysis

The MARSSIM approach to soil sampling uses a two-dimensional random-start systematic grid to select the surface soil sampling locations. A similar approach to choosing the sample location was used in the three-dimensional survey unit volumes. Within each survey unit, six sample positions will were randomly selected in the horizontal plane using a random number generator. At three locations, a single sample was taken from the surface of the pile. At each of the other three locations, three subsurface samples were taken at three randomly-selected depths. As a result, a total of 12 samples from each survey unit volume (three from the surface and nine from the subsurface). While this approach did not result in a systematic grid sampling pattern, it was assumed that each sample location had an equal probability of producing a sample with elevated activity because the soil was randomly placed on the soil pile and it was often moved around as the pile was leveled out to make room for more soil. Furthermore, because the soil is to be used as backfill, the location of the sample in the pile is not relevant to the final position of the material.

Soil Sample Analysis and Data Evaluation

A high-purity germanium (HPGe) gamma spectroscopy system was used to analyze the 500 ml samples from the soil piles were analyzed on-site using a HPGe gamma spectroscopy system and approved sample preparation and analysis procedures. The detector and samples were placed in a shielded sample counting well to reduce the contributions of background and lower the minimum detectable concentrations (MDC). The detector was used in a standard laboratory counting arrangement and calibrated using a 500 ml standard with multiple gamma energies. The sample count time was determined to ensure that the MDCs were not greater than 50% of the contaminate DCGLs.

Previous sample analysis indicated that qualification of Th-232+D and Ra-226+D daughter products could be used to quantify the parent isotope’s concentration. The weighted average Ac-228 and Bi-214 activities were used to estimate the Th-232 and Ra-226 activities respectively. Data collected during other site characterization activities also showed that U-238 concentrations are always less than the Th-232 concentrations. Therefore, U-238 concentrations were conservatively assumed to be equal to the Th-232 (Ac-228) concentration in each sample.

Because multiple isotopes are present, the unity rule was used to assess the sample concentrations against the normalized DCGL of 1. For example, sample A1 had an Ac-228 (Th-232+D) concentration of 0.0577 Bq/g (1.56 pCi/g) and a Bi-214 (Ra-226+D) concentration of 0.037 Bq/g (1.0 pCi/g); applying sum of the fractions rule gives a normalized value of 0.34 as shown below.

$$\left(\frac{0.0577 Bq/g}{0.26 Bq/g}\right)_{Th-232+D} + \left(\frac{0.037\, pCi/g}{0.36 Bq/g}\right)_{Ra-226+D} + \left(\frac{0.0577\, pCi/g}{6.2 Bq/g}\right)_{U-238} = 0.34 \qquad \text{(Eq. 4)}$$

Table II provides the results of the soil sample analysis for the three survey unit volumes and a background reference area. The maximum Th-232+D concentration was 0.283 Bq/g (7.65 pCi/g) and the maximum Ra-226+D concentration was 0.107 Bq/g (2.90 pCi/g). The sum of the fraction exceeded 1.0 in two of the 36 samples (samples A6 and C6).

Table II. Soil Pile Sample Results (Bq/g)

Sample Number	Th-232+D (Ac-228)	Ra-226+D (Bi-214)	U-238	Sum of Fractions
A 1	0.058	0.037	0.058	0.34
A 2	0.077	0.042	0.077	0.43
A 3	0.135	0.061	0.135	0.71
A 4	0.162	0.060	0.162	0.82
A 5	0.159	0.056	0.159	0.80
A 6	0.283	0.096	0.283	1.41
A 7	0.083	0.063	0.083	0.51
A 8	0.041	0.044	0.041	0.29
A 9	0.082	0.043	0.082	0.45
A 10	0.087	0.048	0.087	0.48
A 11	0.084	0.036	0.084	0.43
A 12	0.073	0.032	0.073	0.38
B 1	0.062	0.034	0.062	0.35
B 2	0.079	0.032	0.079	0.41
B 3	0.155	0.046	0.155	0.75
B 4	0.086	0.055	0.086	0.50
B 5	0.070	0.045	0.070	0.41
B 6	0.083	0.046	0.083	0.46
B 7	0.071	0.060	0.071	0.45
B 8	0.087	0.046	0.087	0.48
B 9	0.077	0.037	0.077	0.41
B 10	0.062	0.027	0.062	0.32
B 11	0.073	0.046	0.073	0.42
B 12	0.100	0.040	0.100	0.50
C 1	0.067	0.054	0.067	0.42
C 2	0.151	0.074	0.151	0.81
C 3	0.133	0.076	0.133	0.75
C 4	0.191	0.073	0.191	0.97
C 5	0.177	0.099	0.177	0.99
C 6	0.271	0.107	0.271	1.39
C 7	0.067	0.041	0.067	0.38
C 8	0.080	0.043	0.080	0.44
C 9	0.074	0.037	0.074	0.40
C 10	0.071	0.034	0.071	0.37
C 11	0.063	0.038	0.063	0.36
C 12	0.066	0.029	0.066	0.34
Bkgnd 1	0.019	0.021	0.019	0.14
Bkgnd 2	0.026	0.015	0.026	0.15
Bkgnd 3	0.025	0.018	0.025	0.15
Bkgnd 4	0.025	0.016	0.025	0.15
Bkgnd 5	0.022	0.017	0.022	0.14
Bkgnd 6	0.026	0.018	0.026	0.16
Bkgnd 7	0.013	0.019	0.013	0.11
Bkgnd 8	0.028	0.019	0.028	0.17
Bkgnd 9	0.026	0.016	0.026	0.15

From Table I.4 for MARSSIM, the critical value for a WRS test with 12 survey unit samples and 9 reference area samples with an alpha error rate of 0.05 is 122.[1] Applying the WRS test to the survey unit volume data sets and the reference area data set, the resulting sums of the ranks of the reference area data are 144, 153, and 144 for survey unit volumes A, B, and C respectively. While the WRS test is not shown here, it is important to recall that the combined data set ranked during as part of the test includes the survey unit volume data (sum of the fraction values) and the reference area data plus the DCGL (sum of the fraction values plus 1.0). Because the sum of the ranks of the reference area data plus the DCGL is greater than the WRS critical value in each case, the null hypothesis (H_o) can be rejected with a 95% confidence even with samples that exceeded the DCGL in two of the survey unit volumes. Therefore the soils in the soil pile meet the DCGLs and can be left on site.

SLAG SURVEY DESIGN

Low-to-no activity slag material, referred to on-site and in site documents as Type 3 material, was segregated from higher activity material and stockpiled based on remediation support surveys conducted as the material came out of the mechanical screener and collected in the bucket of a front loader. If health physics technicians detected activity with a 2x2 NaI detector in the loader bucket after it was full, the entire bucket load was directed to separate stockpile for slag materials greater than the DCGLs. Loader buckets with no activity detected above the screening criteria were directed to the Type 3 slag pile. As such, there was a high probability that the Type 3 slag pile contained only material with Th-232+D and Ra-226+D concentrations less than the DCGLs. As a result, the Type 3 slag pile was classified as Class 2.

The design of the survey to release the Type 3 slag pile was similar to the approach taken for the soil pile. The volume of the Type 3 slag was less than 3,000 m^3 and, therefore, the pile was divided into 2 survey unit volumes. Because the standard deviations of the Th-232+D and Ra-226+D are similar in the Type 3 slag material to the standard deviations in the soil pile material, the number of samples required from each survey unit volume is also eleven samples.

Because of the variability in the different types of slag materials in the Type 3 slag pile, it was determined that a sample of the material would be best represented by a large-volume sample rather than a small 500 ml sample. Therefore, 1.4 m^3 bins were randomly filled with slag material and surveyed using a Canberra *In-Situ* Object Counting System (ISOCS). One 5-minute count was made 2 feet from the sample bin on the side that demonstrated the second highest exposure rate using a microR exposure rate meter. Typically, however, there was very little measurable difference in the exposure rates observed at 2 feet from each of the four sides of the sample bins.

The analysis of the large-volume provided a significant level of confidence that the Type 3 slag materials could be used on site as stabilization materials on excavated embankments during a demobilization period. Eventually the material will be spread over the site in a thin layer as a base for the fill material. At that time, a walk-over survey will be performed finalizing the release of the material.

Slag Sample Analysis and Data Evaluation

As with the soil pile samples, gamma spectroscopy data was evaluated to determine the Th-232+D and Ra-226+D concentrations based on the weighted averages of the Ac-228 and Bi-214 peaks. U-238 concentrations were again conservatively assumed equal to the Th-232+D concentrations because site data indicate that U-238 is always less than Th-232+D in slag samples containing elevated Th-232+D concentrations. The Type 3 slag pile data is presented in Table III. No large-volume slag samples showed concentrations that exceeded the DCGLs and the sum of the fractions exceeded 1.0 in only one of the 24 samples (sample T3B-6).

Table III. Type 3 Slag Pile Large-Volume Sample Results (Bq/g)

Sample Number	**Th-232+D (Ac-228)**	**Ra-226+D (Bi-214)**	**U-238**	**Sum of fractions**
T3A-1	0.084	0.084	0.084	0.57
T3A-2	0.040	0.033	0.040	0.25
T3A-3	0.121	0.037	0.121	0.59
T3A-4	0.101	0.092	0.101	0.66
T3A-5	0.093	0.067	0.093	0.56
T3A-6	0.074	0.056	0.074	0.45
T3A-7	0.043	0.067	0.043	0.36
T3A-8	0.043	0.088	0.043	0.42
T3A-9	0.043	0.085	0.043	0.41
T3A-10	0.158	0.102	0.158	0.92
T3A-11	0.109	0.078	0.109	0.65
T3B-1	0.178	0.069	0.178	0.91
T3B-2	0.060	0.074	0.060	0.45
T3B-3	0.094	0.035	0.094	0.48
T3B-4	0.044	0.035	0.044	0.28
T3B-5	0.122	0.037	0.122	0.60
T3B-6	0.212	0.087	0.212	1.09
T3B-7	0.108	0.067	0.108	0.62
T3B-8	0.173	0.038	0.173	0.80
T3B-9	0.053	0.034	0.053	0.31
T3B-10	0.044	0.028	0.044	0.25
T3B-11	0.121	0.033	0.121	0.58

From Table I.4 for MARSSIM, the critical value for a WRS test with 11 survey unit samples and 9 reference area samples is 116.[1] Applying the WRS test to the survey unit volume data sets and the reference area data set, the resulting sum of the ranks of the reference area data is 144 for both survey unit volumes T3A and T3B. Because the sum of the ranks of the reference area data is greater than the WRS critical value in each case, the null hypothesis (H_o) can be rejected with a 95% confidence. Therefore, with supporting walk-over surveys yet to be conducted, the slag materials in the Type 3 slag pile meet the DCGLs and can be left on site.

CONCLUSION

It was important to site operations that the stockpiles of fill materials be released or at least sampled in a manner such that there was a high level of confidence that the materials met the DCGLs prior to backfilling or use in site stabilization. Therefore, a modified MARSSIM survey and sampling approach was designed and implemented. The approach used randomly collected samples, on-site gamma spectroscopy analysis, scan surveys, and a proven statistical test with properly sized data sets to prove with a 95% confidence that the material in the Type 3 slag pile and the soil pile met the DCGLs.

REFERENCES

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