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# CHALLENGES OF NON-DESTRUCTIVE ASSAY WASTE MEASUREMENT* (U)

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## Abstract

Historically, the Savannah River Site (SRS) routinely produced special nuclear material (SNM), which provided stable measurement conditions for the non-destructive assay (NDA) methods. However, the main mission of SRS has changed from the production of SNM to the processing of waste and material stabilization. Currently, the purpose of processing is to recover the SNM from the waste and stabilization materials, much of which is from other DOE facilities. These missions are usually of a short duration, but require non-destructive assay (NDA) accountability measurements on materials of varying composition and geometric configuration. These missions usually have cost and time constraints, which sometimes require re-application of existing NDA methods to waste measurements. Usually, each new material or re-application of the NDA method to a different SNM campaign requires new standards and timely re-calibration of the method. These constraints provide numerous challenges for the NDA methods, particularly in the area of measurement uncertainty. This paper will discuss the challenges of these situations, mainly from a measurement and statistical point of view and provide some possible solutions to the problems encountered. Specific examples will be discussed for the segmented gamma scanner (SGS), neutron multiplicity counter (NMC) and passive neutron coincidence counter (PNCC), which are some of the most common NDA instruments at SRS.

## Introduction

A result of the changed mission at SRS is increased challenges in maintaining adequate method standards, calibration and uncertainties due to increased variation in waste and material stabilization measurements. There are also challenges due to increased time and cost constraints, and detection limit concerns for some instruments. These challenges and some possible solution will be discussed from a statistical point of view using specific NDA instruments as examples.

## Increased Variation

Previously, production of SNM provided a very consistent sample for measurement. The samples were consistent with regard to the amount of SNM and geometry, and fairly consistent for impurity level. Today's samples from waste and stabilization materials have little of these consistencies. A result has been an increase in the random and systematic variation seen from the NDA methods. As can be seen from the following two examples, one solution to the systematic variation is an adjustment to the instrument calculation parameters or to the final inventory based on the systematic effect.

The first example of increased variation is from the Mark 42 campaign of several years ago. Nine of these items were calorimetered to produce reference values and then run multiple times on the NMC. The percent differences from the calorimeter value are shown in Figure 1. For this data, which is based on 3600-second counts, the short-term random uncertainty (within item) is 2.3% and the systematic uncertainty is 6.2%. This short-term variation is within the percent relative standard deviation (%RSD) expected for plutonium metal (3.3%), or that expected for stabilization material (4.5%).[1] However, the random uncertainty seen in the deviations from the calorimeter values is 6.9%, which is above that of 4.5% expected for stabilization materials. This increased variation is thought to be caused by item to item differences. The 6.2% systematic uncertainty is above that expected for plutonium metal (1.8%) or stabilization material (1.5%).[1] The systematic uncertainty was particular troubling and is thought to be due to aluminum contamination. As a result, adjustments were make to some of the instrument multiplicity parameters to reduce the bias to near zero. This is detailed in reference 2.

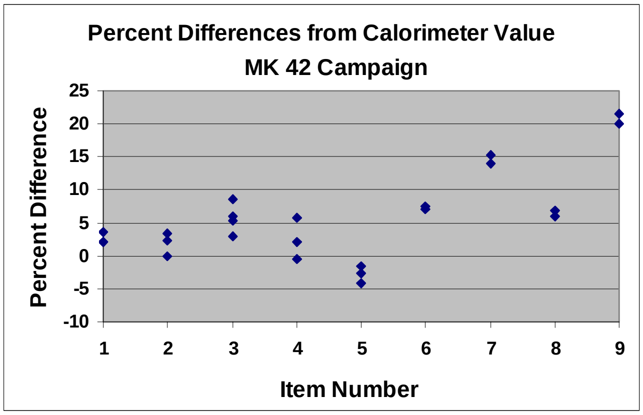

**Figure 1 – NMC Variation in MK 42 Campaign**

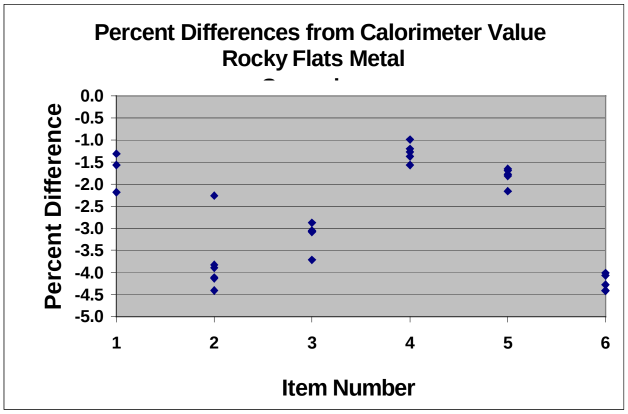

**Figure 2–NMC Variation in Rock Flats Metal Campaign**

A more recent example of increase variation is from the Rocky Flats metal campaign. To provide reference values, six items were calorimetered for comparison to multiple NMC measurements. See Figure 2. For these 3600-second counts, the short-term random uncertainty of this data was 0.4 %, the systematic uncertainty was 2.9% and the random uncertainty from the calorimeter values was 1.2%. Both random uncertainties are within the expectation of the instrument for metal, which is 3.3%, but the systematic uncertainty of nearly 3% is above the expected limit of 1.8% for metals with 3600 second counts.[1] The 2.9% systematic uncertainty was particularly troubling due to the large grams quantities of SNM involved. Later comparison of more items to the calorimeter confirmed the systematic uncertainty at the same level. A geometric effect is thought responsible for the systematic uncertainty. The campaign is continuing and final inventory levels adjustments may need to be made based on the systematic effect.

Even though not completely desirable, particularly in the second example, a feasible solution in both of these examples was an adjustment to the method or the measurements based on the systematic uncertainty determined by a comparison of items to calorimetered values.

## Standards

Historically, standards for the NDA methods were obtained by calorimetry and gamma isotopics. Even though the measurements were made on oxide and metal, the standards were matrix matched in both cases. However, with waste measurement several new problems were encountered. First, the waste container could not be measured by the calorimeter, since it was too large to fit into the instrument. In addition, since most waste contains smaller quantities of SNM, the calorimeter would not have been capable of providing a good standard value measurement. Other difficulties involve the moderating material found in waste, the lack of uniform density and the non-uniform distribution of SNM in the container.

In an attempt to solve the matrix problem for the PNCC, special standards were fabricated using the normal standards along with a wax medium. The wax simulated the hydrocarbon moderation of the paper and plastic found in waste. Moderation was not a problem for the SGS, but it was necessary to space the normal standards from the center of the waste container to simulate the non-uniform density and distribution of SNM found in waste. See Figure 3 for more detail on the fabricated standards. Using the fabricated standards resulted in more variation in the data. This uncertainty was included when re-estimating the total uncertainty of the PNCC and SGS.[3] However, calibrations using the normally configured standards were used for the actual prediction.

## Calibration

For calibration, the challenges of measuring waste and stabilization material, in addition to the lack of adequate standards mentioned above, include frequent calibrations, non-uniform material within a campaign and the inability to change the calibration of the NDA instrument itself. Most of the waste and stabilization campaigns at SRS are of a relatively short duration, sometimes lasting only months. This is in sharp contrast to previously long periods of SNM production, which provided stability with regard to impurity levels, geometric configuration, metal or oxides. Each waste campaign usually requires the re-calibration of one or more NDA instruments. As a result, calibrations for these short campaigns are at a challenging pace.

The material used to calibrate any instrument should be of the same type, geometry and matrix as the material for which the calibration is used. This is a basic assumption of any calibration. When measuring waste material, one is not always sure that this assumption is being satisfied. The lack of matrix matched samples for the SGS and PNCC were already discussed above. An additional example for the NMC is now presented.

As previously discussed, the NMC is used to quantify SNM, which was re-packaged Rocky Flats metal in one of the campaigns. Due to time constraints, we were only able to compare six representative re-packaged items with the calorimeter as a calibration check. As stated above, analysis of the data from these items indicated a significant systematic error estimate of 2.9%, which was most likely due to geometric differences between SRS production material and the Rocky Flats re-packaged material. Since the instrument was not configured to be easily re-calibrated for geometric effects, and since there were extreme time constraints, it was impossible to "calibrate out" the systematic uncertainty. This is shown in Figure 4 for most of the campaign.

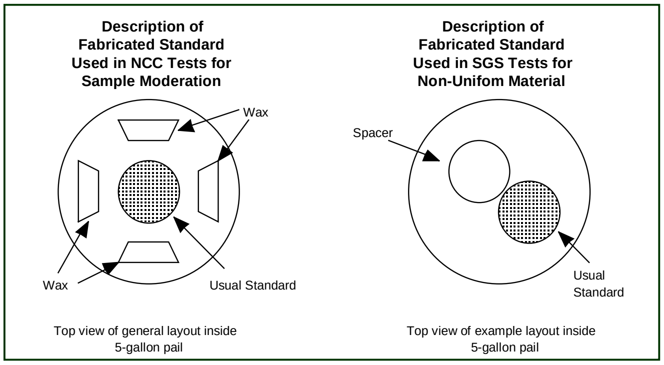

**Figure 3 –Fabricated Standard Layouts**

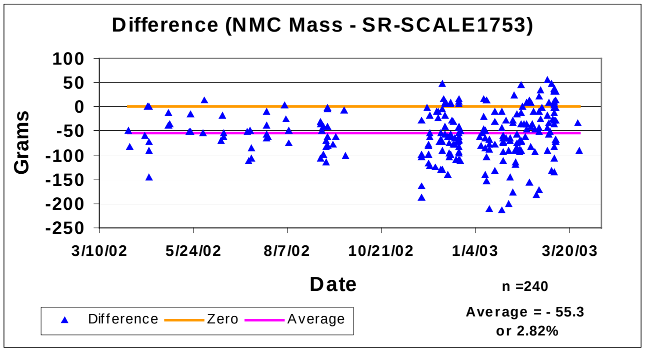

**Figure 4- NMC for Rocky Flats Campaign**

## Time and Cost Constraints

We have already discussed briefly about the need for frequent calibrations due to short waste campaigns, but additional effects of time and cost constraints will now be given.

From a purely statistical viewpoint, one effect of time constraints is small sample sizes for calibration, and determination of uncertainties. We have previously discussed several examples in which time was available to calorimeter and perform NDA comparative measurement on only 6 to 9 items. Estimates of uncertainty, particularly random, are not reliable with such small sample sizes. For example, with the Rocky Flats metal campaign already discussed, the estimate of systematic uncertainty was not adequately estimated until additional items were calorimetered and measured on the NMC after the campaign had already begun.

Sometimes, time and cost constraints lead to success stories. An example of this is the confirmatory measurement counter (CMC), which was qualified for reducing unmeasured inventory of waste. The instrument was previously used for confirming shipment quantities of SNM. A picture of the CMC is shown in Figure 5.

Time and cost constraints prevented the utilization of existing instrumentation for measuring the waste inventory. The SGS and PNCC were normally used for this purpose, but CMC provided much faster results. Qualification of the instrument involved re-calibration and calculation of detection limits, which will be more fully addressed below. After qualification, significant progress is being made in reducing the unmeasured waste inventory. Even though the uncertainty of the CMC was greater for waste measurement than the SGS and PNCC, it was adequate for the intended purpose.

## Detection Limits

Previously, when SRS was producing large quantities of SNM, the gram quantities in items measured by NDA methods was well above the detection limits of the instruments. However, the missions of material stabilization, and particularly waste recovery, often involve gram quantities at or below the detection limit.

**Figure 5 – Picture of CMC**

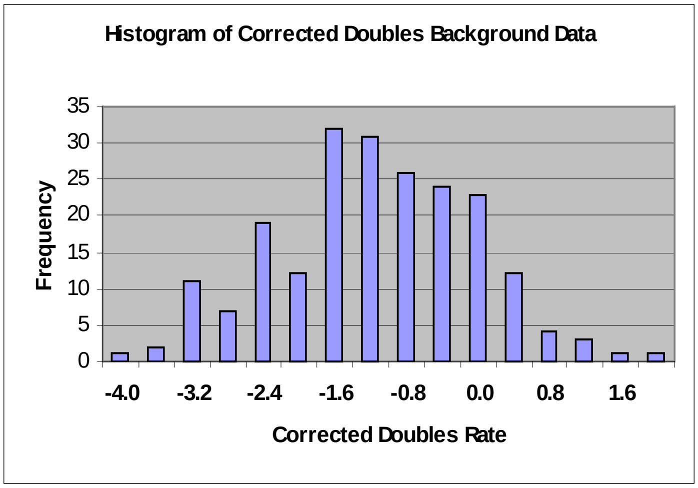

**Figure 6 – Doubles Background Histogram**

Basically, a detection limit is the point below which an instrument cannot give reliable measurements. Because of the detection limit definition, the uncertainty at the detection limit is near +/- 100%. As a result, measurements that are below the detection limit are assigned the detection limit as a conservative estimate. However, measurements at or slightly above the detection limit could be considerably larger due to large uncertainty. In fact for the CMC example discussed below, this was the case.

To solve this problem, the limit of quantification is proposed. This limit is discussed in Currie's paper, which is generally accepted technology for limit determination for counting methods.[3] This limit is around 3 to 4 times further from zero than the detection limit. At this point, the uncertainty of most NDA methods has improved to around 20 to 30% with a high degree of confidence, usually 95%. Currie recommends that measurement between the detection limit and limit of quantification is reported only as "detected". But from a practical standpoint, a value of the measurement is usually desired. Conservatively, the measurement could be given a value equal to the quantification limit. It should also be noted that detection and quantification limits for analytical methods by Taylor is generally in agreement with these principles.[4]

To illustrate the calculation of detection and quantification limits, we will discuss an unusual example from the CMC. This example will also illustrate the need to understand the type of data distribution involved so appropriate statistics can be applied.

Values from a CMC are corrected doubles rates. Therefore, detection limit was determined in doubles rate units before conversion to grams. Since the rate was an average rate based on ten, 60-second counts that had been corrected by background subtraction, the data no longer has the usual distribution for counting, which is a Poisson distribution. This can be observed from the histogram in Figure 6. If fact, the data appears to have an approximate normal distribution. Since Currie's method assumes a Poisson distribution, another method for normal distributions from Taylor was used to determine the limits. This method is typically used for analytical chemistry methods, assumes a normal distribution and uses the ordinary standard deviation, s, of the distribution. Taylor's method is simply 3s for the detection limit and 10s for the quantification limit. For this example s was calculated as 1.1 doubles/second (d/s), which yields a detection limit of 3.3 d/s and a quantification limit of 11.0 d/s. [5]

## Summary

Measurement of stabilization materials and waste at SRS is not business as usual. In fact, these measurements are made with increased difficulty and increased uncertainty when compared to previous measurement of SNM production material. In spite of these challenges and increased need for statistical support, many ways of overcoming these challenges has been shown by the discussion and examples above. In some cases, using production NDA instrumental has resulted in a more timely and cost savings approach than doing otherwise. Even though uncertainty has increased in some cases, the methods are still adequate to meet accountability and other requirements.

## References

1. N. Ensslin, Harker, Krick, Langner, Pickrell and Stewart, Application Guide to Neutron Multiplicity Counting, Los Alamos National Laboratory, LA13422-M Manual, 79, November 1999.

2. Samer D. Kahook & Linda B. Baker, α-Based, Self Correction to the Neutron Multiplicity Coefficients for Assay of SNM with Known Impurities, Presented to NDA users group meeting, Knoxville, TN, November 2001.

3. TSD-CLS-2001-00013, Rev.1, An Improved Set of Measurement and Measurement Uncertainty Correction Factors for the FB-Line SGS, March 2001.

4. L. A. Currie, Limits for Qualitative Detection and Quantitative Determination, Analytical Chemistry, Volume 40, No.3, 586-593, 1968.

5. FSS-CLS-2002-00084, FB-Line Confirmatory Measurement Counter Material Control & Accountability Qualification for Transuranic (TRU) Waste in 5-Gallon Red Pails, September 2002.

6. John L. Jaech, Statistical Methods in Nuclear Materials Control, Technical Information Center, Atomic Energy Commission, 1973.

7. Walpole and Myers, Probability and Statistics for Engineers And Scientists, Macmillan Publishing Company, 1972.

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