

Application of NPL Radioactive Waste Packages Standards at AWE - 11004

T.J.Miller
AWE, Aldermaston,
Reading, Berkshire,
England, RG7 4PR
Tel: 0118 982 7449
Fax: 0118 982 5796
e-mail: Timothy.Miller@awe.co.uk

ABSTRACT

A variety of radioactive waste package standards, prepared by the National Physical Laboratory (NPL) in the United Kingdom (UK), have been applied to test the accuracy of a number of assay techniques routinely used at the UK Atomic Weapons Establishment (AWE).

The packages include standard 200 l waste drums, 320 l boxes and 1l cylinders containing either homogeneous or heterogeneous waste and photon emitting radionuclide distributions. Isotopes measured include Am-241, Cs-137 and Co-60 at concentrations ranging from sub Bq/g to a few Bq/g total activity. The assay techniques employed High Resolution Gamma Spectrometry (HRGS) (with and without transmission measurements) and gross counting using plastic scintillation detectors.

In most cases accurate results were obtained. Any discrepancies were easy to rationalise and regular checks, with the NPL standards, are now an important part of the ongoing Quality Assurance (QA) measurements that form an integral part of the assay process.

INTRODUCTION

During 2007 AWE participated in a gamma assay proficiency testing exercise run by NPL. This involved measurements on a NPL prepared 200 l waste drum, at 16 UK nuclear sites, without knowledge of the isotopic activities (1). A second exercise was run during 2009 (2) and there are plans to run a third exercise in 2011.

The exercises confirmed that most techniques gave accurate results and highlighted a few discrepancies that were easy to rationalise. Subsequently AWE procured the NPL drum, used in the 2007 exercise, together with other NPL waste package standards, in order to institute ongoing QA checks for a range of assay techniques.

This paper examines the NPL packages, measurement protocols and results achieved for each AWE assay technique that was used to measure the isotopic activities for each NPL standard. Discrepancies are highlighted and rationalised and early results for newly instituted ongoing QA checks are presented.

ASSAY MEASUREMENTS

Drum 2007

The radiograph (figure 1) was consistent with the information supplied by NPL. This indicated that the contents were five layers of 48 plastic bottles with each bottle containing 190 g of resin. Total contents mass was 62.4 kg and empty drum mass was 15.43 kg. Total activity (Bq/contents mass) was declared to be < 0.4 Bq/g for Am-241, Cs-137 and Co-60 combined (evenly distributed throughout the resin).

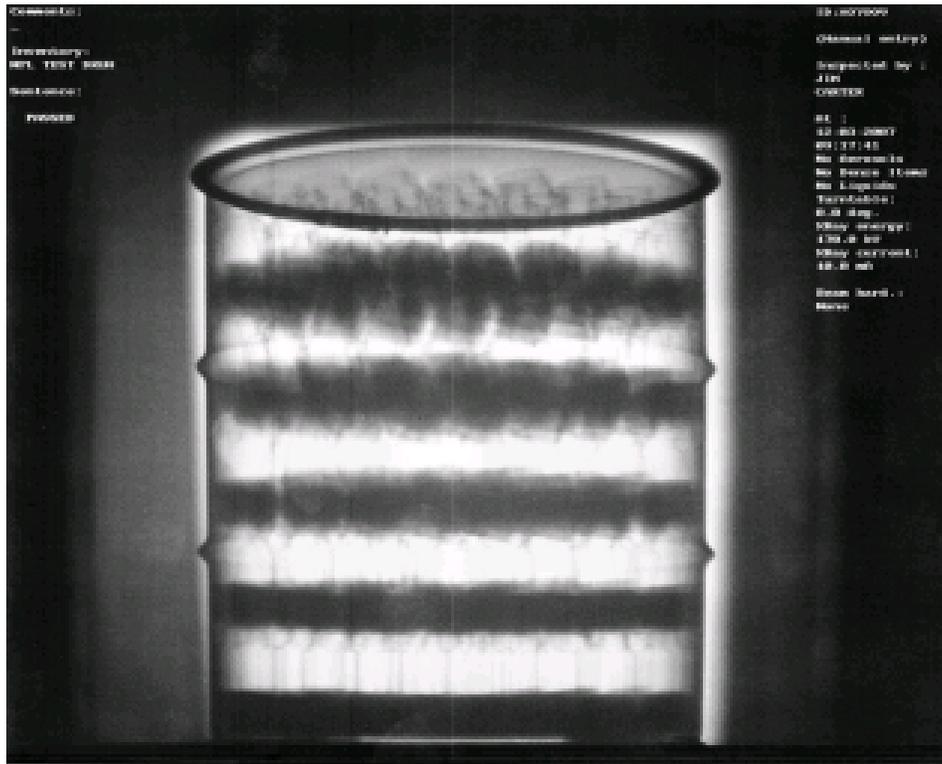


Figure 1. Radiograph of NPL drum 2007

The drum was monitored using two HRGS techniques: SNAP and AQ2. SNAP measurement protocol involved positioning a single HPGe detector either 30 cm or 60 cm from the centre/middle of the drum and counting until a few thousand net counts had been acquired for each photopeak. In contrast AQ2 was configured with three detectors (close to the top, middle and bottom of the drum) aligned with three transmission sources. The uniform matrix and activity distribution meant that drum rotation was optional.

The AQ2 operation was fully automated and produced a results summary for each isotope measured. In contrast SNAP required the detector calibration to be corrected (using SNAP software) for the drum dimensions, detector positioning, detector used, waste density and composition and drum wall thickness and composition. The reader is referred to the section entitled SNAP software for a detailed description of this process.

Four assays were done using SNAP. First, using detector A, calibration A and the non-standard drum wall thickness (0.9 mm) used by NPL. Second, a repeat following repackaging of the drum contents into a standard drum and purchase by AWE. Third, a repeat with a recalibrated detector A and fourth, a repeat with detector B and calibration B. Table 1 summarises the decay corrected results as a percentage of the activity declared by NPL.

Table 1 Percentage of declared NPL activity for drum 2007

Isotope	AQ2	SNAP1	SNAP2	SNAP3	SNAP4
Am-241	790	81	89	88	90
Cs-137	95	96	92	91	91
Co-60	75	97	94	94	95

The overestimation of Am-241 by AQ2 was attributed to the transmission measurement overestimating the drum wall thickness. All SNAP results were consistent and one of the recently instituted QA checks involves plotting the results achieved for the repackaged drum, using detector A and calibration A, over several months, on a Shewart chart (figure 2). The green, orange and red lines on the chart are the established mean with, +/- 2x SD and +/- 3x SD limits. The elevated result observed, at 260 days, was due to a drum movement in the facility. Otherwise results were within established limits for the first 8 results.

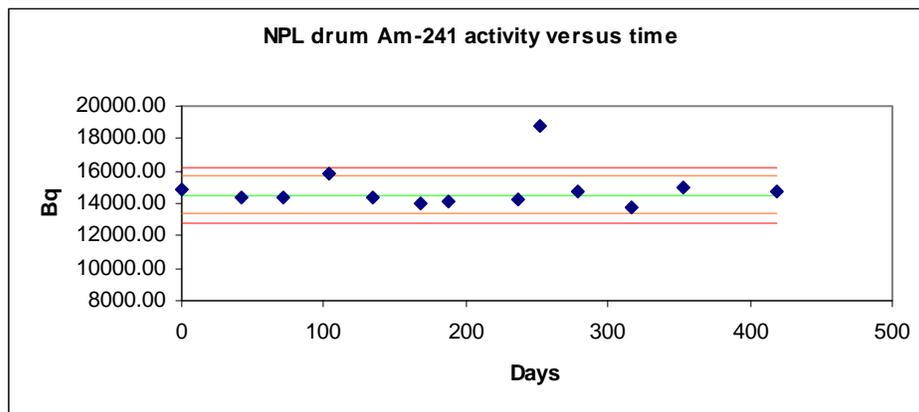


Figure 2 Shewart chart for NPL drum 2007

Drum 2009 A

Drum 2009A was filled with plastic bottles containing vermiculite to give an overall density of 150 kg m^{-3} . The Am-241/Cs-137/Co-60 activity ratios were different from the 2007 drum, but the total added activity was still below 0.4 Bq/g . This activity was contained within a single plastic bottle containing ion-exchange resin with a density of 800 kg m^{-3} . The drum radiograph shows that the location of the denser resin filled bottle was just inside the drum wall and one layer of bottles down from the top (figure 3).

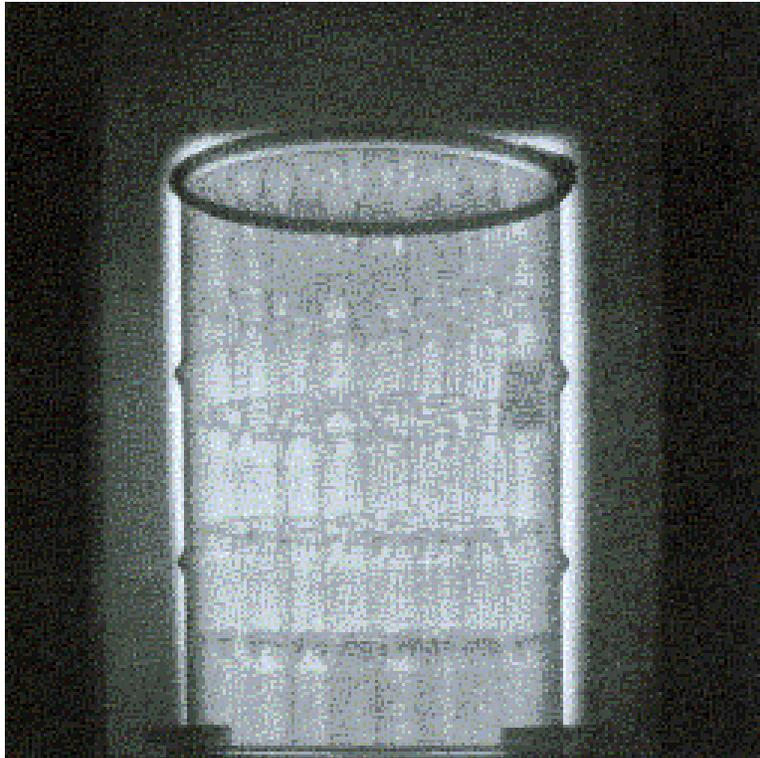


Figure 3. Radiograph of NPL 2009 drum A

Two techniques: SNAP and a Segmented Gamma Scanner (SGS) were applied and the drum was counted on a rotating turntable because of the non-uniform activity distribution. The SGS detector had a cadmium filter which precluded measurement of Am-241. Table 3 shows that both techniques gave a close match to the declared NPL activities.

Table 2 Percentage of declared NPL activity for drum 2009A

Isotope	SGS	SNAP
Am-241	-	94
Cs-137	86	105
Co-60	71	101

SNAP modeling was based on a uniform distribution of matrix activity so peripheral source location led to overestimation, but this was offset by the high local density of the 'hot spot' compared to the average matrix density.

DRUM 2009B

Drum 2009B was created by making 3 changes to drum 2009A. The active bottle was replaced with one containing more activity (a few Bq/g total), different isotope ratios and a location one bottle in from the drum wall. Again this is shown by the drum radiograph (figure 4).

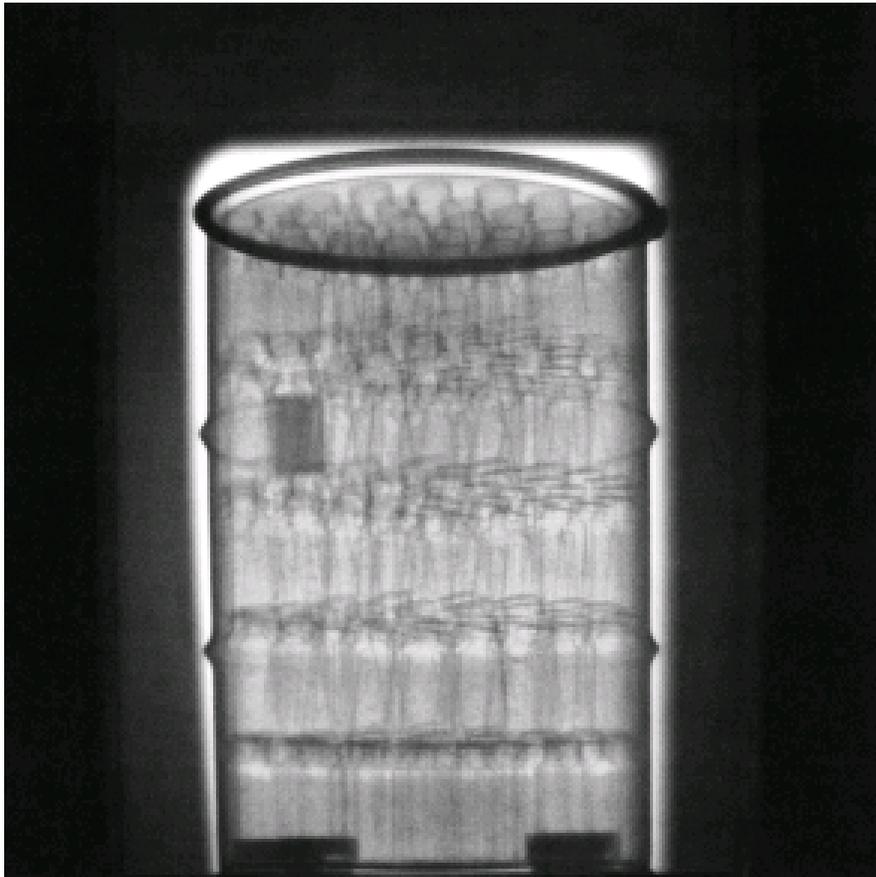


Figure 4. Radiograph of NPL drum 2009B

Comparative results for the SGS and SNAP are given in table 3.

Table 3 Percentage of declared NPL activity for drum 2009B

Isotope	SGS	SNAP
Am-241	-	74
Cs-137	78	94
Co-60	71	97

The SGS results were relatively unaffected by repositioning the 'hot spot' deeper within the drum. However, the SNAP results were all reduced compared to the more peripheral location in drum 2009A.

320 I Box

Two boxes were prepared: a blank containing inactive polythene slabs and a sample spiked uniformly with Am-241 (figure 5). This consists of a corrugated carton (62x62x84 cm), containing a series of nine filter papers (each 58x58 cm), each spiked uniformly with a standard solution of Am-241 and each separately laminated. The filter papers were interspersed within the carton between a series of ten inactive polythene slabs (each 60x60x8 cm). The total Am-241 activity was certified at 2857 +/- 12 Bq at 1 January 2010 with no other gamma emitting contaminants detected.



Figure 5. NPL 320 I box

The blank and sample were counted using SNAP at 30 cm standoff and a bag monitor (figure 6) that had been previously calibrated using a traceable Am-241

point source positioned at volume weighted locations within bags of low bulk density waste stimulant (3).

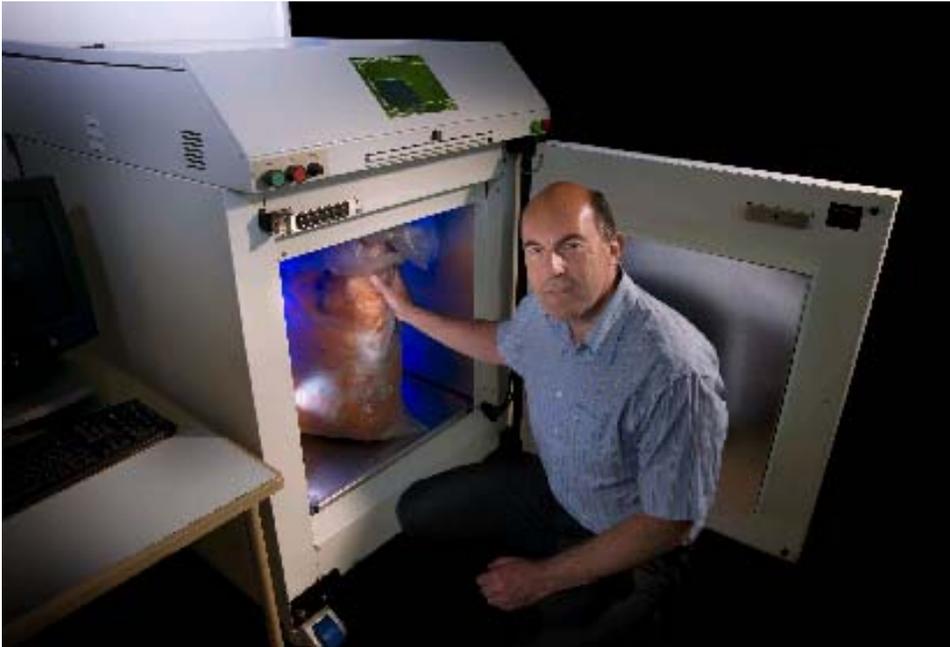


Figure 6. Bag monitor

The blank box gave no measurable activity with SNAP or the bag monitor and the comparative results for the sample are given in table 4.

Table 4 Percentage of declared NPL activity for 320 l box

Isotope	Bag monitor	SNAP
Am-241	101	105

One liter cylinder

An aqueous solution of Am-241, containing nitric acid at a concentration of 0.5 mol in one liter (figure 7), was assayed in a plastic bottle (17x9 cm diameter) using SNAP at 9 cm standoff. The certified Am-241 activity was 103.6 +/- 1.8 Bq at 1 January 2010. SNAP estimated 96.1 Bq (i.e. 93 %) of the declared activity.



Figure 7. NPL 1 l cylinder

SNAP SOFTWARE

The gamma spectrum, for the waste drum, is stored in the form of a Region of Interest (ROI) report (.Rpt) file containing the counting data for the photons of interest (e.g. Am-241 @ 60 keV) and imported into SNAP for radionuclide identification, modelling, assay calculation and reporting. Radionuclide identification can be performed using a full radionuclide library or a sub-library pertaining to the waste-stream of interest (e.g. plutonium). Each ROI in the spectrum is selected, by the analyst, from the library and the spectrum saved as a RPu file.

The RPu files can then be imported into the SNAP modelling. Early versions of the SNAP software have two models: a cylinder or a box. More recent versions have additional models, such as a disk. The dimensions of the cylinder (height and diameter) or the box (height, width and depth) pertain to the dimensions of the waste material within the waste package. For a completely filled waste drum this would be the internal dimensions of the drum that define the size of the item being assayed.

The detector location, relative to the waste, is defined by three measurements: the detector to item distance is measured from the detector end cap to the surface of the waste material; the detector height is measured from the base of the waste to the detector axis and the left of centre is measured from the centre of the waste to the detector axis. The detector calibration (e.g. Coldfinger, Old

Probe or New Probe) is selected together with the collimator position (e.g. flush with the detector end cap).

The waste material is defined by three or four variables: the matrix mass, primary matrix percentage by volume, primary matrix material and (if applicable) secondary matrix material. Three layers of shielding material and thickness may be applied, but for a 200 l drum 0.11 cm of iron is used with the other two layers set to 0 cm of 'none'.

Modelling is completed by entering the count time, height above sea level, detection limit required (i.e. Critical Level or Minimum Detectable Amount) and number of sides of the waste package counted (2 or 4) for computation of the Geometric Attenuation (GA) error. This is the percentage difference in activity between a uniformly distributed matrix and activity compared to a single point source of activity at a 'worst case' location.

Assay calculations are performed once all of the above modelling information has been entered. This gives radionuclide activities, based on uniform activity and matrix distribution, for each photon measured. All photon energies, for a given radionuclide, should yield consistent results. If not the modelling is adjusted to get the best agreement. For example, if the 60 keV signature from Am-241 is underestimating by a factor of two, compared to the 662 keV Am-241 photon, increasing the steel shielding thickness by 1 mm would give better agreement.

Another feature is the lump correction routine for uranium and plutonium (Pu) waste-streams. For example, the main photons from Pu are at 129 and 414 keV. Underestimation at 129 keV compared to 414 keV is often an indication of photon self-absorption within Pu. The software allows the analyst to progressively increase the size of the Pu until consistent results are obtained at 129 and 414 keV.

When the analyst is satisfied with the modelling the software can be used to generate an htm report file. This summarises all of the sample details, modelling and results for presentation to the customer as an electronic or paper copy

DISCUSSION

Any HPGe detector may be calibrated by Eberline Services (at LANL) for SNAP using the gamma spectra acquired from certified traceable sources positioned along the detector axis. The SNAP software is then used to correct the calibration, for the counting scenario of interest, based on uniform activity and matrix distribution. In the case of waste drum assay most of the modelling parameters, listed in table 1, are normally fixed and the only variable affecting the detector response is usually the waste matrix density. Hence, simple software

generated calibration curves (e.g. figure 8) may be used to achieve accurate results against certified waste drum standards.

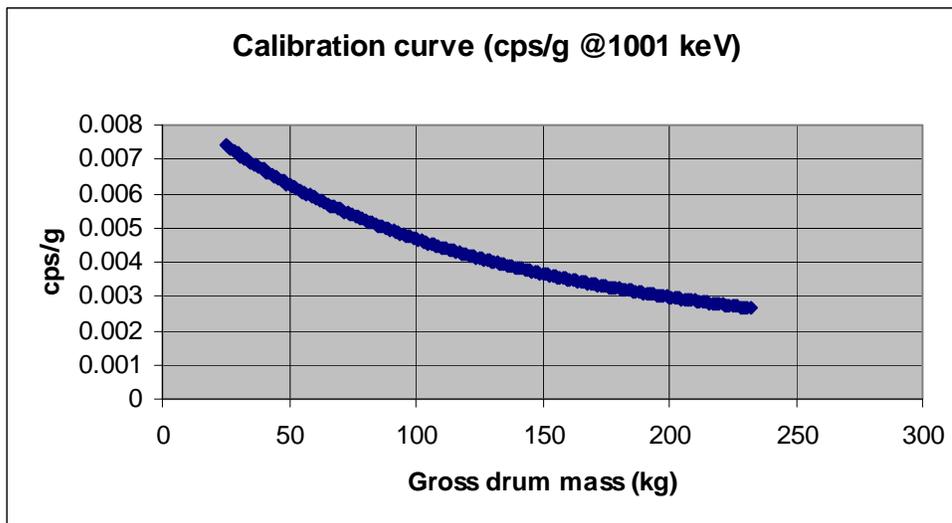


Figure 8. Software generated calibration curve

All of the results for the NPL 2007 drum were a few % below NPL activities, with the degree of underestimation reducing with photon energy. This observation was attributed to the relatively high density of the resin containing the activity (800 kg m^{-3}) compared to the average matrix density (300 kg m^{-3}) used by the SNAP modelling. Better agreement, between SNAP and NPL, would be expected if the matrix was truly uniform, rather than layered as shown in figure 1.

Results for drum A were relatively higher, but still within a few % of the NPL activities. The peripheral location of the active bottle tended to result in overestimation of activity because this was based on the SNAP uniform activity distribution modelling. However this effect was offset by the relatively high density of resin (800 kg m^{-3}) in the active bottle compared to the much lower average matrix density (150 kg m^{-3}) used by SNAP.

Drum B results were lower than drum A due to the repositioning of the active bottle deeper within the drum. As noted for the 2007 drum, underestimation was greatest at the lower photon energies.

It was notable that all 3 calibrations gave similar results for the 2007 NPL drum procured by AWE. The Am-241 values were higher than originally measured. This was attributed to the sensitivity of the 60 keV measurement to drum wall thickness since the drum was repackaged prior to purchase by AWE. The calibrations for detector A were within 1 % of each other despite the eight year period between the calibrations. The detector B calibration, applied to a more recently procured detector, was also within 1 % of the detector A calibration

despite the shorter detector length and lower counting efficiency at higher photon energies compared to detector A.

The results highlighted the effect of non-uniform matrix and activity distributions because SNAP modelling assumes uniform matrix and source distribution. Placing the activity in denser regions, further from the detector resulted in underestimation. However, the Measurement Good Practice Guide (4) states that any underestimation, even in the most severely attenuating matrix, is unlikely to exceed a factor of 2 or 3 if the activity is present in the form of even a small number of randomly positioned point sources. Hence, AWE has recommended that future proficiency testing exercises utilise drums containing a matrix and activity distribution that is a close match to those routinely encountered from waste generating facilities. Previous studies have indicated that the SNAP methodology is applicable to AWE wastes because non-uniform matrix/activity effects tend to cancel out when averaged over the whole drum (5).

CONCLUSIONS

The SNAP calibrations gave excellent results when applied to scenarios where the matrix and activity were truly uniformly distributed. Non-uniform distribution effects can result in over or underestimation. However, these effects are likely to cancel out when averaged over a drum containing randomly distributed contamination.

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