SuperHENC: Final Performance and Certification Summary

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The Super High Efficiency Neutron Counter (SuperHENC) mobile assay system for Standard Waste Boxes (SWBs) was designed, fabricated, calibrated, qualified for material accountability measurements, and certified for WIPP use under a joint team comprising Los Alamos National Laboratory (LANL), BNFL Instruments, and Rocky Flats Environmental Technology Site (RFETS) personnel. Calibration measurements include chamber efficiency mapping, establishment of a doubles calibration curve using plutonium standards, development of the Add-A-Source (AAS) correction factor (CF) curve, establishment of multiplicity-based matrix correction equations for metallic and high hydrogen matrices, and development of cosmic ray background suppression techniques. Validation activities on SWB standards demonstrated that the measurement method met accountability criteria established for 208-L (55-gal) drums for a large range of matrix materials (combustibles, plastics, metals, and heterogeneous mixtures). Minimum Detectable Concentration (MDC) characterization of these material types has also indicated that the SuperHENC can perform a TRU/LLW sort at the 100 nCi/g threshold.

1. INTRODUCTION

The SuperHENC assay system for Standard Waste Boxes (SWBs) is a joint project among LANL, BNFL Instruments Inc. (BII), and Rocky Flats Environmental Technology Site (RFETS). It was deployed at RFETS in January 2001. The system was anticipated to be capable of (1) substantially lower detection limits than traditional passive-only neutron systems; (2) meeting WIPP quality assurance objectives (QAOs) for 208-L (55-gal) drums or much larger containers; (3) meeting RFETS DOE Material Control & Accountability (MC&A) QAOs for 208-L (55-gal) drums or much larger containers; and (4) assay performance that is insensitive to mixtures of matrix materials due to LANL innovations in hardware and physics design. Many of the details of this system have been presented previously.[1] This paper summarizes the performance parameters relevant to WIPP certification of the system, including a calibration summary, matrix specific performance/QAO data, Performance Demonstration Plan (PDP) data for the first cycle of the box PDP, and Minimum Detectable Concentration (MDC) methodology and results.

2. SYSTEM DESCRIPTION

The SuperHENC is an entirely self-contained mobile system. It is mounted on a 48-ft commercial "low boy" trailer that is 8 ft 6 in. wide and just under 13 ft 6 in. high. The forward third of the trailer contains the control room. The middle third contains the SuperHENC. The SuperHENC trailer has also been outfitted by BII with a gamma spectroscopy system (the SGEAS) for the identification of other elements of interest. The aft section is used for the BII gamma spectroscopy system.

The trailer is designed for rapid setup and leveling. Waste packages are loaded from the right side of the trailer with the long axis of the SWB parallel to the SuperHENC onto a fold-down drawbridge. The drawbridge has an integrated load cell, and uses a motorized pallet to introduce the sample into the assay chamber. Figure 1 shows the SuperHENC trailer and the SWB loading dock.

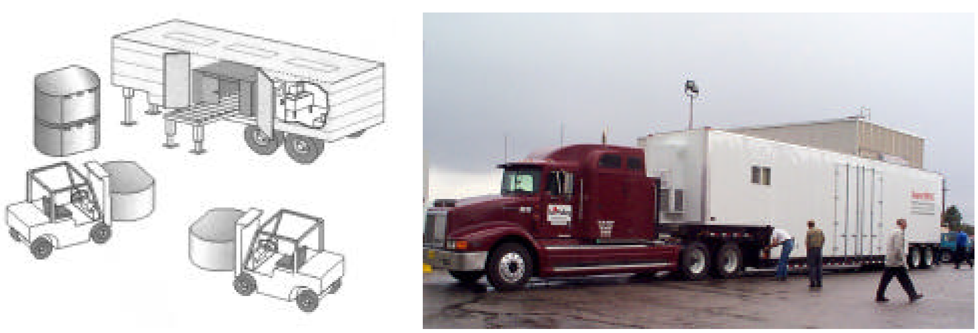

FIG. 1. Standard Waste Box (SWB) loading configuration and SuperHENC trailer.

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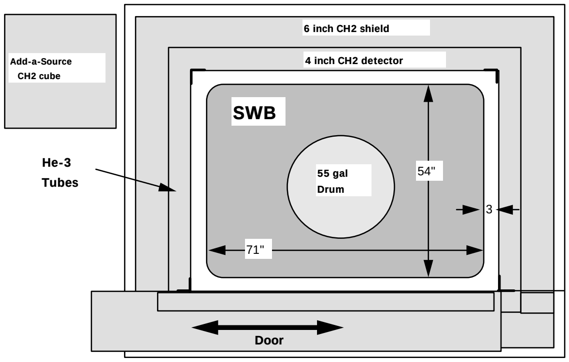

FIG. 2. Top view of the SuperHENC detector body showing the exterior CH_2 shielding and the SWB or drum sample locations. The SWB has 7.5 cm of clearance to the walls and roof.

The assay chamber is sized to accommodate at most an SWB; it is intended to measure one SWB or one 208-L (55-gal) drum at a time, but not the larger waste crates such as the IP-2 box. Figure 2 shows a diagram of the detector body with an SWB container inside the measurement cavity.

SuperHENC measures the $^{240}Pu_{eff}$ content using passive neutron time-correlation counting and calculates the total plutonium content using accepted isotopic knowledge or independent gamma-ray spectroscopy measurements. The SuperHENC is operated by a custom tailored version of the LANL NCC software.[2] The software controls the data collection and analyses and the metrical movement, and contains the calibration and error handling functions. After the passive neutron count, a small ^{252}Cf add-a-source (AAS)[3] used for matrix correction is moved into the assay chamber automatically to pre-specified positions. After about a four-minute AAS count, the source is then returned to its shielded storage area in the aft of the trailer. The chamber door opens and the package exits and is off-loaded using a fork-lift. The entire assay system is constructed to minimize presence of high atomic weight material, minimizing background from cosmic ray induced neutron cascades.

The package typically then is loaded on the SGEAS turntable, and a 20-min (10 min per side) gamma measurement is performed. The SGEAS software[4] uses the BII Efficiency-Times-Attenuation technique, which is optimized for the sparse counting signal characteristic of large packages and dense matrices. The SGEAS software also automatically integrates the SuperHENC neutron data with the gamma data collected, and calculates the sample-specific MDC from the SuperHENC data. Automatic quality checks on the gamma result are also performed.

3. CALIBRATION & VALIDATION

The calibration was conducted in several steps: (1) constructing a Monte Carlo Neutron/Photon (MCNP) model for the system, (2) mapping chamber response with a neutron source, (3) obtaining calibration measurements and establishing the coincidence calibration curve, (4) establishing the AAS correction factor calibration, (5) implementing background reduction techniques for samples with high backgrounds and low signals, and (6) validating the calibration on different plutonium standards at the final location (RFETS). The calibration and validation measurements are discussed in more detail below.

A. Calibration Measurements

The calibration used on the SuperHENC is fundamentally very simple. It measures the $^{240}Pu_{eff}$ content using passive neutron time-correlation counting ("standard" neutron doubles analysis) and calculates the total plutonium content using accepted isotopic knowledge. In waste measurements, corrections are made to the count rate to account for the count rate difference if the truncated doubles (ones)[5] are used, background and matrix effects, to return the sample measurement to the empty SWB case. In the final analysis, all measurements use the doubles calibration curve described in this section to determine the sample $^{240}Pu_{eff}$ mass.

As the empty SWB case is the baseline case, the SuperHENC was calibrated with plutonium standards placed in an empty SWB in the detector. Six replicate measurements at eight different plutonium loadings were taken at the volume average position for all calibration measurements. The volume average position is a virtual surface where about half of the volume of the SWB is inside the surface and half outside the surface. In practice, this surface is at about one-third of the distance between the outside and the center of the SWB. For an empty SWB, any central position qualifies for calibration because of the uniform efficiency, and the calibration standards were randomly placed in the central region so that they did not get closer than 5 cm to each other to avoid multiplication. Measurements were taken for a set time period of 1000 s. A blank (no plutonium) set of six replicates was also taken. A set of certified PDP-type standards was used to give the final calibration for the doubles rate.

No matrix corrections were applied to the calibration data because the standards are measured in an empty SWB. The net count rate was determined after the subtraction of the cosmic ray background. Because there is negligible multiplication over the plutonium mass range of interest (0–350 g) and no neutron self-shielding (due to exclusive use of the passive mode), the calibration curve is expected to be a straight line through the origin. In principal, one standard can establish the calibration slope with a zero intercept.

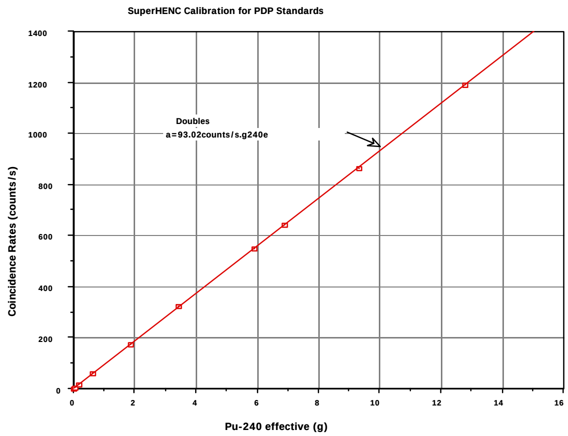

FIG. 3. Doubles calibration for a wide range of plutonium standards. The maximum plutonium mass was 187 g for the available PDP-type standards.

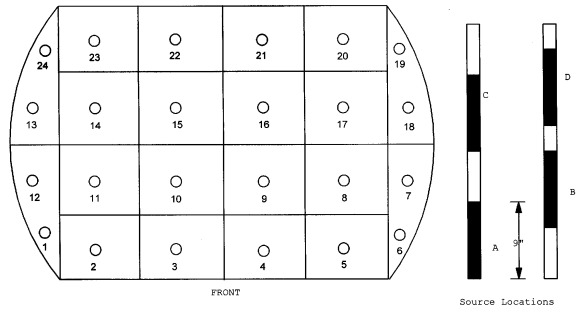

FIG. 4. Diagram of SWB matrix cubes (60) with 24 vertical tubes for the introduction of plutonium standards. The 24 tubes and 4 vertical heights (A, B, C, and D) provided 96-source position options for the positioning of PDP-type standards.

Figure 3 shows the calibration of SuperHENC using PDP-type standards with a mass loading from 0.5 to 187 g of plutonium. The doubles curve has a slope of 93.02 counts/s•g 240e. This line is the calibration curve used for all matrices and all container types. As stated above, matrix-specific corrections are handled in the count rate adjustment using the AAS or the multiplicity matrix corrections. Complete equations for calculating final assay values are in the software documentation.[2]

B. Validation Measurements

Validation measurements were collected on a non-interfering matrix (empty) standard for WIPP method performance demonstration, and on a variety of material types (light metals, mixed matrices, and plastics) that are representative of the waste stream at RFETS. Six replicates on three plutonium loadings (1, 10, and 320 g) were collected. Sources were located in the approximate volume average position. This plutonium range characterized the entire expected range, from near-MDC levels to the SWB criticality-loading limit. The matrix standards are described below, and a summary of the validation data presented.

C. Validation Matrix Standards

Figure 4 shows a diagram of the standard SWB matrix cubes designed by INEEL. There are 24 vertical tubes that can accommodate the 5-cm diameter PDP-type standards. There are three layers of the cubes for a total of 60 cubes or modules. These modules have been filled by INEEL to represent RFETS materials such as IDC 480 (mixed metals), IDC 330 (dry combustibles), and IDC 337 (plastics).

Decontamination & decommissioning (D&D) activities at RFETS have resulted in the dismantlement and size-reduction of numerous glove boxes, process piping, tanks, and other equipment. RFETS has traditionally segregated its waste streams into very narrowly defined IDCs. The benefit of less segregation (in terms of human exposure, cost, schedule, and difficulty) was recognized early in the D&D program. Consequently, two new TRU mixed matrix IDCs for D&D activities were defined: IDC 3010 (inorganic matrix with less than 10% by weight of organic constituents), and IDC 3011 (inorganic matrix with greater than 10% by weight of organic constituents). As these IDCs represented a substantial change in RFETS' way of doing business, MC&A qualification of these materials was specifically required. Mock-up standards of IDC 3010 and IDC 3011 were constructed by combining cubes of metal and plastic matrices to achieve 10% and 30% by weight organic content, respectively. Qualification data on these new mixed materials is presented in the next section.

D. Validation Measurements

Table 1 summarizes the measurements taken at RFETS on independent plutonium standards to validate the calibration. All the validation measurements used RFETS PDP standards that were traceable to NIST. None of the validation standards was used to establish the calibration curve.

Data was collected with the standard operating procedure and assay parameters used in routine operations. All the measurements included in Tbl. 1 passed the applicable WIPP or MC&A data quality objective for 208-L (55-gal) drums.

E. PDP Results for SWBs

The first cycle of the SWB Performance Demonstration Testing was conducted in June 2001. Two samples were tested: a combustibles matrix box and a stainless steel box. Six SuperHENC and SGEAS replicates of each sample were taken and integrated in accordance with the standard operating procedure. Final

TBL. 1. Validation measurement summary.

Matrix (IDC)	1-g WG %R	Pu (0.91) %RSD	10-g WG %R	Pu (9.0 g) %RSD	320-g WG %R	Pu (320 g) %RSD
Metals (480)	140%	6.43%	109%	4.07%	102%	0.89%
Mixed (3010)	110%	3.46%	93.9%	1.03%	98.9%	0.82%
Mixed (3011)	91.9%	4.00%	94.6%	0.78%	102%	1.07%
Plastics[a] (337)	–	–	96.1%	2.08%	102%	2.17%
Zero[b] (000)	126%	13.42%	106%	1.49%	103%[b]	0.60%[b]

[a] Plastics were not evaluated at the 1-g level.

[b] The zero matrix box was tested at 1, 10, and 160 g per the WIPP WAC; the last column of data reflects the 160 g nominal compliance point.

TBL. 2. Performance Demonstration Program (PDP) Cycle 1 summary.

Matrix (IDC)	7-g WG %R	Pu %RSD	14-g WG %R	Pu %RSD
Metals (480)	140%	6.43%	109%	4.07%
Combustibles (330)	126%	13.42%	106%	1.49%

results are presented in Tbl. 2. The results indicated in Tbl. 2 pass the PDP SWB cycle targets.

4. MINIMUM DETECTABLE ACTIVITY FOR THE SuperHENC

The minimum detectable concentration (MDC) for the SuperHENC has been determined by applying group statistics to a sample of repeat assays carried out on blank waste matrices contained within an SWB. A blank matrix is defined as a simulated waste matrix that contains no added activity. The assay results that are obtained when such an SWB is placed within the SuperHENC chamber are characterized by the background conditions.

WIPP provides guidance for how the MDC of an assay system should be determined. This guidance states, “The MDC is defined here as that radioactivity concentration which, if present, yields a measured value greater than the critical level with 95% probability, where the critical level is defined as that value which measurements of the background will exceed with 5% probability.” This definition is best represented graphically as in Fig. 5.

WIPP requires that the MDC be based on a determination of the detection limit as discussed above. The determination of the MDC must also account for interferences from different matrix conditions or radiation backgrounds that occur in the waste. In the case of the SuperHENC, the largest impact on the MDC are the neutronic properties of the matrix and specifically the effect that these have on the background. The effects of the matrix properties on the MDC are accounted for by calculating a matrix-specific MDC. Additionally, it is important to account for the effect of altitude on the MDC; this effect is caused by the fact that the cosmic ray background varies with altitude. To account for this effect, the MDC for each matrix-specific will be based on measurements of the background recorded at the same location as will be used for routine assays. In the calculations that follow, it has been assumed that the standard deviation does not vary with WGPu loadings in the gram range from zero to L_D.

The assays used to quantify the MDC for each matrix type were acquired by RFETS operators using the normal operating procedure. This ensured that the variability observed in the data set was representative of the variability that one would expect during normal operations. It does however lead to the observation that the variability in the background measurement, which is updated for each 4 hours of operation, will not be adequately captured. This is a consequence of the fact that the seven repeat assays for each IDC were acquired over time periods of about 4 hours that each required only one background measurement. To account for this, the statistical factor k used in the equations below has been set to 2.325. This factor is required to account for the fact that none of the variability in the background assays has been captured; instead, the only manifest

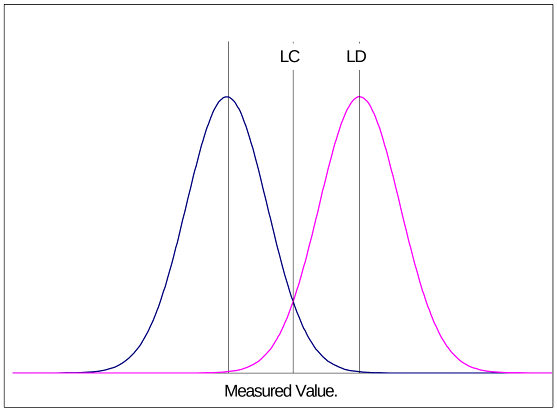

FIG. 5. The critical level L_C is defined where there is 95% confidence that measurements of the background are below L_C. The detection limit L_D is defined such that for a true value that is equal to L_D there is 95% confidence that the measured value is greater than L_C.

variability is in the assays of the blank. This procedure is equivalent to setting the detectability limit to be a factor 4.65 times greater than the standard deviation (sigma) of the seven repeat runs of the blank.

Finally, the effect of matrix composition on MDC has been considered. It is clear that the composition of a waste matrix has a significant effect on the background. The SuperHENC uses a specific surrogate for each matrix type and then makes a correction for the difference in weight between the unknown waste crate and the corresponding surrogate. The background correction routine involves regular assays of an empty SWB background; these results are then scaled to determine the background for the corresponding surrogate. There are three sources of uncertainty that arise as a result of this background correction technique:

- The uncertainty in weighting the crates has been demonstrated to be ~1.5%.
- The scaling factors used to determine the matrix-specific background based on the measured empty SWB background have been measured with an uncertainty of <1%.
- The final uncertainty component addresses the extrapolation from the surrogate to the unknown waste sample; it is the most involved aspect and will, therefore, be discussed in detail below.

To estimate the third uncertainty component, listed above, we will look at the equation used to calculate the background for the unknown waste sample. The equation used is:

$$B_U = B_{SWB\ 000}\left[a + bm\right]$$

where:

BU is the background associated with the unknown waste sample

a is the ratio $\dfrac{B_{SWB\,000}}{B_{SWB\,000}}$, where BSWB000 is the background associated with an empty SWB. This ratio, by definition, has a value of 1

b is given by, $\dfrac{\left[\dfrac{B_S}{B_{SWB\,000}} - \dfrac{B_{SWB\,000}}{B_{SWB\,000}}\right]}{M}$, where BS is the background associated with the surrogate matrix standard and M is the net weight of the surrogate matrix standard in kilograms

m is the net weight of the unknown waste crate in kilograms.

The background ratios have been determined by taking the average of four repeat measurements and the associated uncertainty has been estimated as about 1% based on the relative standard deviation of the repeat measurements.

The difference in background between the surrogate matrix and the unknown sample is manifest in the bm term, given in the equation above. A sensitivity analysis has been carried out to assess the impact of an error in this term on the estimate of the background associated with the unknown waste matrix. Two cases have been considered; these represent assumed errors of 5% and 15% in the bm term. The uncertainty associated with weighing the crates of ~1.5% is included in these assumed uncertainties and the uncertainty in the background ratios, <1%, is insignificant in comparison with the assumed uncertainties and has therefore been ignored.

The results of this analysis are summarized in Tbl. 3. This table includes the errors in both the WGPu mass and the activity concentration that result from the 5% and 15% errors in the bm term.

A 15% error in the bm term is considered to satisfactorily capture the uncertainty between the surrogate matrix and unknown waste samples. This uncertainty, referred to as the σ_F, is included in the determination of MDC by adding it in quadrature to the observed standard deviation in the replicate assays.

Equations

Critical level $$L_C = \overline{B} + \sqrt{(k \times s_R)^2 + s_F^2}$$

Detection limit $$L_D = \overline{B} + 2 \times \sqrt{(k \times s_R)^2 + s_F^2}$$

MDC $$MDC = \frac{L_D \times Act_{spec}}{W_{net}}$$

where

$\overline{B}$ is the average background corrected assay value recorded from repeat assays of the blank.

σ_R is the standard deviation of 7 repeat assays of the blank matrix.

σ_F, is the uncertainty due to matrix composition.

k is a statistical constant that relates to the one tailed 95% confidence level when a normal distribution of assay results is assumed.

W_{net} is the net weight of the blank matrix.

Act_{spec} is the specific activity of the isotope or mixture of interest.

Quantification of MDC

Table 4 lists the results of the assays used to quantify MDC. The data corresponding to IDC 480, IDC 330, IDC 3010, and IDC 3011 are summarized in Tbl. 5. The RSD for the empty SWB000 has also been determined and is included in the table since it is used to calculate sample-specific MDCs.

The total MDC is calculated by combining the statistical component "A" and the "F" component in quadrature in units of gWGPu. The conversion to nCi/g is dependent on the net weight of the unknown.

5. SUMMARY

The calibration of the SuperHENC was performed using NIST traceable plutonium standards in an empty SWB. The calibration is a straight line through the origin. For metals and other non-interfering matrices, the measured net doubles (or ones) rate is directly compared to the calibration line to give the $^{240}Pu_{eff}$ mass. The matrix corrections are only dependent on measured sample parameters.

The verification measurements were performed on independent standards at the final deployment site. All MC&A and WIPP criteria for assay of 208-L (55-gal) drums were met for the SWBs. Qualification of the new mixed-matrix waste streams crucial to the successful D&D of RFETS was demonstrated, achieving a new milestone in waste management at RFETS. PDP Cycle data for the first box assay also met certification criteria.

TBL. 3. MDC uncertainty component from surrogate matrix.

	Change in WGPu mass (g)		Change in activity concentration (nCi/g)	
IDC	5%	15%	5%	15%
480	0.054	0.162	7.2	21.6
330	0.014.	0.041	3.6	10.9
3010	.037	0.099	4.9	14.7
3011	0.014	0.042	2.5	7.5

A detailed MDC assessment was performed and demonstrates that the SuperHENC is capable of performing TRU/LLW sorting for the SWB waste streams of interest at RFETS.

The SuperHENC/SGEAS assay system is now fully qualified for MC&A measurements, fully certified for WIPP measurements, and has demonstrated TRU/LLW sorting capability. Thus, the design basis for the instrument has been achieved, and the assay system successfully deployed for all intended uses.

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2. W. Harker and M. Krick, *Mobile, High Efficiency Neutron Counter Software Design Document*, LA-CP-00-446 (LANL, 2000).
3. H. O. Menlove, *Passive Neutron Waste Drum Assay with Improved Accuracy and Sensitivity for Plutonium Using the Add-a-Source Method*, JNMM 17, 17–26 (July 1992).
4. P. Simpson, *SuperHENC Gamma Energy Analysis System Calibration and Validation Plan*, BII-5111-C&VR-001 (BNFL Instruments, 2000).
5. M. S. Krick, "Thermal Neutron Multiplicity Counting of Samples With Very Low Fission Rates," Nucl. Mater. Manage. **XXVI**, CD-ROM (1997).

Table 4(a). Results of the assays used to quantify MDC – replicates.

Case (IDC / Matrix / Net Mass (kg))	Replicate WGPu (g)						
	1	2	3	4	5	6	7
Case 1: IDC 480; Matrix: light metals, no voids; Net mass = 599 kg	-0.0137	0.0158	0.0193	-0.0243	0.0018	0.0252	0.0233
Case 2: IDC 330; Matrix: dry combustibles, no voids; Net mass = 300 kg	0.0861	0.1144	0.0857	0.0608	0.0108	0.0628	0.0488
Case 3: IDC 3010; Matrix: mixed metals, no voids; Net mass = 535 kg	-0.1005	-0.1215	-0.0635	-0.0205	-0.0462	-0.0814	-0.0467
Case 4: IDC 3011; Matrix: mixed metals, no voids; Net mass = 449 kg	-0.1149	-0.0592	-0.0795	-0.0802	-0.038	-0.1002	-0.0449

Table 4(b). Results of the assays used to quantify MDC – results summary.

Case (IDC / Matrix / Net Mass (kg))	Results Summary								
	Average WGPu (g)	Adjusted average WGPu (g)	SD WGPu (g)	Critical level WGPu (g)	Detection limit WGPu (g); (4.65 σ)	Replicate analysis (nCi/g)	F factor WGPu (g)	F factor (nCi/g)	MDC (nCi/g)
Case 1: IDC 480; Matrix: light metals, no voids; Net mass = 599 kg	0.0068	0.0000	0.0226	0.05262	0.10523	14.05	0.162	21.6	25.8[a]
Case 2: IDC 330; Matrix: dry combustibles, no voids; Net mass = 300 kg	0.0671	0.0000	0.0384	0.08927	0.17854	47.61	0.041	10.9	48.8
Case 3: IDC 3010; Matrix: mixed metals, no voids; Net mass = 535 kg	-0.0686	0.0000	0.0407	0.09456	0.18912	25.3	0.0986	14.7	29.2
Case 4: IDC 3011; Matrix: mixed metals, no voids; Net mass = 449 kg	-0.0738	0.0000	0.0329	0.07659	0.15318	27.3	0.042	7.5	28.3

a. The MDC quoted in Case 1 is calculated using WGPu isotopics (including approximately 30-year ^{241}Am in-growth) and the net weight of the simulated waste matrix blank that was used for the replicate assays. This MDC has been calculated with zero substituted for the average WGPu mass. The justification for this substitution is that a background corrected measurement of the background should, by definition, be zero.

TBL. 5. MDC data summary.

IDC	IDC Matrix weight (kg)	Sigma (gWGPu)*	4.65 sigma (gWGPu)*	F sigma (gWGPu)	MDA for Surrogate (gWGPu)	MDC for Surrogate kg (nCi/g)
SWB000	0	0.0244	0.113	0	0.113	N/A
IDC480	599	0.0226	0.105	0.162	0.193	25.8
IDC330	300	0.0384	0.179	0.041	0.183	48.8
IDC3010	535	0.0407	0.189	0.099	0.219	29.2
IDC3011	449	0.0329	0.153	0.042	0.159	28.3