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Total Measurement Uncertainty in Holdup Measurements at the Plutonium Finishing Plant

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Project Hanford Management Contractor for the
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ABSTRACT

An approach to determine the total measurement uncertainty (TMU) associated with Generalized Geometry Holdup (GGH) [1, 2, 3] measurements was developed and implemented in 2004 and 2005 [4]. This paper describes a condensed version of the TMU calculational model, including recent developments.

Recent modifications to the TMU calculation model include a change in the attenuation uncertainty, clarifying the definition of the forward background uncertainty, reducing conservatism in the random uncertainty by selecting either a propagation of counting statistics or the standard deviation of the mean, and considering uncertainty in the width and height as a part of the self attenuation uncertainty. In addition, a detection limit is calculated for point sources using equations derived from summary equations contained in Chapter 20 of MARLAP [5].

The Defense Nuclear Facilities Safety Board (DNFSB) Recommendation 2007-1 to the Secretary of Energy identified a lack of requirements and a lack of standardization for performing measurements across the U.S. Department of Energy (DOE) complex. The DNFSB also recommended that guidance be developed for a consistent application of uncertainty values. As such, the recent modifications to the TMU calculational model described in this paper have not yet been implemented. The Plutonium Finishing Plant (PFP) is continuing to perform uncertainty calculations as per Reference 4. Publication at this time is so that these concepts can be considered in developing a consensus methodology across the complex.

ASSAY CONVENTIONS

The primary detection systems are sodium iodide (NaI) detectors with a peak region-of-interest (ROI) approximately 405 to 435 keV. Spectral Compton background is subtracted from an unequal width ROI from approximately 440 to 450 keV.

Most gloveboxes are assayed by either placing the detector inside gloveports to assess each surface or by placing the detector back from the front of the glovebox and modeling the entire glovebox as a single plane of activity coinciding with the centerline of the glovebox. Linear systems such as vacuum piping, ventilation ducts, and some gloveboxes are assayed from the far field and calculated as line sources. Special items such as valves and elbows are assessed separately as point sources. Waste packages and individual items are also assayed as point sources. Self-attenuation and finite width corrections are made.

TOTAL MEASUREMENT UNCERTAINTY

Systematic uncertainties are evaluated by segregating individual measurements into distinct populations with similar characteristics. Each distinct population group is assigned an uncertainty

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that represents each member in the group. Generally, the population group size is an entire surface, line length, or item.

The systematic uncertainty of the measurement is calculated as follows:

$$\sigma_{systematic} = \sqrt{\begin{array}{l} \left(m_{tot}\times\sigma_f\right)^2 + \left(m_{tot}\times\sigma_K\right)^2 + \sum_p\left(m_p\times\sigma_{CF(AT)_p}\right)^2 + \left(m_{tot}\times\sigma_{CF(AT)_{general}}\right)^2 + \\ \sum_p\left(m_p\times\sigma_{ICF_p}\right)^2 + \sum_p\left(m_p\times\sigma_{dist_{Line,p}}\right)^2 + \sum_p\left(m_p\times\sigma_{glove_p}\right)^2 + \sum_p\left(m_p\times\sigma_{equip}\right)^2 + \\ \sum_p\left(m_p\times\sigma_{ledges_p}\right)^2 + \sum_p\left(m_p\times\sigma_{mat\,distrib_p}\right)^2 + \sum_p\left(m_p\times\sigma_{f\,bkg_p}\right)^2 + \sum_p\left(m_p\times\sigma_{Sorenson_p}\right)^2 \end{array}}$$

where:

m_{tot} and m_p	is the calculated total and population mass, respectively.
p	represents the population group for the given uncertainty.
σ_f	is the uncertainty for the mass fraction of ^{239}Pu.
σ_K	is the calibration uncertainty.
$\sigma_{CF(AT)}$	is the attenuation uncertainty for each shield.
$\sigma_{CF(AT)general}$	is the general attenuation uncertainty that is judged to be 10 percent, due to additional sources of attenuation uncertainty that are difficult to quantify (e.g., poorly known material densities, use of empirically determined coefficients).
σ_{ICF}	is the uncertainty associated with the item correction factor (ICF)[†]. It is due to both the uncertainty of the position of the deposit within the line width or point source and the uncertainty in the line width or point source size.
σ_{dist}	is the distance uncertainty. It is a systematic uncertainty in relation to line sources, because the detector is held a consistent distance from the surface of an item for multiple measurements.
σ_{glove}	is the contaminated glove uncertainty. Assays are made through glove ports, bagout ports, and windows, which are assumed to be clean, but may be contaminated.
σ_{equip}	is the intervening equipment uncertainty. Assays made through glove ports assume the activity is located on the opposite surface. Intervening process equipment may be included in assay measurements from both sides resulting in assaying the item twice.
σ_{ledges}	is the ledges uncertainty. When a glovebox is assayed through the glovebox wall, steel framework between panels and around edges is difficult to assess and could represent significant shielding on the near surface, but not on the far surface.

[†] The ICF is the historical Plutonium Finishing Plant name for the Finite Width Correction Factor.

$\sigma_{mat\ distrib}$	is the material distribution uncertainty. Measurements of area and line sources have sensitivity to nonuniform material distributions. There may be a diminished response at the edge of a surface or line, and it is not generally practical to space adjacent measurement shots uniformly.
$\sigma_{f\ bkg}$	is the forward background uncertainty due to non-Poisson background interference from plutonium deposits forward of the detector and which are generally difficult to accurately account for in a background measurement.
$\sigma_{Sorenson}$	is the Sorenson[‡] uncertainty. Measurement assume the detector is aiming directly at the object. This is not always the case and results in a slight underestimation of the activity due to a diminished response at the edge of the field of view.

The basis for the material distribution uncertainty is depicted in Figure 2. Figure 2 shows the effects of uneven detector spacing in relation to counting a single point source located on the X-axis. The Y-axis represents the individual detector responses to a point source located accordingly on the X-axis. The thick black line represents the average response of the overall measurement system (measurements made at each detector location). Detector spacing is in terms of the effective length, *Eff L*, which is a geometrical constant in the GGH model and is approximately equal to the full width at half maximum of the radial response. Figure 2 is taken to represent the extreme case of positioning effects due to assaying a localized deposit when the detector placement deviates from normal spacing. In reality, material distributions tend to be spread out. The minimum and maximum values of Figure 2 are taken to be the lower and upper ends of a normal probability distribution at the 99 % confidence level, or six standard deviations. These curves were used to support the default guidance for $\sigma_{mat\ distrib}$ listed in Table 1. The two dimensional uncertainty is estimated by root sum square of each dimension.

The basis and values for $\sigma_{CF(AT)}$, σ_{ICF} , σ_{dist}, $\sigma_{Sorenson}$, σ_{ledges}, σ_{equip} ,σ_{glove}, $\sigma_{f\ bkg}$ are described:

$\sigma_{CF(AT)}$	Attenuation correction factors are calculated for both a most representative shielding and for a largest plausible shielding configuration. The uncertainty is estimated from the range between the largest and most representative correction factors. The difference represents two standard deviations (one sided distribution).
σ_{ICF} , σ_{dist}	Estimated from the range of plausible parameters. The difference between the largest and smallest plausible parameters represents four standard deviations (two sided distribution).
$\sigma_{Sorenson}$	Underestimation of 11% (effect of being 15° off center) represents three standard deviations and is assigned a value of 4% (one sided distribution).
σ_{ledges}	Underestimation of 50% represents two standard deviations and is assigned 25% (one sided distribution).
σ_{equip}	Overestimation of 50% represents three standard deviations and is assigned 17% (one sided distribution).

[‡] The term was named after Donald L. Sorenson, a senior NDA technician at the Plutonium Finishing Plant.

σ_{glove} Overestimation of 50% represents three standard deviations and is assigned a value between 0 and 17%. (one sided distribution).

$\sigma_{f\,bkg}$ Estimated from the mass difference observed between subtracting ambient background versus zero background, which represents two standard deviations (one sided distribution).

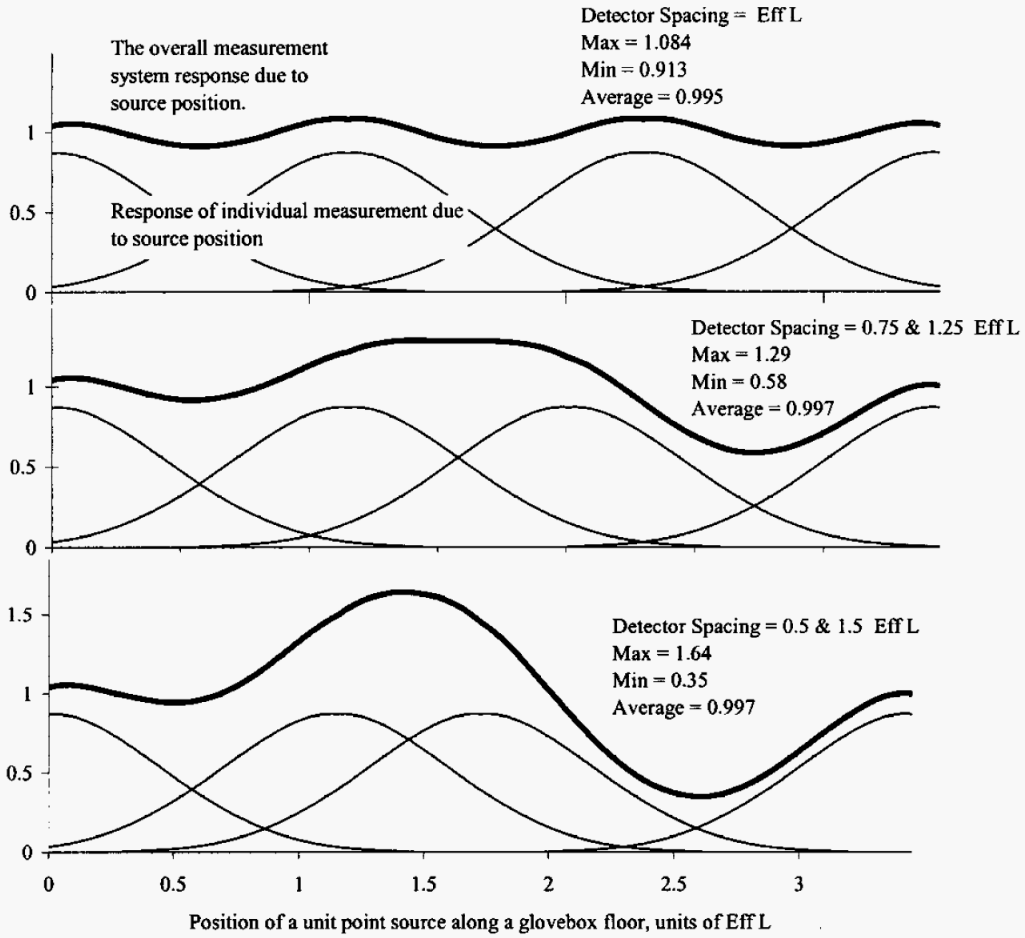

Figure 2. The Individual Detector and Overall System Response to a Single Point Source Located on the X-axis.

Table 1. Default Guidelines for $\sigma_{mat\ distrib.}$

Spacing of measurements	One dimensional uncertainty	Two dimensional uncertainty
Near Eff L	3%	4%
Less than +/- 25% of Eff L	12%	16%
Greater than +/- 25% of Eff L	22%	31%

The random uncertainty is represented by either the standard deviation of the mean or a propagation of the counting statistics adjusted for self attenuation (Reference 2). The standard deviation of the mean is chosen when the measurement shots of an item represent a sampling of the entire item. A propagation of the counting statistics adjusted for self attenuation is chosen when the measurement shots are located such that the entire surface or line segment, is measured. The random uncertainty is estimated using the following reduced equation:

$$\sigma(random)=\sqrt{\sigma_{rand}^2+\sigma_{area}^2+\sigma_{length}^2+4\sigma_{dist_{Point}}^2}$$

where:

σ_{rand}	is either the uncertainty due to counting statistics adjusted for self attenuation or the standard deviation of the mean.
σ_{area}	is the area uncertainty for area source items.
σ_{length}	is the length uncertainty for line source items.
σ_{dist}	is the distance uncertainty for point source items. It is considered random as the distance bias to the actual deposit varies depending on the rotation of the object.

The overall TMU is the root sum square of the random and systematic components.

SELF ATTENUATION

The self attenuation correction is a nonlinear exponential function. At high degrees of self attenuation, the uncertainty associated with an individual measurement point is nonlinear for both systematic and random uncertainties. Traditional error propagation is not practical. This section describes the total uncertainty associated with a single measurement point. When the single measurement point contains significant self attenuation and is inside of a larger system, a judicially determined method uncertainty should additionally be assigned to the set of measurements.

The corrected linear concentration (line sources) and mass (point sources) is given as follows:

$$L_{SAcorrPu}=\frac{-w}{\mu/\rho(1+f_U)}\times ln\left(1-\mu/\rho\times\frac{L_{Pu}}{w}(1+f_U)\right)$$

$$P_{SAcorrPu}=\frac{-w\times h}{\mu/\rho(1+f_U)}\times ln\left(1-\mu/\rho\times\frac{P_{Pu}}{w\times h}(1+f_U)\right)$$

where:

$L_{SAcorrPu}$	is the linear concentration of plutonium corrected for self attenuation.
$P_{SAcorrPu}$	is the plutonium mass for a given point source corrected for self attenuation.
L_{Pu}	is the linear concentration of plutonium for a given measurement.
P_{Pu}	is the plutonium mass for a given point source measurement in grams.
w	is the width of the "fat" point or "wide" line in inches.

h is the height of the "fat" point in inches.

μ/ρ is the mass attenuation coefficient with the value of 0.0388 in^2/g for PuO_2.

f_U f_U is the weight percent of uranium in the system relative to plutonium.

The variance at each measurement location is derived from a partial derivative. Independent variables are assumed. Uranium must be absent or have a negligible uncertainty in the isotopic concentration in comparison to the overall measurement uncertainty. The source must also be counted at a sufficient distance such that the deposit width and height results in a negligible uncertainty related to the ICF.

$$\sigma^2_{L_{SAcorrPu}} = \left(\frac{\partial L_{SAcorrPu}}{\partial L_{Pu}}\right)^2 \sigma^2_{L_{Pu}} + \left(\frac{\partial L_{SAcorrPu}}{\partial w}\right)^2 \sigma^2_W$$

$$\sigma^2_{P_{SAcorrPu}} = \left(\frac{\partial P_{SAcorrPu}}{\partial P_{Pu}}\right)^2 \sigma^2_{P_{Pu}} + \left(\frac{\partial P_{SAcorrPu}}{\partial w}\right)^2 \sigma^2_W + \left(\frac{\partial P_{SAcorrPu}}{\partial h}\right)^2 \sigma^2_h$$

The solution is an absolute uncertainty, where $\sigma_{L_{Pu}}$, $\sigma_{P_{Pu}}$, σ_w and σ_h are all relative.

$$\sigma_{L_{SAcorrPu}} = \sqrt{\left[\frac{L_{Pu}}{1-\frac{\mu/\rho L_{Pu}}{w}(1+f_U)}\right]^2 \sigma^2_{L_{Pu}} + \left[L_{SAcorrPu}\frac{L_{Pu}}{1-\frac{\mu/\rho L_{Pu}}{w}(1+f_U)}\right]^2 \sigma^2_w}$$

$$\sigma_{P_{SAcorrPu}} = \sqrt{\left[\frac{P_{Pu}}{1-\frac{\mu/\rho P_{Pu}}{w\times h}(1+f_U)}\right]^2 \sigma^2_{P_{Pu}} + \left[P_{SAcorrPu}\frac{P_{Pu}}{1-\frac{\mu/\rho P_{Pu}}{w\times h}(1+f_U)}\right]^2 \left[\sigma^2_w+\sigma^2_h\right]}$$

POINT SOURCE DETECTION LIMIT

The value of the Critical Level, S_c, refers to the number of net counts in a counting period which, when exceeded, is taken to indicate that a sample contribution exists with a specified probability, 1 - α. The MARLAP critical level definition considers both a normal counting background and an external interference background:

$$S_c = z_{1-\alpha} t_s \sqrt{\frac{r_B+r_I}{t_s} + \frac{r_B}{t_B} + \xi^2_B + \sigma^2_{r_I}}$$

where:

t_s is the sample count time.

t_B is the background count time.

r_B is the mean count rate of the blank.

r_I is the mean count rate of interference background.

ξ_B is the non-Poisson variance in the blank.

$\sigma^2_{R_I}$ is the variance estimator for interference background count rate.

PFP holdup measurements include a peak measurement and a Compton scattering background. The Regions-Of-Interest (ROI) for parameters are unequal in width. There may be multiple measurements of the item and also an unequal number of interference background measurements (room background). The MARLAP equation is re-written as:

$$S_c = z_{1-\alpha}\sqrt{2\sigma_B^2 + \sigma_I^2 \frac{t_s}{t_I}\left(1+\frac{t_s}{t_I}\right) + \sigma^2_{f\,bkg}}$$

assuming:

$t_S = t_B$ Peak and Compton background counts are from the same spectrum.

$\sigma^2_B = r_B t_B$ The variance in the background is due to Poisson statistics.

$\sigma^2_I = r_I t_I$ The variance in the interference background (of the above equation) is equal to the total counts in the interference background.

$\sigma^2_{RI} = \sigma^2_I / t_I^2$ The variance in the interference background rate (of the above equation) is equal to the variance in the interference background divided by the square if the interference count time.

$t^2_s \xi^2_B = \sigma^2_{f\,bkg}$ The non-Poisson variance in the blank rate multiplied by the count time squared is taken to be the forward background uncertainty.

The net counts for the measurement is:

$$NET = \left(C_S - R \times B_S\right) - \left(C_I - R \times B_I\right)\frac{T_S}{T_I}$$

where:

R is the channel ratio of the peak ROI to the Compton background ROI.

B_S is the summation of the Compton background for multiple measurements.

C_I is the summation of the interference peak background for multiple measurements.

B_I is the summation of the interference Compton background for multiple measurements.

T_S is the summation of the sample count times, t_S, for multiple measurements.

T_I is the summation of the interference count times, t_i, for multiple measurements.

The uncertainty σ_{fbkg} is equal to ½ the difference between the net counts and the net count calculated without the interference background. This leads to the following equation for the critical level:

$$S_c = 1.645\sqrt{2R^2 B_S + \left(C_I + R^2 B_I\right)\frac{T_s}{T_I}\left(1+\frac{T_s}{T_I}\right) + \left[\frac{1}{2}(C_I - RB_I)\frac{T_s}{T_I}\right]^2}$$

The limit of detection in terms of net counts, S_D, refers to the smallest quantity of an analyte (plutonium) which is expected to yield a net count that exceeds the Critical Level with a specified probability, 1-β. The MARLAP method is used to approximate the limit of detection for $z_{1-\beta} = z_{1-\alpha} = 1.645$:

$$S_D = \frac{2.7 + 2S_C}{1 - 2.7a}$$

where a is the relative variance of the overall sensitivity, or the variance of the systematic components and the distance:

$$a = \hat{\sigma}_f^2 + \hat{\sigma}_K^2 + \hat{\sigma}_{CF(AT)}^2 + \hat{\sigma}_{CF(AT)_{general}}^2 + \hat{\sigma}_{ICF}^2 + \hat{\sigma}_{glove}^2 + \hat{\sigma}_{Sorenson}^2 + 4\hat{\sigma}_{dist}^2$$

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