Collated Experience of the Use of Multi-Group Analysis (MGA) in Non-Destructive Waste Assay Systems – 15257

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ABSTRACT

The quantification of plutonium is an important aspect of non-destructive assay of Special Nuclear Material (SNM). The calculation of %^{240}Pu$_{eff}$ can be done through prior knowledge of the composition of the waste but more often is done through the analysis of a measured plutonium gamma-ray spectrum performed using an isotopic analysis code. In this work, the %^{240}Pu$_{eff}$ results from a variety of non-destructive waste assay systems in the UK are presented and compared using MGA v9.63C and v10. The body of the results include real measurement results of a variety of waste materials containing plutonium, the results of reanalysis of these initial waste measurements with changes to certain setup parameters and the results of reanalysis with a later version of MGA (v10). Additionally, measurements of test matrices with known source isotopics are measured and the results analysed and presented here. The use of the 'waste' flag parameter introduced to allow measurement of unusual spectra in v10 is also evaluated.

When using MGA as part of the waste assay characterisation process in automatic or batch mode, results have proven to be very dependent on the quality of the spectra measured and on the setup parameters that the code is reliant upon. With a modern version of the code, the likelihood of under-reporting the total plutonium content of a 200 litre/55 gallon drum using PNCC and MGA isotopics in combination is low. Additionally, the latest 'hardened' code will also reduce the number of cases where MGA fails to run and the use of default isotopics results in an over-pessimistic evaluation of the total plutonium mass.

INTRODUCTION

The quantification of plutonium is an important aspect of non-destructive assay of Special Nuclear Material (SNM). Non-destructive assay techniques used to measure SNM include Passive Neutron Coincidence Counting (PNCC) and High Resolution Gamma-ray Spectroscopy (HRGS). The PNCC technique exploits the fact that multiple neutrons from spontaneous fission in SNM are emitted essentially simultaneously. Plutonium in particular usually contains several isotopes that have large spontaneous fission yields; ^{238}Pu, ^{240}Pu, and ^{242}Pu so the observed coincidence neutron response is due to all three isotopes. ^{240}Pu is typically the dominant contributor for materials in the civilian reactor and weapons fuel cycles, so traditionally the strength of the spontaneous fission signal is expressed in terms of the effective mass of ^{240}Pu present (^{240}Pu$_{eff}$), which can be calculated as a linear sum of all three isotopes (see Eq. 1). However, to provide a total plutonium mass, this effective mass needs to be corrected by using the ^{240}Pu$_{eff}$ weight fraction (%^{240}Pu$_{eff}$).

The calculation of $\%^{240}Pu_{eff}$ can be done through prior knowledge of the composition of the waste but more often is done through the results of analysis of a measured plutonium gamma-ray spectrum performed using an isotopic analysis code. One code in particular is Multi-Group Analysis (MGA), which was developed for Nuclear Safeguards applications by Ray Gunnink and co-workers at the Lawrence Livermore National Laboratory (Canberra has since procured exclusive rights for this version of the code from Ray Gunnink). The MGA code analyses a number of regions of the gamma-ray spectrum, including the 90 to 110 keV region which contains a number of the highest yield but often strongly overlapping photopeaks. The precision of the MGA analysis is very dependent on quality of the spectrum, which is, for a detector with good energy resolution, strongly influenced by the counting statistics in the 90 to 110 keV region. The results of the analysis are then used to calculate $\%^{240}Pu_{eff}$, which in conjunction with $^{240}Pu_{eff}$ from PNCC measurement can be used to calculate the total plutonium content where $\%^{240}Pu_{eff}$ is calculated by (Eq. 1) [1]:

$$\%^{240}Pu_{eff} = k_{238}\%^{238}Pu + k_{240}\%^{240}Pu + k_{242}\%^{242}Pu \qquad \text{(Eq. 1)}$$

where the weighting factors k_{238}, k_{240}, and k_{242} are 2.66, 1.00 and 1.64 respectively as specified by the customer for the HRGS.

Note that $\%^{240}Pu_{eff}$ can be described in terms of an isotopic correction factor, F_{iso} as shown in (Eq. 2) below and used as part of the total plutonium calculation in (Eq. 3).

$$F_{iso} = 100/\%^{240}Pu_{eff} \qquad \text{(Eq. 2)}$$

$$\text{Total plutonium mass} = F_{iso} \times {}^{240}Pu_{eff} \qquad \text{(Eq. 3)}$$

In this work, the $\%^{240}Pu_{eff}$ results from a variety of non-destructive waste assay systems in the UK are presented and compared using MGA v9.63C and v10. The body of the results include real measurements of a variety of waste materials containing plutonium, the results of reanalysis of these initial waste measurements with changes to certain setup parameters and the results of reanalysis with the latest version of MGA (v10). Additionally, results of the measurements of test matrices and plutonium sources with known isotopics are analysed and presented here. The use of the 'waste' flag parameter introduced to allow measurement of unusual spectra in v10 is also evaluated.

The MGA code is a non-destructive analytical method of assessing the isotopic makeup of radioactive substances from their gamma-ray spectra. It has been widely used in nuclear waste treatment and handling in order to assess the quantities of nuclear materials such as plutonium's fissile isotopes (primarily ^{239}Pu), for nuclear material accountancy and safety concerns [2].

Many of the systems that use MGA as a method to determine $\%^{240}Pu_{eff}$ have a default set of isotopics defined so that, should MGA fail to provide results or should the quality of the results not meet certain figures of merit, then a conservative value of $\%^{240}Pu_{eff}$ is used in order to determine the total plutonium content. However, the use of a default $\%^{240}Pu_{eff}$ means that, for example, $^{240}Pu_{eff}$ mass result from PNCC would combine to produce in excess of 150 g total plutonium with only a few grams of $^{240}Pu_{eff}$ measured. Excessively large assay results such as these can cause issues with plant operation & require rework and they should therefore be minimised.

When using MGA as part of the waste assay process in automatic or batch mode, results have proven to be very dependent on the quality of the spectra measured and on the setup parameters that the code is reliant upon. With a modern version of the code, the likelihood of under-reporting the total plutonium content of a 200 litre/55 gallon drum using PNCC and MGA isotopics in combination is low. Additionally, later, 'hardened' code will also reduce the number of cases where MGA fails to run [3].

MEASUREMENT METHOD

The underlying data files and results of 1000 real assay measurements have been provided to Canberra UK (CUK) by Sellafield Ltd in order to verify the improvement in system performance of one of their HRGS systems used for the assay of drums of plutonium contaminated material (PCM) following a minor software modification. The HRGS assays 200 litre PCM drums that have arisen from operations at Sellafield. These drums, all of which were produced in areas containing plutonium of varying concentrations and isotopics, contained a variety of waste matrices and radionuclide compositions.

Description of the Drum HRGS

The Drum HRGS is designed to carry out an assay of the gamma-ray emitting nuclides present in PCM drums. The HRGS provides the measurement data used to determine the isotopic composition of the plutonium in the waste and also an inventory of activity of other radioactive isotopes of interest.

The Drum HRGS uses four electrically cooled high purity germanium (HPGe) detectors mounted on a fixed support structure to view a 200 litre drum on a turntable, which rotates the drum throughout the measurement. The detectors are collimated and have cadmium filters installed to reduce deadtime by reducing the count rate at 60 keV from drums containing significant amounts of [241]Am, see Fig. 1. The summed spectra four gamma-ray spectra (one for each detector) are summed.

The detectors are Canberra Broad Energy Germanium (BEGe) detectors covering the energy range of 3 keV to 3 MeV. The BEGe detector has a short, 'fat' shape designed to offer a high resolution at low energies (below 200 MeV) whilst also optimising efficiency below 1 MeV for typical sample geometries, both of which benefit the MGA algorithm.

Fig. 1. The four detector Drum HRGS used for the measurements. The left image shows the collimators pointing toward the drum cavity. The light gray rectangle above the white label is the Cd filter. The right image shows the electrical cooler.

Description of Drum HRGS System Software

The software is a collection of MS-DOS based executables which includes all the functions required to produce the assay result for the HRGS including handling item information, spectrum acquisition, MGA, analysing and reporting the results.

MGA v9.63

MGA v9.63C was introduced around the year 2000 and was the latest commercially available code at the time of the initial Drum HRGS installation. v9.63C was primarily intended for use within the Safeguards community for verification measurements and was not initially designed for analysis of spectra from waste drums and mixed-contaminant samples. Spectra from these sources can have low counting statistics, high attenuation and poor sorting, including high ^{241}Am content 'heat source' Pu samples such as ^{238}Pu and fission product contamination. Over its commercial life, the code was developed with v9.63H hardened to accommodate more complex spectra such as these but still having some shortfalls in capability. A key issue to note is that MGA was initially developed for use with low-energy germanium detectors (LEGe), which are of low sensitivity and have poor efficiency at higher energies. While acceptable for Safeguards purposes, this was inadequate for waste categorisation. The v9.63C code can fail to report a result for various reasons such as software and hardware limitations – primarily resolution

and some intrinsic limitations. MGA v10 was developed to improve the robustness of the analysis of challenging spectra.

Anomalous Drum Assay Results

Occasionally, the Low Energy (LE) region of the spectrum from a drum is obscured by high levels of interfering gamma-rays (e.g. from fission products also present in the drum) or is heavily attenuated by the presence of dense materials within the drum. In such cases, the HRGS software proceeds to run the MGA algorithm in its High Energy (HE) mode – using the higher energy gamma-rays produced by plutonium to determine F_{iso}. However, because these higher energy gamma-rays are emitted by plutonium at much lower abundances, the HE mode result is often of poor quality (with large uncertainties) or the analysis is not possible at all. When neither mode of MGA analysis gives a satisfactory result, the software automatically uses a conservative default F_{iso} value to calculate the total plutonium mass content of the drum. However, this situation is undesirable as it can cause significant overestimation of the plutonium content of the waste drum. In these cases, the mass content reported may exceed the safe fissile mass limit for a drum, even though the true mass of plutonium present is well below the limit.

Whilst carrying out reviews of these drums, Canberra found that the NQFIT, which is a normalised estimate of the goodness of the fit that MGA models to the true spectrum, could be drastically improved in a number of cases by a single parameter change. The parameter changed was the detector crystal volume which in v9.63C is used by MGA to define the generic detector efficiency curve. In many cases, using this curve as an initial starting point, the code is able to iterate towards a realistic detector efficiency curve, however, in the case of complex spectra with poor statistics, this was not the case and the use of a default value of 5 cm^3 was seen to contribute to many of the failures above. Changing this parameter to a more realistic value was seen to improve the performance of the code in these challenging cases. Note in v9.63C the detector crystal volume cannot be set to the true detector volume of 95 cm^3 as it is internally limited to a maximum of 20 cm^3 – a volume of 100 cm^3 was specified approximating the true detector volume.

DATA ANALYSIS METHOD

Detector Volume Change MGA v9.63C

Initially, the results of 27 anomalous drum assays were reanalysed individually offline with the detector volume parameter set to 100 cm^3 although v9.63 can only accommodate detector volumes up to 20 cm^3. These showed good results overall with all but one assay having the total reported plutonium content reduced. However, to implement the software change in the automatic HRGS program, a full analysis of the effect of the parameter change was required.

A random selection of the results from 900 normal assays by the Drum HRGS and from 101 anomalous drum assays, which consisted of multiple assays of the 27 drums discussed earlier, were used in an automatic reanalysis using the same operating system and MGA version with only one change in the setup (the detector volume parameter as discussed above).

MGA v10

Outside of this project, a separate installation of a Canberra Auto Q2 (AQ2) waste assay system has been carried out at Sellafield that uses a similar analysis method and the same BEGe detector types. As this installation was completed at a later date, it was supplied with MGA v10 which has greater code hardening for use in waste assay applications and allows a larger value to be entered and used for the detector volume parameter, consistent with that for detector types such as the BEGe, see [3].

MGA v10 with 'Waste' Flag Set

This is a specific parameter flag to allow poor or unusual spectra to be analysed without the software raising serious error flags and terminating the analysis. It is expected that if the spectra is of poor quality, then the results may be questionable as uncertainties may not be fully bounding. This feature has been tested offline using two different methods:

1. **Statistical testing**. This involved using a series of manipulated synthetic data where the isotopics and the number of counts in the region of interest are randomly varied from an original set of real measurements of test sources.
2. **Reanalysis of 1000 assays from the Drum HRGS**. The same method was repeated as for the detector volume change but using the MGA v10 code with the waste flag on.

RESULTS

Detector Volume Change MGAv9.63C

The results of the reanalysed data are compared with those for the original assay report files (called 'HR' files, x-axis) in Fig. 2. Generally the results are consistent however a number of outliers where the results are not consistent are highlighted for further discussion.
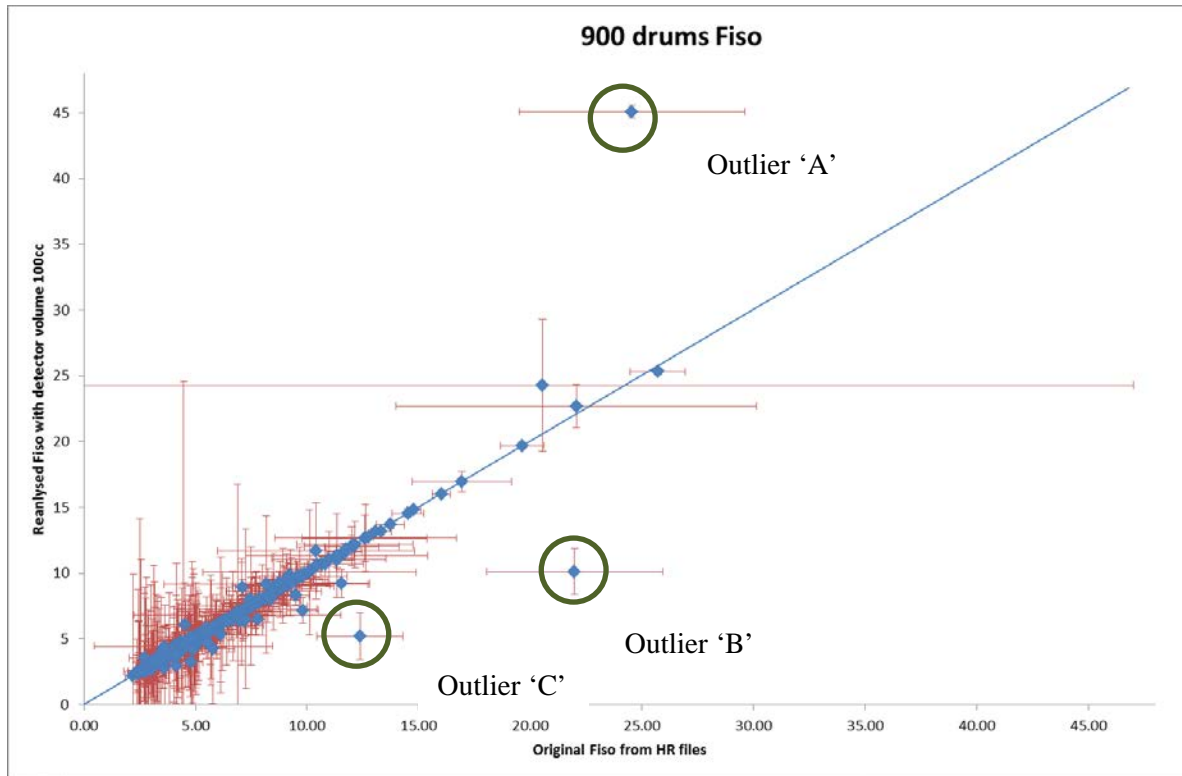
Fig. 2. Detector volume change effect on 900 randomly sampled assays for the F_{iso} (MGA v9.63C)

Detailed analysis of outlier 'A' as shown on Fig. 3 shows that the overall count rate is very low and that there is little ^{241}Pu and ^{241}Am present so the majority of the gamma-ray signal in the 94-104 keV region is result is dominated by those from ^{239}Pu.
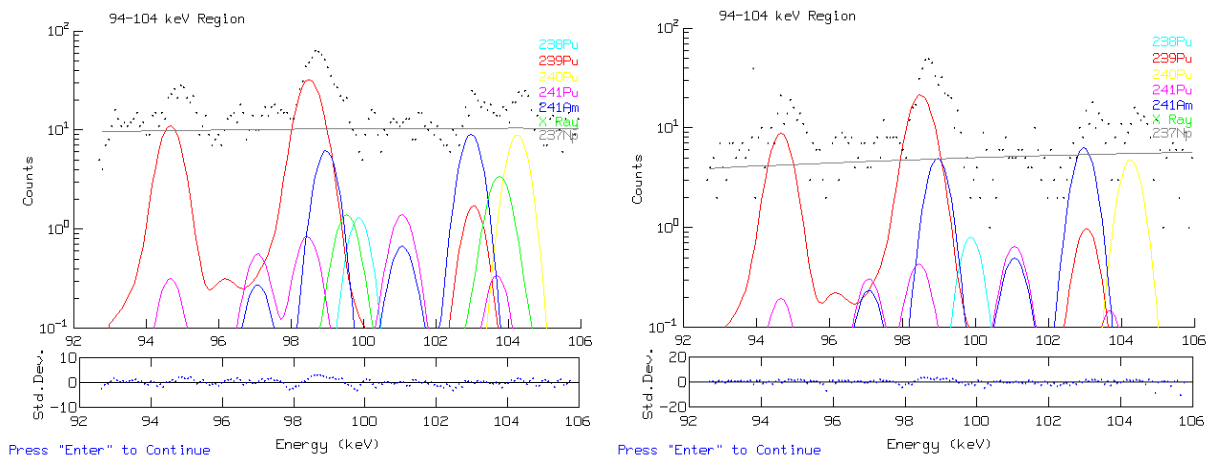

Fig. 3. Analysis of (LHS) outlier 'A' showing low counts in the 94 keV peak (count time 600 sec) and (RHS) analysis of outlier 'B'

It would be recommended to re-assay such a drum for at least 2400 seconds to improve the statistics in the 94 to 104 keV region. Currently, assay of material such as this where the plutonium composition is

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dominated by one isotope will be subject to variable results, especially when F_{iso} is large (i.e. small amounts of ^{238}Pu, ^{240}Pu and ^{242}Pu are present). The data from outlier 'B' in Fig. 3 follows the same pattern as that from outlier 'A'.

Outlier 'C' appears to be easily identified as a lower F_{iso} result when reanalysed with the larger detector volume. The plutonium gamma-ray photopeaks in the sum spectra (black dots) are clearly visible in the 94 keV to 104 keV region as shown in Fig. 4. This indicates that use of the correct detector volume can allow better peak fitting in the MGA v10 algorithm.
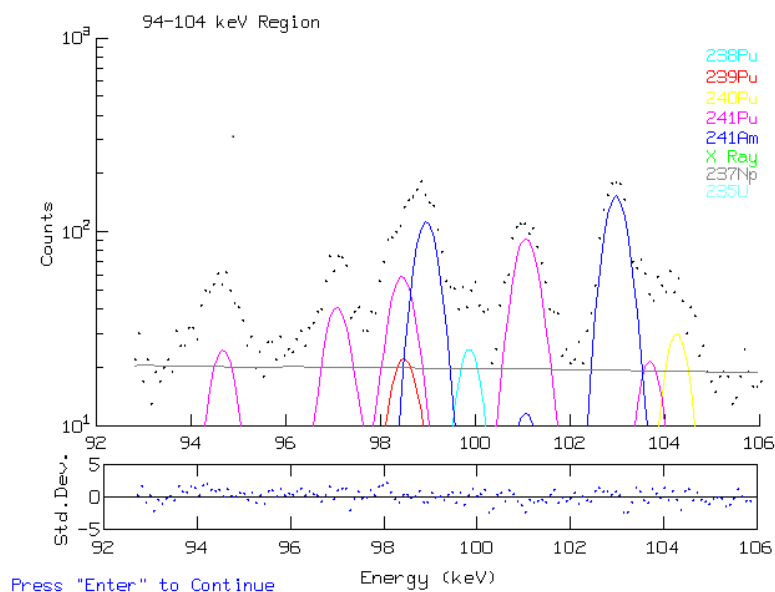
Fig. 4. Analysis of outlier 'C' (count time 600 sec)

Generally speaking, although there are a few outliers, none of these are unexpected as the change in detector volume parameter was thought to have more effect on spectra with low counting statistics and are therefore more challenging to analyse.

Fig. 5 shows the detector volume change effect on the anomalous assays with defaulted/unsuccessful analyses removed. Following the parameter change, no previously successful measurement has now failed but a number of previously unsuccessful measurements have now been successfully analysed although there is no value in comparing those to previously failed results. In Fig. 5 there are no results deviating greatly from a 1:1 ratio, the major trend is that they are indicative of waste from the UK civilian fuel cycle.

Although Fig. 5 shows good correlation between the original and reanalysed results, some data points have high uncertainty such as the highest point on the chart, marked as outlier 'D', examined in Fig. 6.
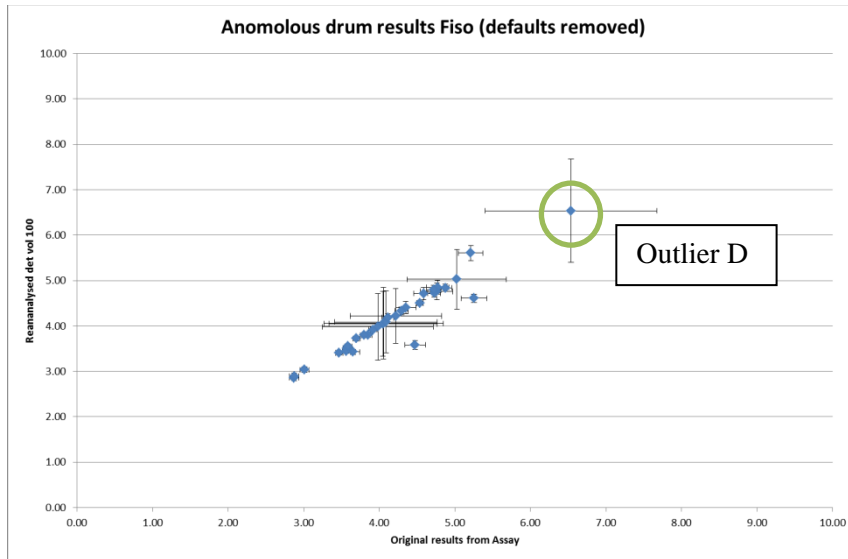
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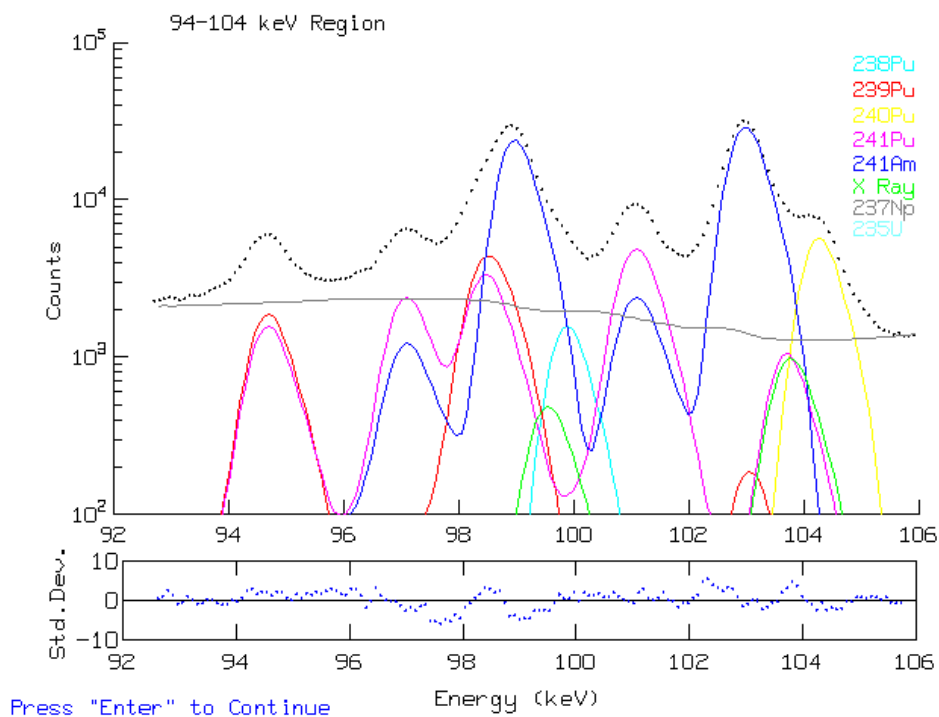Fig. 5. Detector volume change effect on 101 anomalous assays for the F_{iso} (MGAv9.63C)

Fig. 6. Analysis of outlier D showing high amounts of [241]Am (count time 600 sec)

The standard deviation from the modelled peak shape is shown at the bottom of Fig. 6 and this increases significantly where the strong peaks from [241]Am are present, masking plutonium isotope signals. The increased uncertainty in this measurement reflects the high [241]Am content. A number of the other data points in Fig. 5 show the same behaviour and have similar, high [241]Am content.

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Over the 1001 assays analysed with the revised detector volume parameter, the number where MGA fails to run are reduced from 405 to 365 which is a reduction of 9.88%. Specifically, of those anomalous assays where $^{240}Pu_{eff}$ is measured in the corresponding PNCC, the reduction in failed MGA measurements is 36.22%.

MGAv10

Statistical Testing

In a customer requirement as part of the formal testing of a new build, an AQ2 (three detector shielded HRGS) system was tested for a large number and range of different assay conditions. In order to fully test the algorithm's capability, 'Synthetic' assay raw data files were created which mimicked potential real assay data sets. The synthetic data was generated from real assay data files produced from the measurement of sources with known plutonium isotopic compositions. Different measured source assay data files were then randomly mixed in varying proportions such that the full range of F_{iso} could be analysed with varying count rates, thus also allowing the behaviour of the algorithm with low and high counting statistics to be seen.

The results of the measured F_{iso} vs expected F_{iso} can be seen in Fig. 8 where 405 of the assays gave isotopics results and 63 of them failed to run MGA. Generally good agreement is shown, although there are 6 cases where the measured value is more than 3 standard deviations below the expected value. The general trend however in Fig. 7 is consistent with the expected value as indicated by the blue straight line.
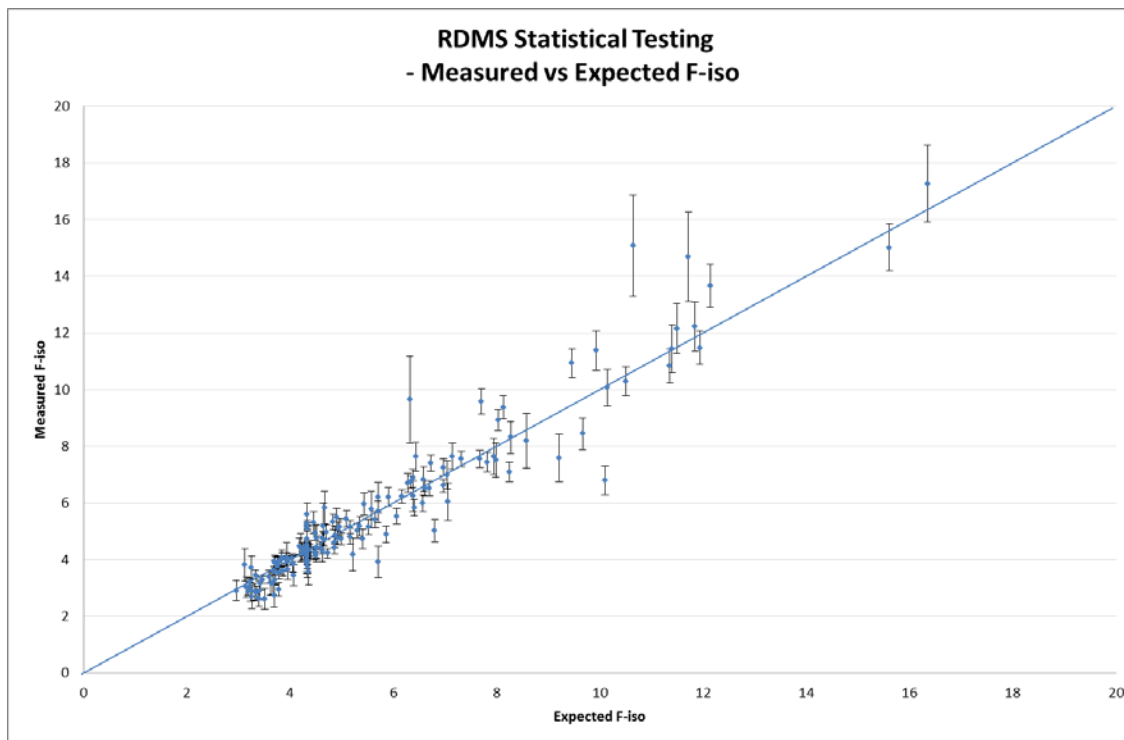

Fig. 7. F_{iso} results for statistical testing in the RDMS AQ2 system with error bars indicating 1 standard deviation.

In order to address the issue of under-reporting, the same data was then reanalysed with a change to the waste flag parameter. The waste flag parameter was set to 'on', which allowed the MGA algorithms to use more relaxed criteria when analysing waste or unusual spectra. The results in Fig. 8 now show a general bias of 1.103 in comparison to the expected results however, there are less instances of under-reporting and far fewer cases of MGA failing to run.
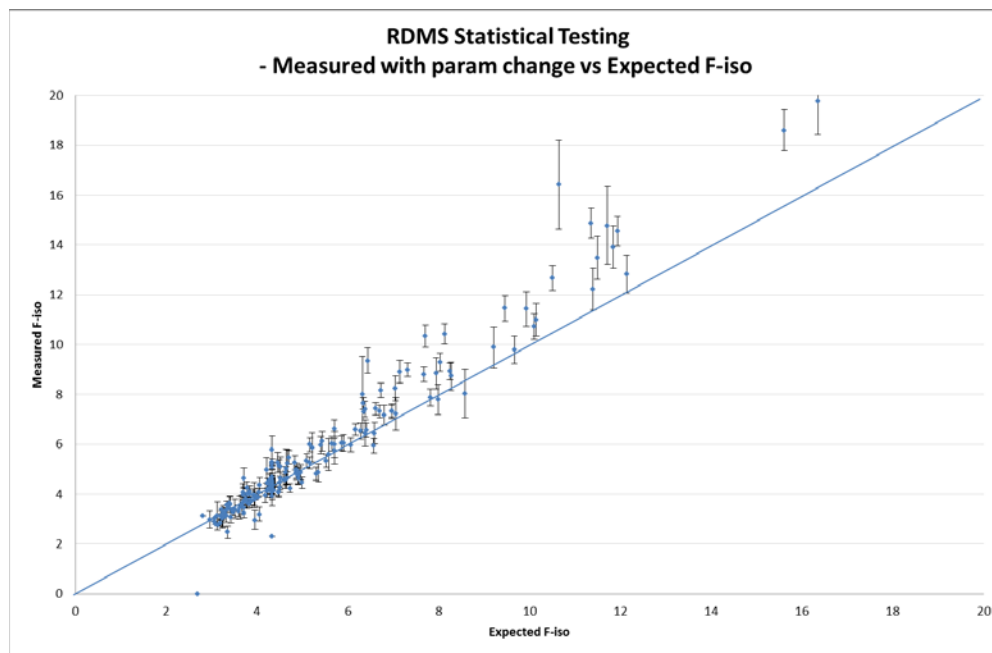

Fig. 8. F_{iso} results for statistical testing in the RDMS AQ2 system with the waste parameter changed to 'on'

The statistical testing results indicate that using the waste flag when measuring waste in automatic mode did reduce the number of assays where default isotopics were applied and may well increase performance, however, the results may be more questionable in terms of quality.

Table 1. Summary of $F_{isotopic}$ Results with Modified MGA Waste Flag

MGA Waste Flag setting	<-3σ	>+3σ	Failed to run
Waste=0 (off)	6	7	63
Waste=1 (on)	5	10	45

Reanalysis of 1001 Real Assays

The data from the same set of 1001 real assays were reanalysed using MGA v10 and the results are shown in Fig. 9.
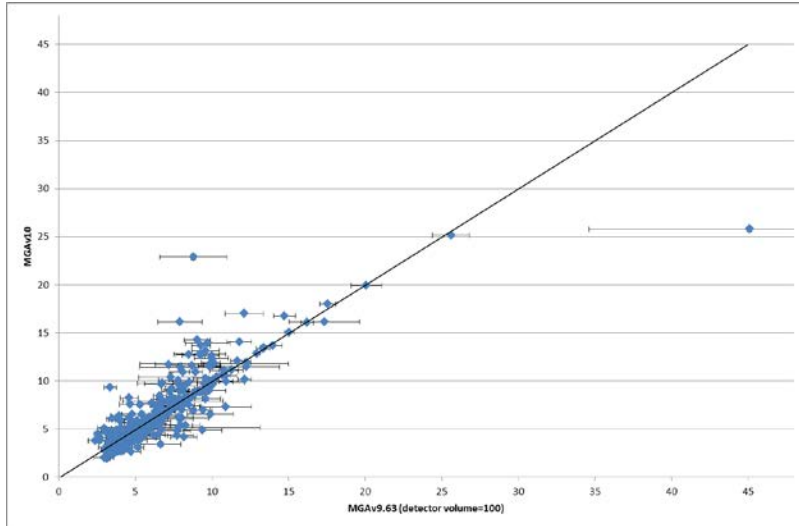
Fig. 9. F_{iso} for MGA v10 results in comparison to MGA v9.63C with the detector volume at 100cc

Note the wide spread of results seen in Fig. 9 where it can be seen that the uncertainties have reduced with the use of MGA v10. It is recommended that further measurements on known samples should be undertaken to determine the accuracy of lower reported statistics.

MGAv10 Waste Flag On

The data from the same 1001 assays were then reanalysed using MGA v10 with the waste flag on compared to the setting without are shown in Fig. 10.
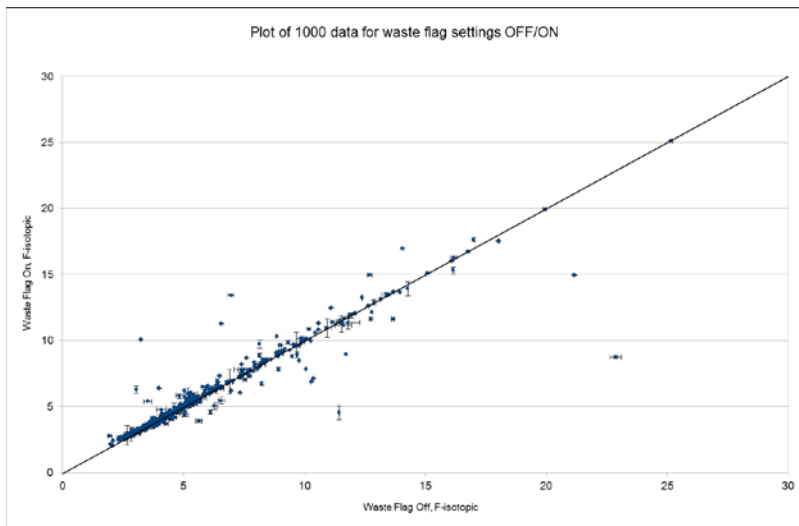

Fig. 10. F_{iso} plot for the 1001 assays with the waste flag on and off using v10 of MGA

Again a great variation in results is shown between the two settings with another reduction in the number of defaulted measurements. The uncertainties for MGA v10 have been reduced significantly as shown below in Table 2 where the mean uncertainty has reduced by a factor of 10 from v9.63C to v10.

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The detector volume parameter used for both MGA 10 runs was 100 cm^3.

Table 2. Summary of statistical uncertainties from all run sequences

F_{iso} uncertainty (units: % isotopic)	MGA9.63C -DV 5cc	MGA9.63C -DV 100cc	MGA10 -waste flag off	MGA10 -waste flag on
Mean	0.2932	0.3222	0.03158	0.04601
Median	0.05890	0.08514	0.01354	0.01703
Max	26.47	34.58	0.5142	0.9904
Min	0	0	0	0

Performance with Known Sources

The results reported so far raised questions about the variability between the results from MGA v9.63C, v10 and the effect of the waste flag in v10. An additional series of measurements has therefore been conducted with known standards.

The precision of the results from an analysis is strongly dependent on quality of the spectrum, which is, for a detector with good energy resolution, strongly influenced by the counting statistics in the 100 keV region. In this work we have taken the pragmatic approach of analysing a wide variety of spectra collected using a set of Plutonium Isotopic Determination Intercomparison Exercise (PIDIE) sources [4] under different conditions of collection time.

For three of the available sources, an initial measurement was performed with no attenuation. Due to the age of the sources used, this introduces a high count rate from gamma emissions of [241]Am. Further measurements were conducted with 1.8mm of cadmium placed in front of the detector to filter out the intense 60 keV gamma-ray from [241]Am. The use of this filter also closely reflects the fact that the installed Drum HRGS has cadmium filters installed in the front of the detector collimators as can be seen in Fig. 1. The three sources chosen for these measurements have isotopic compositions that cover the range of the PCM found in the waste typically seen in the customer's plant with F_{iso}=2.64, 4.72 and 16.48. The detector used for the measurements was of the same electrically cooled BEGe type with the same short shaping time settings to allow it to cope with the sometimes high count rates from the waste.

The results of the measurements are shown in Figures 11-13 where the analysis results are shown for the three different isotopic compositions respectively.
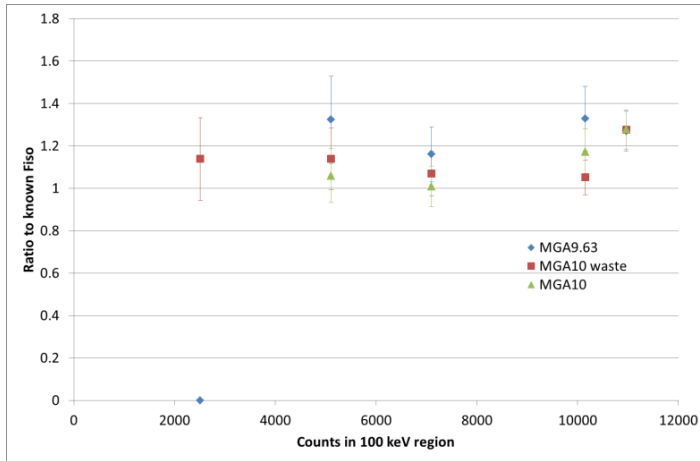
Fig. 11. Analysis for true F_{iso} of 16.48 with MGA v9.63, v10 and v10 with 'waste' flag set
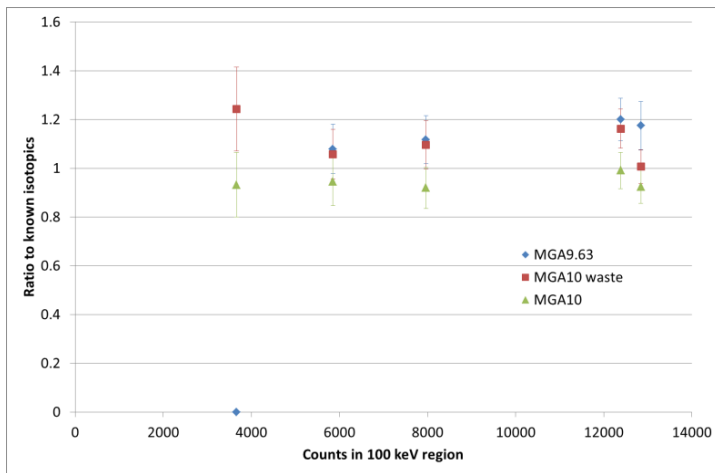

Fig. 12. Analysis for true F_{iso} of 4.72 with MGA v9.63, v10 and v10 with 'waste' flag set
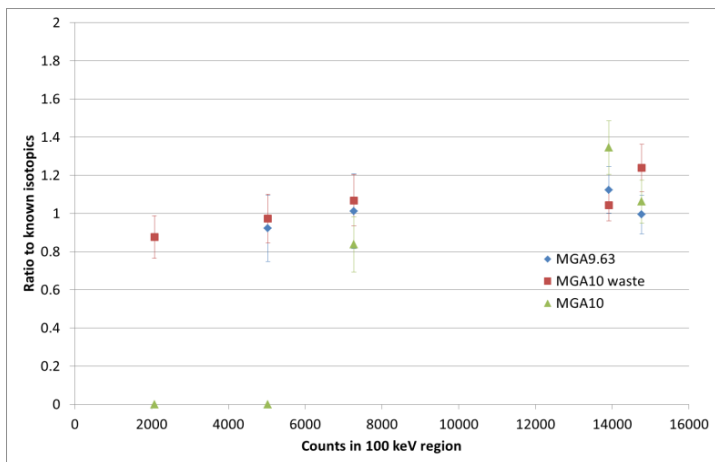

Fig. 13. Analysis for true F_{iso} of 2.64 with MGA v9.63, v10 and v10 with 'waste' flag set

Note the following from the three sets of measurements:

- The results show good comparison to the true F_{iso} and all results for all analyses that ran gave 100% agreement within 3 σ

- MGA v9.63C and v10 in normal mode both failed to run for three of the lower count rate spectra.

- MGA v10 with 'waste' flag set ran for all measurements with no apparent reduction in accuracy although precision did decrease as expected with lower counts in the 100 keV region.

- For some of the measurements in particular, there is a noticeable consistent difference between the results from MGA10 with and without the 'waste' flag - this was also seen in the results from statistical testing in Fig. 8. The effect is to overestimate the true F_{iso} with the 'waste' flag on by approximately 9%.

- Similar overestimates (of approximately 14%) are seen in the results from MGA v9.63C compared with the true F_{iso} value, however this is based on a small sample of measurements (only 15) so further corroborating measurements are needed.

- The reduction in the uncertainties reported with v10 in Table 2 is not replicated with these measurements

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CONCLUSION

A study has been carried out on the performance of the MGA v9.63C analysis software over a set typical plutonium waste spectra. The results were examined in detail and the accurate setting of the detector volume parameter was found to result in an improvement in performance in the automatic waste assay setting. Additionally the performance of MGA v10, the current commercially available release of MGA has been examined with the same data and improved performance was seen both in the reduction of the number of assays which failed to produce a plutonium isotopic result and a reduction in the associated measurement uncertainties reported. It's recommended that further investigation is undertaken with a wider range of measurement conditions with known plutonium standards in order to further evaluate the observations reported in this paper.

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