DEVELOPMENT OF SOIL ACTIVITY CORRELATIONS AND SURROGATE VALUES FOR PERFORMING IN SITU SOIL MEASUREMENTS

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ABSTRACT

An investigation was conducted at the West Valley Demonstration Project (WVDP) to determine if Cs-137 could be used as a target radionuclide when performing in situ measurements of contaminated soil with a hand-held NaI scintillation detector operated as a count rate instrument. Derived concentration guideline levels (DCGLs) and location-specific Cs-137 surrogate values were developed based on site characterization data. Cs-137 was chosen as the target radionuclide because of the relative ease of detecting the associated 662 keV gamma with field instrumentation and the relatively high abundance of Cs-137 as compared to other gamma-emitting radionuclides at the WVDP.

As part of the WVDP's Contaminated Soil Management Program, contaminated soil samples were obtained and analyzed in early 1998. Results of the soil analysis produced characterization information including Cs-137 content (µCi/g), percent moisture content, etc. These soil samples represented 14 distinct areas of the site where soil was previously excavated and containerized. Residual soil from the samples was labeled, containerized, and stored. Fifty-seven direct radiation measurements were taken on samples of this previously characterized soil using a shielded NaI detector. The results were graphed to display net counts per minute (ncpm) verses volumetric activity of Cs-137 in the first 15 centimeters (six inches) of soil. Calculations produced the linear regression line describing the relationship between ncpm (the x variable) and the concentration of Cs-137 (the y variable) in picocuries per gram (pCi/g). Biased, in situ gamma measurements and soil sampling were performed to validate the correlation line. The validation regression line produced values that remained within ± 10% of the correlation line. Statistics were applied to the correlation line producing a final correlation equation of:

$$\text{Cs-137 [pCi/g]} = (0.0215 \cdot \text{ncpm}) + 19.6,$$ representing the +95% confidence level.

Location-specific surrogate values were developed for Cs-137 using site characterization information and defined DCGLs. The Cs-137 surrogate DCGLs ranged from 224 pCi/g at the Low-Level Waste Treatment Facility (LLWTF) to 661 pCi/g at the Vitrification Test Facility (VTF). Location-specific NaI field instrument response, corresponding to the surrogate values, were based on the correlation between NaI count rate and Cs-137 activity concentration in the soil, and ranged from 9,525 ncpm at the LLWTF to 29,883 ncpm at the VTF.

With the development of NaI instrument correlation and surrogate values, Radiation Protection personnel can use a NaI detector in the field in a majority of cases to determine if soil meets or exceeds the containerization criteria. The ability to make these field decisions will improve efficiency and provide WVNS with more options for managing contaminated soil.

INTRODUCTION

The West Valley Demonstration Project (WVDP) occupies approximately a 200-acre area that was originally the site of the United States' first commercial nuclear fuel reprocessing plant. The plant reclaimed uranium and plutonium from spent nuclear fuel through dissolution and chemical extraction. Through the course of its operation, the site accumulated approximately 660,000 gallons of liquid high-level radioactive wastes (HLW) that were stored in underground tanks. The West Valley Demonstration Project Act was passed in October 1980 directing the United States Department of Energy (DOE) to develop and demonstrate technology for solidifying the liquid HLW into a form suitable for long-term storage (1). Vitrification of the liquid HLW in a borosilicate glass matrix was the technology chosen. As a result of these operations, low-level waste (LLW) and contaminated soils were generated at the site.

The WVDP currently stores approximately 4,300 cubic yards of radiologically contaminated soil in steel boxes and in tarp-covered rolloff dumpsters on site. The contaminated soil was found during site construction activities necessary to complete the WVDP mission. According to site procedures, all soils must be surveyed when excavated. WVDP's Radiological Controls Manual requires containerization of soil when specific activity levels reach 10,000 disintegrations per minute per gram (dpm/g) gross beta-gamma or 2,000 dpm/g gross alpha (2). Soils with specific activity levels less than the containerization criteria may be stored on site, uncontainerized under certain conditions. Soil contamination area posting requirements are set at 100 dpm/g gross beta-gamma and 20 dpm/g gross alpha. Routine practice at the site has been to containerize soils that exceeds the soil contamination area posting requirements. Soil samples are routinely screened by Radiation Protection Technicians using a frisker searching for areas of potential contamination as indicated by an increased count rate on the instrument. Soil samples are also analyzed on a low-background proportional counter (Tennelec®).

Revised soil containerization limits have been developed using dose-based, site-specific, Derived Concentration Guideline Levels (DCGLs) for individual radionuclides (3). As part of this effort, a correlation equation for determining Cs-137 activity in the first 15 centimeters of soil based on field NaI instrument response was established. Cs-137 was chosen as the target radionuclide because of the relative ease of detecting the associated 662 keV gamma with field instrumentation and the relatively high abundance of Cs-137 as compared to other gamma-emitting radionuclides at the WVDP. Since Cs-137 is not the only radionuclide in WVDP's soil, calculations were performed using guidance contained in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) to establish surrogate values for Cs-137 based on site soil characterization data (4).

It should be noted that not all areas of the WVDP can be characterized using Cs-137 as the target radionuclide. Certain areas of the WVDP may be contaminated with Sr-90 with very little to no Cs-137 present thereby making the instrument correlation impossible.

DEVELOPMENT OF NAI INSTRUMENT RESPONSE CORRELATION EQUATION TO CS-137 SOIL ACTIVITY

Initial Development of Correlation

Contaminated soil was removed from storage boxes and placed into labeled, five-gallon buckets in early 1998 as part of the WVDP Contaminated Soil Management Program. Aliquots of the soil were removed from the buckets and sent to an off-site laboratory for isotopic analysis. The buckets holding the remaining soil were placed into three containers and stored on site (LSA3). Results of the soil characterization were published in WVDP-308, Final Report: Contaminated Soil Management Program (5).

In August 1998, two storage containers holding the buckets of soil were transported to an area east of the plant for surveying. Surveys were performed with a shielded NaI scintillation detector (Detector/probe, Ludlum 2241/44-10, serial number 132188/RP15194, 5.9% efficiency at 662 keV). Ten one-minute background area measurements were taken prior to introducing the storage containers/buckets into the survey area. Each bucket was removed from its storage container, moved to the survey location, opened, and a one-minute integrated count was taken using the NaI detector. The side walls of the NaI detector were shielded with lead to minimize background from lateral sources and skyshine from existing structures. The NaI detector was placed in the center of the bucket in direct contact with the poly bag containing the soil. Each bucket was filled with soil to a depth of approximately 15 centimeters.

All soil characterization data was adjusted for moisture using Equation (1).

$$Moisture\&Corrected\ \ Concentration\ \ ' \ \frac{reported\ \ \frac{\mu Ci}{g_{dry}}}{1\&MC\frac{g_{dry}}{g_{total}}} \qquad \text{(Eq. 1)}$$

where: MC = moisture content

The Cs-137 concentration determined from laboratory analysis was plotted verses NaI field readings as shown in Figure 1. Linear regression was applied to the data yielding a line represented by the equation: y = 0.0163x - 8.7354, which describes the relationship between NaI instrument response in net counts per minute (ncpm) and the concentration of Cs-137 in picocuries per gram (pCi/g) of soil. Statistics of the regression line show a correlation coefficient (r) of 0.944. The indicated uncertainty in Cs-137 concentration, as obtained from laboratory analysis, was determined as two standard deviations and uncertainty in ncpm was ± 10% based on instrument calibration criteria.

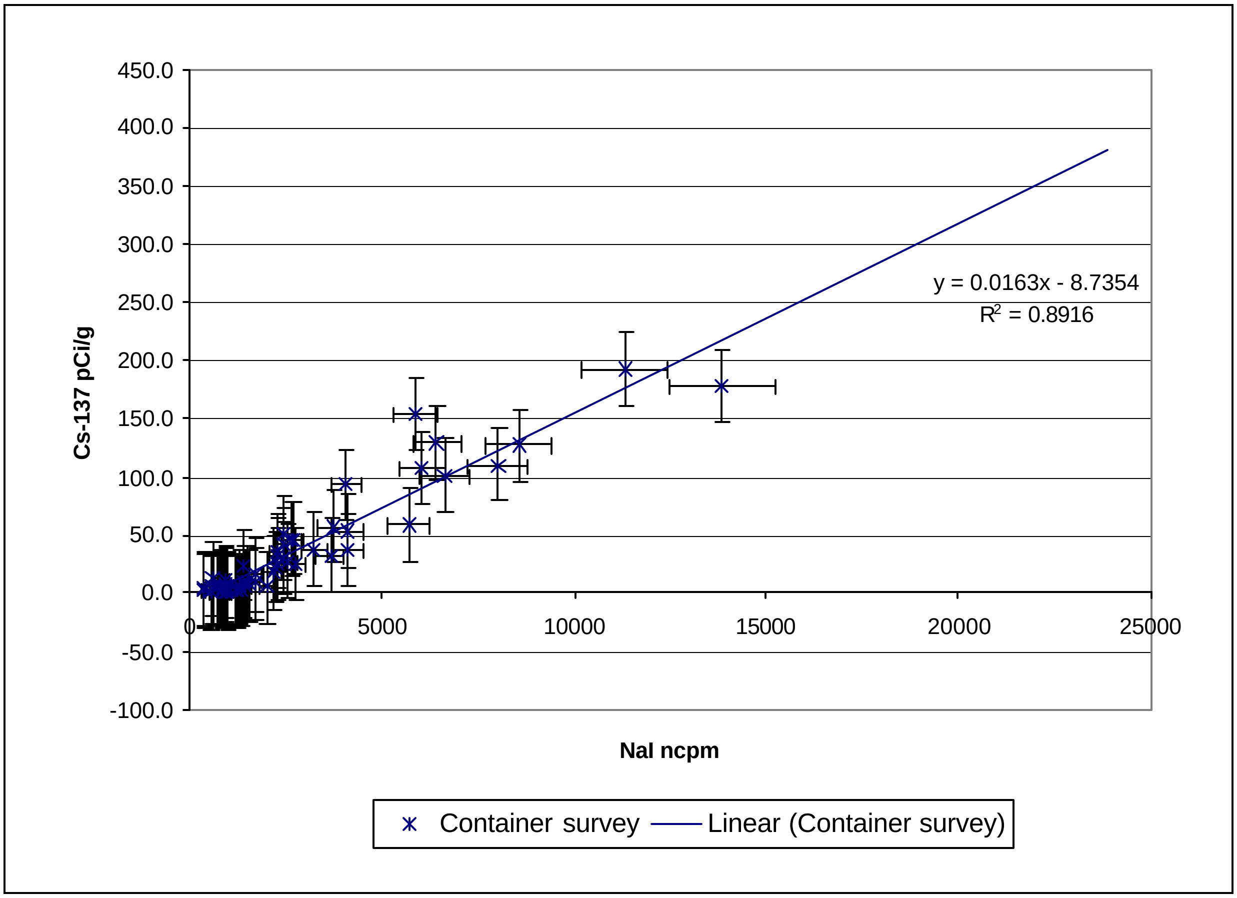

Figure 1. Container survey correlation results

Validation of Correlation Line

Data Quality Objectives (DQOs) were developed to validate the correlation equation (6). The DQOs defined the number of survey points to take in order to validate the correlation line. A DCGL of 710 pCi/g Cs-137 was generated by the RESRAD computer code as part of the technical basis document for soil containerization criteria and is used in this report as an upper bound. A lower bound of 50 pCi/g Cs-137 was chosen based on the soil contamination area posting requirements of 100 dpm/g beta-gamma.

Twelve field soil samples were obtained using a biased sampling approach. Sample areas were identified using the NaI detector looking for indications of elevated soil activity. When the sampling area was chosen, background levels were determined by taking ten one-minute counts with the NaI detector. The in situ soil measurements were performed by taking a series of three one-minute counts at a distance of 2 inches from the surface of the soil. Soil was then excavated using a 3.5 inch diameter, 2 inch deep stainless steel sampler following the steps outlined in EM-5, Environmental Monitoring Procedure (7). Soil was excavated to a depth of 6 inches and placed in a labeled sample container. Approximately 500 ml of soil was excavated from each sample location. Soil gamma scans and percent moisture content determinations were performed by the WVDP Environmental Lab.

Results of the validation survey were plotted yielding a regression line of y = 0.018x - 10.553 as shown in Figure 2. Examining the regression data shows that the initial correlation and validation lines are strongly correlated with correlation coefficients of 0.944 for the container data and 0.977 for the field data. The two regression lines start out close together and diverge. However, evaluation of the regression line over the range of interest (50 - 710 pCi/g) shows the two lines varying by a maximum of 10.1%. Values of 50 pCi/g yield ncpm values of 3,603 and 3,364, a difference of 7.1%. Values of 710 pCi/g yield ncpm values of 44,094 and 40,031, a difference of 10.1%.

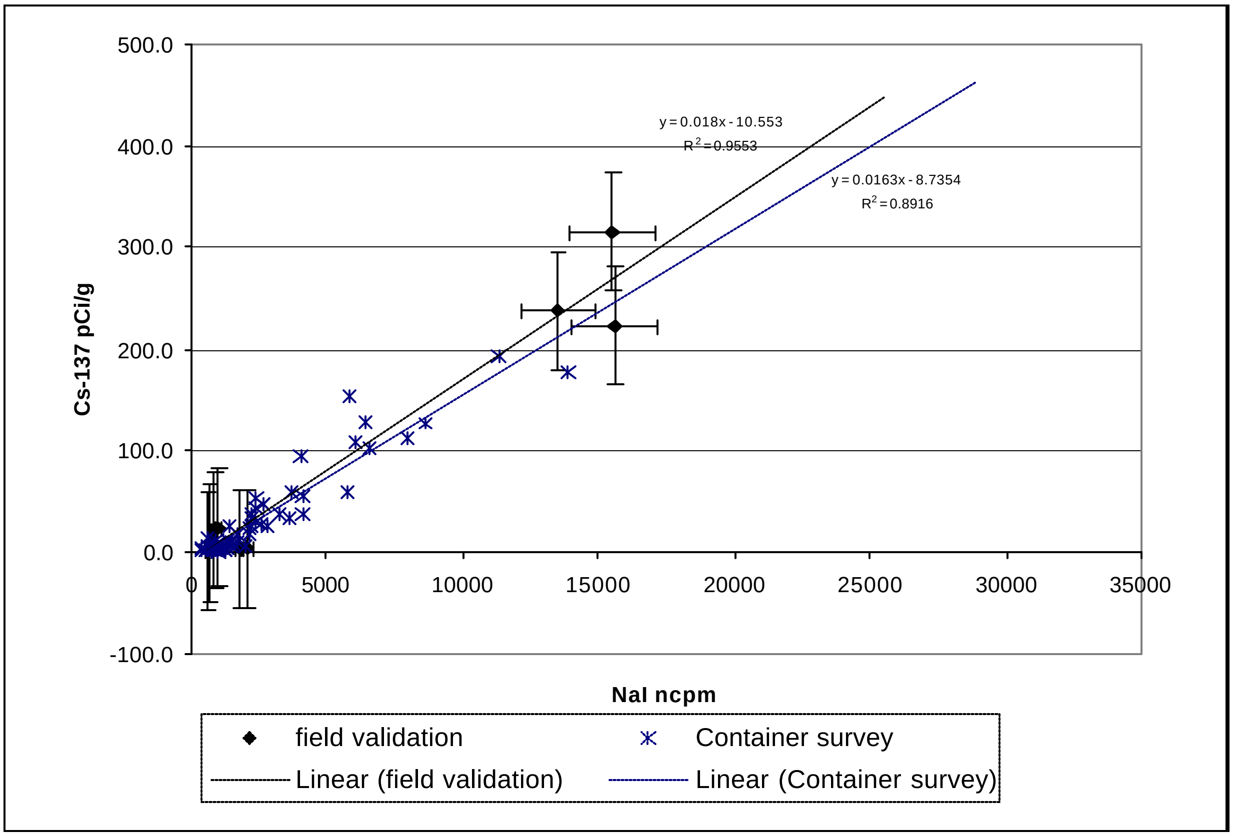

Figure 2. Validation of container survey data with field survey data

Three of the 12 validation samples (NDA1, UR1, and UR3) were significantly lower than predicted when compared to the initially developed validation line. These soil samples were retrieved from storage and surveyed with the same NaI detector used in the in situ survey. The three soil samples all produced significant differences between initial and secondary measurements. Based on the additional survey, the three survey points were classified as outliers and excluded from the data set used in the final regression analysis. A possible explanation for the difference is that in situ results were influenced by contamination deeper in the ground than was excavated.

Validation criteria for the container survey regression line was defined as the two regression lines agreeing to within 20% of each other over the range of interest. Data analysis results from the field survey validate the container correlation line.

Final Correlation Equation

The field validation regression line was chosen as the line to represent the instrument response to Cs-137 soil activity because it was slightly more conservative and had a higher correlation coefficient. Lines representing the 95% confidence level of the correlation line were drawn as shown in Figure 3. All data, with the exception of one point, fit within the ± 95% confidence level lines. The final equation chosen to represent the instrument response to Cs-137 concentration in soil is expressed as Equation (2) and represents the + 95% confidence level.

$$Cs\text{-}137\,pCi/g = 0.0215 \times (ncpm) + 19.6 \qquad \text{(Eq. 2)}$$

Using Equation (2), 50 pCi/g correlates to 1,413 ncpm and 710 pCi/g correlates to 32,110 ncpm using the NaI field instrument.

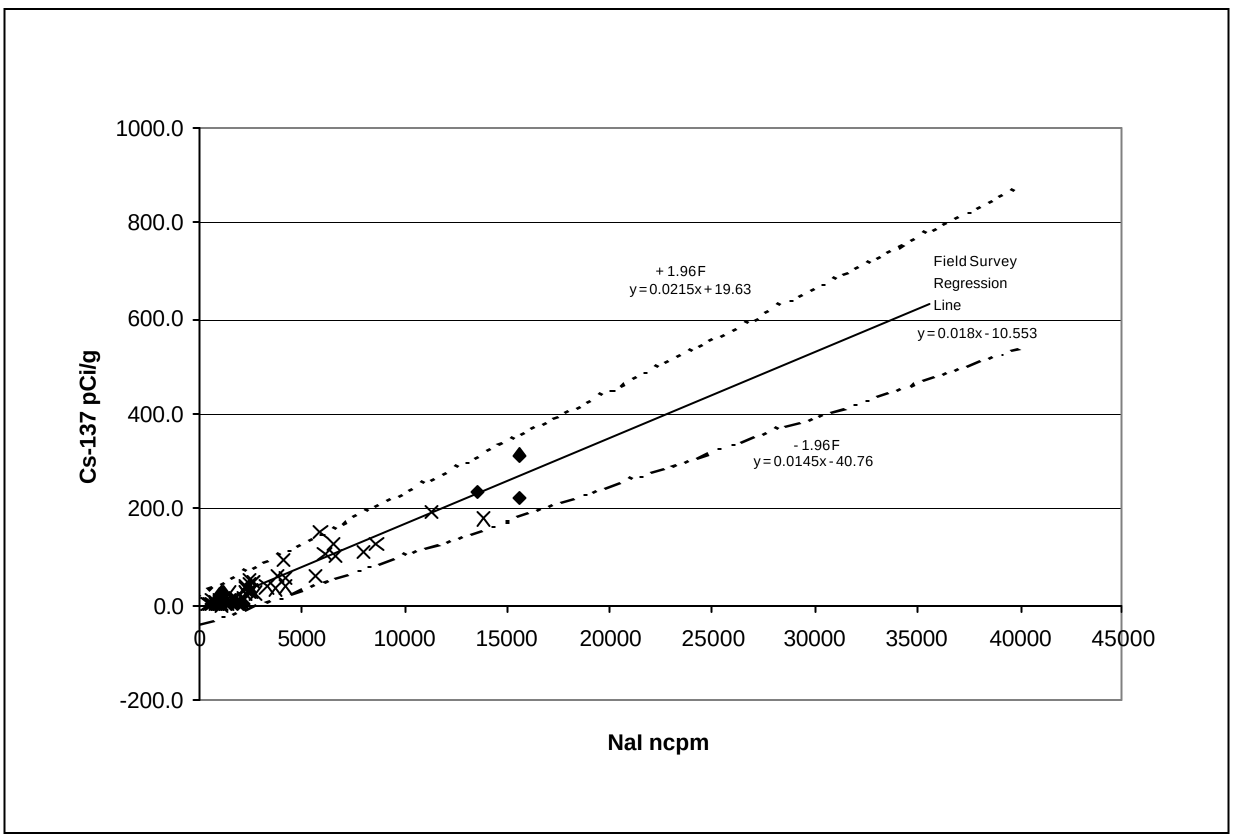

Figure 3. Final correlation line showing 95% confidence intervals

Interference from other Gamma Emitters on Instrument Response

The primary gamma emitter in soil at the WVDP is Cs-137 (Ba-137m). Other gamma emitters do exist in the soil in concentrations large enough to affect the NaI field instrument response, especially at lower energies. In an attempt to quantify this effect, the MicroShield® (8) program was run using existing soil analysis data contained in WVDP-308, Final Report: Contaminated Soil Management Program. Soil data was selected based on the lowest ratio of Cs-137 to other gamma emitting radionuclides in order to produce the most conservative results. Sample data selected is shown in Table 1. Results of the analysis were used with conversions contained in NUREG-1507 to determine the effect each individual isotope had on the NaI detector response (9). The overall effect is shown in Table 2, with Cs-137 gammas accounting for approximately 75% of the total cpm response. Table 3 shows the contribution by isotope. This effect adds an element of conservatism into the results and can be significantly reduced by using a NaI instrument with a window set around the 662 keV peak from Ba-137m.

Table I. NDA B-25 soil

Nuclide	**µCi/g**	**µCi/cm^3**	**Nuclide**	**µCi/g**	**µCi/cm^3**
Am-241	2.92E-5	4.67E-5	Ni-59	<4.43E-6	<7.09E-6
Cm-242	<7.47E-7	<1.20E-6	Ni-63	8.28E-5	1.32E-4
Cm-243/244	7.07E-7	1.13E-6	Np-237	<4.86E-7	<7.78E-7
Cm-245/246	7.92E-7	1.27E-6	Pm-147	<3.12E-6	<4.99E-6
Co-60	6.63E-6	1.06E-5	Pu-238	1.44E-5	2.30E-5
Cs-134	<3.20E-7	<5.12E-7	Pu-239/240	2.03E-5	3.25E-5
Cs-137	1.67E-3	2.67E-3	Pu-241	3.08E-4	4.93E-4
Eu-152	<1.47E-6	<2.35E-6	Ra-226	8.96E-7	1.43E-6
Eu-154	9.12E-7	1.46E-6	Ru-106	<3.28E-6	<5.25E-6
Eu-155	<1.03E-6	<1.65E-6	Sb-125	<1.84E-6	<2.94E-6
Fe-55	<5.48E-5	<8.77E-5	Sr-90	6.49E-4	1.04E-3
I-129	<6.97E-6	<1.12E-5	Tc-99	<2.30E-6	<3.68E-6
K-40	2.41E-5	3.86E-5	U-233/234	1.18E-6	1.89E-6
Nb-94	<1.04E-7	<1.66E-7	U-235/236	<4.41E-7	<7.06E-7
Note: < indicate less than listed Minimum Detectable Concentration (MDC)			U-238	1.01E-6	1.62E-6

Table II. NUREG-1507 (NaI 2x2) instrument dose rate to cpm conversions

MeV	From uShield (mR/hr)	NUREG-1507 conversion (cpm/uR/h)	cpm
0.015	1.20E-09		0
0.020	3.133E-10	2,200	0
0.030	2.455E-03	5,160	12,668
0.040	7.527E-04	8,880	6,684
0.050	2.083E-06	11,800	25
0.060	1.285E-02	13,000	167,050
0.080	2.768E-07	12,000	3
0.100	5.772E-05	9,840	568
0.150	2.794E-07	6,040	2
0.200	2.865E-05	4,230	121
0.300	2.269E-05	2,520	57
0.400	1.721E-06	1,700	3
0.500	6.353E-07	1,270	1
0.600	4.973E-01	1,010	502,273
0.800	1.709E-04	710	121
1.000	4.017E-03	540	2,169
1.500	7.670E-03	350	2,685
Total	5.253E-01		694,429

Table III. Specific isotope effect on total cpm

Nuclide	NaI cpm	Nuclide	NaI cpm
Am-241	6,989	Eu-154	746
Cm-242	160,030	K-40	723
Cm-243	401	Pu-238	5
Pu-238	5	Pu-239	7
U-238	1	Ra-226	22
Ba-137m	521,555	U-233	2
Co-60	3,942	U-238	1

Nuclide	NaI cpm	Nuclide	NaI cpm
		Totals	694,429

SURROGATE VALUE DEVELOPMENT

If Cs-137 was the only radioisotope in WVDP soil, the correlation equation developed earlier in this report could be directly applied to establish an instrument reading where soil containerization would be required. Because soil at the WVDP contains a host of radioisotopes that do not emit gamma radiation, a Cs-137 surrogate value needed to be developed to account for the other isotopes. The Cs-137 surrogate DCGLs (D_{total}) were calculated using Equation 3 and the site-specific DCGLs contained in Table 4. Cs-137 surrogates, including correlated ncpm values, are listed in Table 5 and are shown graphically as Figure 4.

If

$$C_2 = R_2 C_1,\ C_3 = R_3 C_1,\ \ldots,\ C_i = R_i C_1,\ C_n = R_n C_1$$

then

$$\frac{C_1}{D_1} + \frac{C_2}{D_2} + \frac{C_3}{D_3} + \ldots + \frac{C_n}{D_n} = \frac{C_1}{D_1} + \frac{R_2 C_1}{D_2} + \frac{R_3 C_1}{D_3} + \ldots + \frac{R_n C_1}{D_n}$$

$$= C_1\left[\frac{1}{D_1} + \frac{R_2}{D_2} + \frac{R_3}{D_3} + \ldots + \frac{R_n}{D_n}\right]$$

$$= \frac{C_1}{D_{total}}$$

where

$$D_{total} = \frac{1}{\left[\frac{1}{D_1} + \frac{R_2}{D_2} + \frac{R_3}{D_3} + \ldots + \frac{R_n}{D_n}\right]} \qquad \text{(Eq.3)}$$

D_{total} = Cs-137 Surrogate DCLG
D_n = the DCGL for radionuclide *n*
C_i = Concentration of radionuclide *i*

Table IV. Site-specific soil containerization DCGLs

Nuclide	DCGL (pCi/g)	Nuclide	DCGL (pCi/g)	Nuclide	DCGL (pCi/g)	Nuclide	DCGL (pCi/g)
H-3	4,500	Sb-125	1,600	Ra-228	210	Pu-241	4,500
C-14	4,500	I-129	4,500	U-232	210	Pu-242	900
Fe-55	4,500	Cs-134	350	U-233	900	Np-237	900
Ni-59	4,500	Cs-135	4,500	U-234	900	Am-241	900
Ni-63	4,500	Cs-137	710	U-235	900	Am-243	900
Co-60	170	Pm-147	4,500	U-236	900	Cm-242	900
Nb-94	350	Eu-152	350	U-238	900	Cm-243	900
Sr-90	4,500	Eu-154	330	Pu-238	900	Cm-244	900
Tc-99	4,500	Eu-155	4,500	Pu-239	900		
Ru-106	3,800	Ra-226	210	Pu-240	900		

Table V. Cs-137 location-specific Cs-137 surrogate values

Location	Cs-137 surrogate (pCi/g)	Cs-137 surrogate (ncpm)
LLWTF	224	9,525
EDR	276	11,915
NDA rolloffs	309	13,451
VIT	361	15,887
NDA S-70	382	16,871
WTF	449	19,952
SPR	466	20,754
UR	490	21,859
MNT	503	22,469
LAG	557	24,988
NW	595	26,768
NDA B-25	616	27,729
CSS	658	29,699
VTF	661	29,833

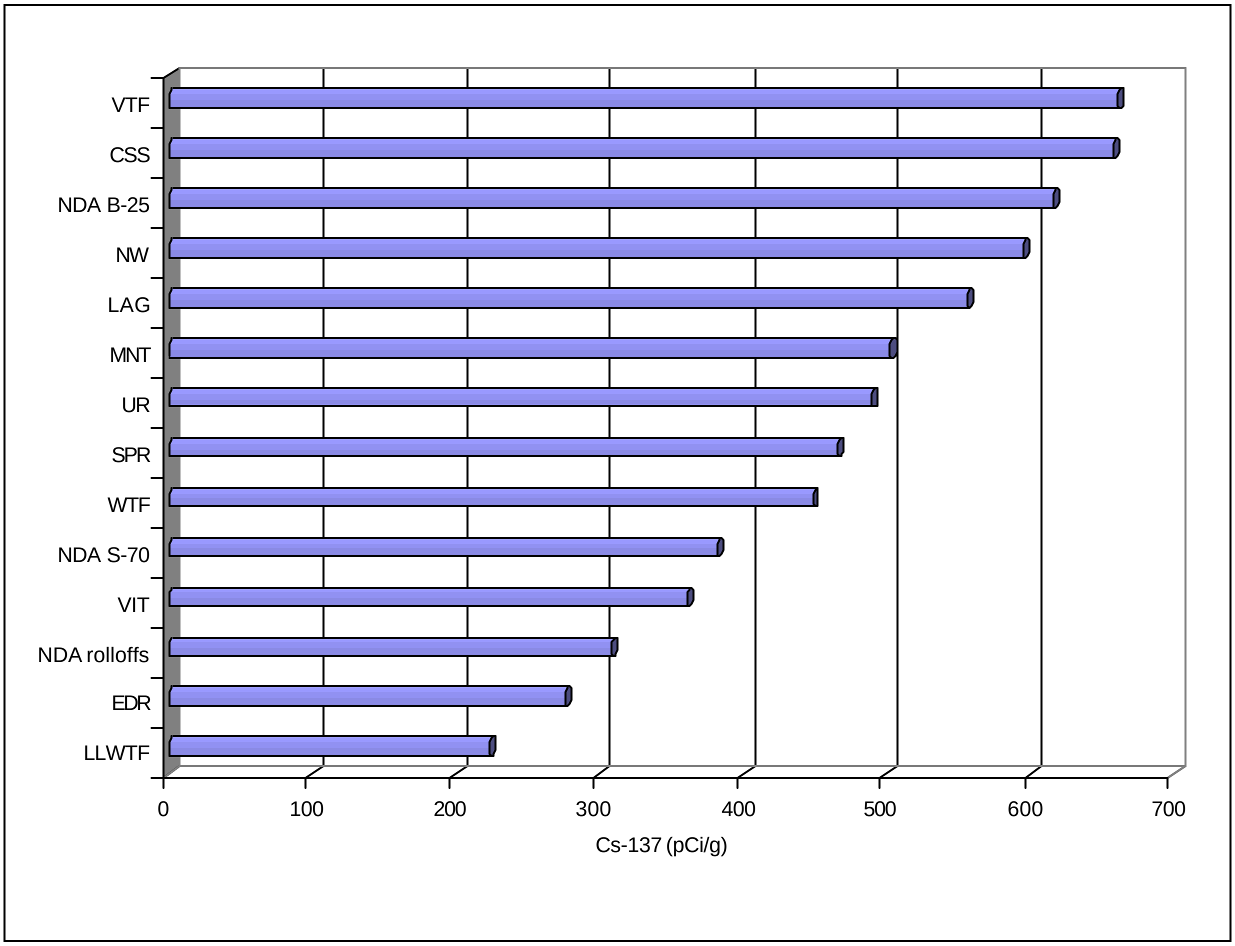

Figure 4. Cs-137 surrogate values

VTF = Vitrification Test Facility
CSS = Cement Solidification System
NDA = Nuclear Regulatory Commission Licensed Disposal Area
NW = New Warehouse
LAG = Temporary Storage Area
MNT = East of Maintenance
UR = Utility Room
SPR = Southeast Plant Road
WTF = Waste Treatment Facility
VIT = Vitrification Load-In Facility
EDR = Equipment Decontamination Room
LLWTF = Low-Level Waste Treatment Facility

CONCLUSION

A correlation line for determining Cs-137 activity (pCi/g) in the first 15 centimeters of soil from in situ NaI counting was developed. The correlation line was validated by performing additional field measurements and sampling for gamma analysis. Validation data results were within 10% of expected values. The final correlation equation at the + 95% confidence level is :

$$Cs\text{-}137\ (pCi/g) = 0.0215 * (ncpm) + 19.6.$$

Cs-137 surrogate values were developed using site-specific soil characterization data with results ranging from 224 to 661 pCi/g. The correlation equation was applied to the surrogate values resulting in NaI values ranging from 9,525 to 29,883 ncpm.

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