Radioactive Waste Measurements: Contributors to Total Measurement Uncertainty

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Abstract

Radioactive wastes that are bound for the waste Isolation Pilot Plant (WIPP0 or other disposal sites are subject to characterization before they can be shipped. Characterization includes site Acceptable Knowledge (AK), Visual Examination (VE), X-ray Nondestructive Examination (NDE), and radiometric Nondestructive Assay (NDA). These characterization methods are developed to meet applicable government regulations, and are usually used together to describe a waste's contents.

NDA techniques include neutron or gamma assays (or both) depending on the nature of the waste. NDA measurement results are generally reported on a radioassay data sheet, formatted to meet the requirements of each site's TRU waste program manager. Results include a list of nuclides, mass, activity, and associated uncertainties, and the minimum detectable activity for each nuclide. The individual nuclide results are used to compute the TRU alpha activity, ^{239}Pu equivalent activity, ^{239}Pu fissile gram equivalent, decay heat, and total Pu mass for each drum. The individual uncertainties are propagated to derive uncertainties for each of these quantities.

Each individual value is reported with a Total Measurement Uncertainty (TMU), into which all the measurement uncertainties are propagated in standard fashion. Typical measurement uncertainties include matrix-source inhomogeneity, source distribution, and lumpiness in addition to counting statistics. The individual uncertainties are evaluated to determine the relative importance of each, so that major contributors to the overall measurement uncertainty can be minimized if possible.

This paper will discuss the contributors to TMU for typical gamma and neutron assays, and discuss the relative importance of each one.

Introduction

Everyone who collects and analyzes radiometric data is familiar with counting statistics and the uncertainties that are derived thereby. However, in complex measurements the statistical uncertainty is not the only source of error, and in many cases it is not even the dominant one. To get a realistic idea of the uncertainty inherent in a complex measurement, one must consider all the variables inherent in the measurement, identify and quantify their related uncertainties, and somehow propagate them to predict an overall uncertainty. This overall uncertainty is referred to as TMU in the regulations that govern radioactive waste disposal [Ref. 1].

Once identified and quantified, in many cases the individual uncertainties may be propagated using the error propagation formula

$$\sigma_u^2 = \left(\frac{\partial u}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial u}{\partial y}\right)^2 \sigma_y^2 + \left(\frac{\partial u}{\partial z}\right)^2 \sigma_z^2 + \ldots$$

where $u = u(x,y,z,\ldots)$,

$x,y,z,\ldots$ are the independent variables from which the overall result is derived,

and

$\sigma_x, \sigma_y, \sigma_z, \ldots$ are the standard deviations of those variables.

The error propagation formula can be simplified, in the case of multiplicative factors, so that the relative uncertainties are added in quadrature to derive the relative uncertainty in the overall result

$$\frac{\sigma_u}{u} = \sqrt{\left(\frac{\sigma_x}{x}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2 + \left(\frac{\sigma_z}{z}\right)^2 + \ldots}$$

The variables of a complex measurement may have either controllable or uncontrollable uncertainties. Examples of controllable uncertainties include uncertainties in calibration and counting statistics. Often these uncertainties can be minimized by improving the measurement process. Examples of uncontrollable uncertainties include matrix inhomogeneity and source distribution within the waste. There is usually nothing the analyst can do to reduce this type of uncertainty. Each measurement variable, along with its uncertainty, must be considered and included in the TMU calculation. Quantifying the uncertainties is, in some cases, as much art as it is science. This paper presents two examples, passive neutron counting and gamma spectrometry, to illustrate typical contributors to TMU.

TMU is applied routinely to radioactive waste measurements. For example, wastes destined for disposal at the Waste Isolation Pilot Plant (WIPP) are governed by the US Department of Energy Order DOE /WIPP-02-3122, which specifies that each waste drum's Radioassay Data Sheet must include a TMU expressed in activity (in Curies) and/or mass (in grams). TMU is routinely assessed to improve the measurement process. By reducing the magnitude of the biggest contributors to TMU, the overall uncertainty is reduced and the measurement as a whole is improved.

Typical Measurement Techniques

Drummed radioactive wastes are normally assayed using neutron or gamma techniques, or a combination of the two (only the case of 200 ℓ drums will be considered here). Results for these techniques are reported, along with their TMUs, on Radioassay Data Sheets as required. Contributors to the TMU, and their relative importance, are greatly dependent on the combination of measurement techniques applied to a waste [Ref. 2].

Neutron measurements may be passive or active. For passive measurements, the coincident neutrons emitted from spontaneous fissions in the waste are counted directly. Active techniques usually use an external neutron source to induce fissions in the sample, and the coincident fission neutrons are then counted. The neutron coincidence count is not usually used by itself, but combined with a measured or assumed set of isotopic fractions to determine the quantities of each fissile nuclide.

Gamma measurements may be used to determine the isotopic fractions of elements such as uranium or plutonium, or may be used to quantify individual radionuclides. Gamma spectrometers use one or more detectors, and may be configured to measure the sample as a whole or in segments. Gamma instruments sometimes have a mechanism for self-attenuation correction, which generally involves specialized software.

Neutron measurement uncertainties

Major variables associated with neutron assays include calibration, isotopic fractions, neutron source strength (active mode only), counting statistics, matrix effects, neutron absorption (active mode only) and moderation, radionuclide distribution, multiplication, and cosmic ray interactions. Typical uncertainties associated with these variables are displayed in Table 1.

Table 1. Neutron uncertainty contributions in typical waste matrices.

Uncertainty Source	Debris Waste 1 σ	Homogeneous Solids 1 σ
Counting Statistics	2.5%	5%
AAS Correction	4%	5%
Source Distribution	2.9%	17%
Multiplication Effects	0%	0%
Background effects	0%	0%
Isotopics Uncertainty	12%	12%
Calibration Uncertainty	2%	2%
Nominal TMU (1-σ)	13.4%	22.1%

Neutron counters are usually calibrated using either ^{235}U (active mode only), plutonium or ^{252}Cf sources. If possible, calibration sources are counted long enough so that the uncertainty in the calibration curve is negligible compared to the other uncertainties. In the example shown in Table 1 (weapons-grade Pu oxide in surrogate waste matrix), the calibration uncertainty is assigned a nominal 2% relative standard deviation.

Passive neutron coincidence counters are usually calibrated in terms of $^{240}Pu_{effective}$, since the observed coincidence response is mainly due to spontaneously fissioning ^{238}Pu, ^{240}Pu, and ^{242}Pu. In order to generate quantitative values for all the plutonium isotopes, and the

total Pu mass, it is necessary to know the isotopic fractions accurately. It is important to note that error in the isotopic fractions is more important for neutron assays than for gamma ray assays. This is because a neutron assay result is based on the $^{240}Pu_{effective}$ content (mostly ^{240}Pu) of a waste and the isotopic fractions, so the ^{239}Pu value (representing most of the Pu mass) is derived using a large scaling factor. An error of a few percent in the ^{240}Pu fraction can therefore cause an error of 10 grams, or more in the Pu mass. In waste measurements, the isotopic fractions are either determined by gamma spectrometry, or assumed as defaults from site AK. Uncertainty in the isotopic fractions is propagated for each isotope and accumulated in the final result.

Most of the variables that introduce variables to a neutron measurement are the same, whether the measurement is active or passive. Two notable exceptions are variability in the external neutron source's emission rate, which depends on the type of neutron source, and neutron absorption by the matrix. In general, spontaneous fission and (α,n) neutron sources have less variability in the neutron emission rate than do accelerator-based neutron generators. However, instruments that use neutron generators are usually equipped with flux monitors to correct for such variability. Active neutron counters usually include 0% to 4% in their TMU to account for neutron source emission variability. Neutron absorption by the matrix comes about for waste in hydrogenous matrices because the interrogating neutrons are strongly moderated by the neutron counter's polyethylene walls and drum matrix contents prior to causing fissions. Corrections to the data are typically applied based on the interrogating neutron flux. The corrections are small for non hydrogenous matrices such as metals but become large for matrices with high water content such as sludge. TMU estimates for this effect are specific to the manufacturers of each type of active neutron system.

Uncertainty due to counting statistics is controlled by the counting time. To maintain reasonable throughput, it is not practical to extend the counting time beyond a preset limit. For transuranic waste measurements, it is only necessary that the count time be long enough to reliably determine whether the waste contains at least 100 nCi/g of Pu. For a 1-g Pu (weapons grade) sample and 1200 s count time, the counting statistics uncertainty is generally about 2.5% relative standard deviation in combustibles or steel, and about 5% in sludges.

Neutron interactions vary widely in different waste matrices, especially if the matrices are moderating or absorbing. Add-a-source corrections are effective in reducing bias caused by the waste matrix, except for highly moderating wastes such as sludge or polyethylene which may suffer from bias in addition to large uncertainty. If the matrix and source are reasonably homogeneous, the add-a-source uncertainty for metal wastes like steel may be as low as 1%. For combustible wastes, it may be 2.5 %, and for sludges, it is estimated as 5%.

If the waste is heterogeneous, neutron measurements will have additional uncertainty. The extreme case is that of a neutron-emitting point source in the waste. Test cases have shown that the position dependence and resulting uncertainty increase dramatically as the amount of moderator increases. This uncertainty component is estimated from two

empirical relationships R_{min} (minimum response/average response) and R_{max} (maximum response/average response) and the Add-a-Source correction factor.

For a typical High Efficiency Neutron Counter, and assuming that R_{max} and R_{min} represent ±3σ extremes for the response of a single point source position, computer models indicate that the source distribution TMU component in waste is:

$$R_{\min} = 0.97 \cdot e^{0.69 \cdot (1 - f_{AaS})},$$

$$R_{\max} = 1.03 \cdot e^{0.22 \cdot (f_{AaS} - 1)},$$

$$\text{and } \sigma_{position} = (R_{\max} - R_{\min}) / 6 / \sqrt{3} \quad \text{[Ref. 3]}$$

Typical source distribution uncertainty is about 2.9% RSD for debris, and up to 17% in dense, moderating matrices like sludge. Although a point source in hydrogenous sludge would be the worst case, such waste is almost always homogeneous with respect to source distribution so the uncertainty is usually overstated. The difference between R_{max} and R_{min} is divided by √3 to account for the probability that all drums in a waste stream will consist of multiple rather than single point sources.

Multiplication effects can be significant in wastes containing plutonium along with appreciable amounts of fluorides in intimate association, or Pu oxides. These increase the probability of (α,n) reactions and increase multiplication in the waste, which in turn creates bias and a one-sided uncertainty. The ratio of (α,n) neutrons to spontaneous fission neutrons is an indicator of multiplication effect; if this ratio is less than 5, the multiplication uncertainty is given by

$$\sigma_{mlt} = 10^{-4} \cdot (m_{Pu})^{1.5}$$

where m_{Pu} is the true plutonium mass in grams.

For low-mass Pu wastes (<1 g Pu), the uncertainty associated with multiplication effects is <0.1%. However for high mass drums the multiplication uncertainty can be the most significant factor. A drum in the range of 200 g using the equation shown above can have an uncertainty of approximately 28%.

Cosmic rays interact with metals in the waste, producing coincident spallation neutrons. If uncorrected, these would lead to a biased result, especially at higher elevations. This effect increases with the amount of metal in the drum, and with the atomic number of the metal. If High-Z corrections are made, the uncertainty in the correction factor is also dependent on the nature and amount of metal present in the waste. At lower elevations, this effect can be neglected. At higher elevations the cosmic ray interactions can significantly affect the precision of the instrument.

Gamma measurement uncertainties

Major variables associated with gamma ray assays include calibration, counting statistics, background fluctuations, matrix non-homogeneity, radionuclide distribution, and isotopic fractions. The uncertainties associated with these variables are displayed in Table 2.

Table 2. 1-σ Gamma uncertainty contributions for assay of 10 g Pu-239 as oxide in typical waste matrices.

Uncertainty Source	Debris Waste (38 kg)	Homogeneous solids (227 kg)
Counting Statistics	0.8%	1.1 %
Matrix Non Homogeneity	5 %	5 %
Source Distribution	9 %	25 %
Calibration Uncertainty	2.5 %	2.5 %
Nominal TMU (1-σ)	10.6 %	25.6 %

Gamma spectrometers used to assay transuranic waste are generally calibrated using either plutonium standards or mixed-nuclide sources that emit gamma rays spanning the energy range of gamma rays expected in the waste. There are several schemes for calibrating gamma spectrometers, but most waste measurements are performed using an external source for transmission correction, or an energy/density/efficiency calibration multi-curve, or both. For the energy/density/efficiency calibration, the calibration sources are mounted in drums containing matrices of various densities to span the range of densities expected in the waste. Calibration uncertainty is controlled by counting the calibration sources long enough so that the uncertainty in the calibration curve is negligible compared to the other uncertainties. In the example shown in Table 2, gamma calibration uncertainty is assigned a nominal 2.5% independent of gamma energy or waste matrix density. This factor also takes into account the uncertainty in the fabrication of the source(s) as indicated in the calibration source certificate(s).

For gamma spectrometry, the uncertainty due to counting statistics is determined by the counting time, the density of the waste, and the amount and identity of the radionuclides present. Dead time and spectral interference corrections and their related uncertainties are propagated with the counting statistics uncertainty. The counting statistics uncertainty can be reduced by extending the assay time, but in practice a maximum assay time is preset to balance assay quality against throughput needs. For WIPP-destined drums, count times must be at least long enough to reliably detect 100 nCi of plutonium per gram of waste.

Fluctuations in background radiation also add to the uncertainty in a gamma measurement, but in working nuclear facilities, these fluctuations are caused by movement of radioactive materials in the vicinity of the spectrometer. These movements are not random events, and it is not possible for NDA personnel to predict or treat them

mathematically. In practice, a gamma ray background measurement is made daily and subtracted from all gamma measurements made that day.

Matrix non-homogeneity is another potential source of uncertainty. Such uncertainties can be minimized by using drum rotation, segmented scanning and transmission correction. If those techniques are not available, it may be necessary to assay the drum in a single count, assuming the drum's contents are uniform, and adjust the uncertainty accordingly. Sludge matrices are generally homogeneous because of the way they are prepared – radionuclides are precipitated from batches of waste liquids, and the solids are filtered and placed into drums. No such statement can be made about debris wastes, which may be accumulations of just about anything. If the source distribution is fairly uniform throughout the matrix, the matrix uncertainty for gamma rays above 100 keV is typically <5%.

Matrix effects are more important for lower-energy gamma rays or for wastes in which the source distribution is non-uniform. Low-energy gamma rays can be corrected for attenuation if there are other gamma rays with higher energies emitted by the same radionuclide. Instrument response to non-uniform source distributions can be mapped with a computer simulation to predict where the minimum and maximum values will occur. A further simplification can be made if more than one point source is considered; if three randomly-distributed sources are assumed, the source distribution uncertainty approaches a normal distribution. For a single detector positioned alongside the waste drum, the minimum and maximum response functions can be expressed as

$$R_{\min} = a_{\min,0} + a_{\min,1} \cdot \rho + a_{\min,2} \cdot \rho^2,$$
$$R_{\max} = a_{\max,0} + a_{\max,1} \cdot \rho + a_{\max,2} \cdot \rho^2,$$
$$\text{and } \sigma_{position} = (R_{\max} - R_{\min}) / 6 / \sqrt{3}$$

where $a_{min,0} = 0.8092$ $a_{max,0} = 1.2189$
$a_{min,1} = -0.8645$ $a_{max,1} = 1.5131$
$a_{min,2} = 0.2439$ $a_{max,2} = -0.2380$

Plutonium uncertainties are usually computed from the measured ^{239}Pu mass and the Pu isotopic fractions. The isotopic fractions may be determined form the gamma spectrum or assumed from site AK. In either case, the uncertainty for each nuclide is propagated from the ^{239}Pu uncertainty and the individual isotopic uncertainties. In some cases, ^{237}Np, ^{241}Am, and ^{243}Am are also determined as weight fractions relative to ^{239}Pu. Coefficients may be geometry-dependent. This example is for an uncollimated detector in close proximity to the waste drum. The difference between R_{max} and R_{min} is divided by $\sqrt{3}$ to account for the probability that all drums in a waste stream will consist of multiple rather than single point sources.

Other uncertainties

There are several parameters with uncertainties that are common to all types of waste measurements.

For example, every container of waste is weighed to determine its net mass. Gross and tare measurements are both needed to determine the net mass, but in practice the combined uncertainties of both are insignificant when compared to matrix, source distribution, and counting uncertainties. Variations in drum positioning are generally negligible.

Drums vary in thickness and coating, and a liner may or may not be present. Unless the drums had been weighed before filling, these variations can give rise to tare weight errors. Some drum liners may require a separate calibration.

Acceptable knowledge is gleaned from a number of sources, including written records, process knowledge, and recollections of technical staff. The quality of AK varies accordingly. Default isotopic fractions assumed from AK may not be reliable estimates in all cases, but may be the only available information about a waste.

Some radionuclides are not directly quantifiable by either neutron or gamma techniques. The presence of ^{90}Sr in a waste is inferred when ^{137}Cs is detected. ^{234}U is inferred from ^{235}U in cases where the waste contains enriched uranium, or from ^{238}Pu in heat source waste. ^{242}Pu is always quantified relative to ^{239}Pu. In these cases, scaling factors are developed from AK, and the derived nuclide's uncertainty is scaled from the uncertainty of the observed nuclide.

In cases where both neutron and gamma results are available, the analyst must select one mode over the other depending on which mode is deemed more reliable. The uncertainty inherent in that selection is not included in the overall uncertainty.

Summary

As shown in Tables 1 and 2, the relative importance of measurement uncertainties is quite different for neutron assays and gamma ray assays. Reliable isotopic fractions are essential for a good neutron assay of Pu, but matrix and source homogeneity are more important for good gamma ray assays that directly measure ^{239}Pu.

It is often possible to improve isotopic measurements, or to develop more credible AK, to significantly improve neutron assays. Sludges are poor candidates for neutron assay because the matrix is highly moderating and the result may be biased unless assumptions of matrix homogeneity and activity uniformity apply.

Because matrix and source distributions are more important in gamma ray assays, segmented scans and transmission correction greatly improve the results over static measurements [Ref. 4, 5]. Multichannel scaling techniques may also be used to identify matrix inhomogeneities [Ref. 6].

Source distribution in sludges is probably not as important as the values in Tables 1 and 2 imply, because sludge matrices are usually homogeneous with the source material evenly distributed throughout. The distribution is important for performance testing of the instrument. If sludge test drums are loaded incorrectly the affects on the measurements can be significant.

References:

1. D.R. Davidson, M.J. Koskelo, B.M. Gillespie, B.D. McElroy, and T. Nishida, "Total Measurement Uncertainty for NDA for SNM in Process Materials and Waste", presented at the INMM-Japan Chapter Meeting, Tokyo (November 1997)

2. J.M. Veilleux, D. Cramer, "Comparison of Gamma and Passive Neutron NonDestructive Assay Total Measurement Uncertainty Using the High Efficiency Neutron Counter", presented at the 46th Annual Meeting of the INMM, Phoenix (July 2005)

3. B.M. Gillespie, "Total Measurement Uncertainty for the CCP High Efficiency Neutron Counter (HENC)", Canberra Industries report no. CCP-INL-HENC-0002 (April 2005)

4. R. Venkataraman, S. Croft, M. Villani, R.D. McElroy, and R.J. Estep. "Total Measurement Uncertainty Estimation for Tomographic Gamma Scanner", presented at the 46th Annual Meeting of the INMM, Phoenix (July 2005)

5. B.M. Gillespie, M. Zebarth, "Comparison Of A Variety Of Gamma Attenuation Correction Techniques for Different Waste Matrices", presented at ESARDA, Salamanca (April 1993)

6. B.M. Gillespie, "Detection and Correction of Inhomogeneities in Drum Waste Assay Systems", presented at Nondestructive Assay and Nondestructive Examination Waste Characterization Conference, Pocatello (February 1994)