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# Errors Associated with the Direct Measurement of Radionuclides in Wounds

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## Introduction

Work in radiation areas can occasionally result in accidental wounds containing radioactive materials. When a wound is incurred within a radiological area, the presence of radioactivity in the wound needs to be confirmed to determine if additional remedial action needs to be taken. Commonly used radiation area monitoring equipment is poorly suited for measurement of radioactive material buried within the tissue of the wound. The Lawrence Livermore National Laboratory (LLNL) In Vivo Measurement Facility has constructed a portable wound counter that provides sufficient detection of radioactivity in wounds as shown in Fig. 1. The LLNL wound measurement system is specifically designed to measure low energy photons that are emitted from uranium and transuranium radionuclides. The portable wound counting system uses a 2.5cm diameter by 1mm thick NaI(Tl) detector. The detector is connected to a Canberra NaI InSpector™. The InSpector interfaces with an IBM ThinkPad laptop computer, which operates under Genie 2000 software.

Fig. 1 – Portable Wound Counting System

The wound counting system is maintained and used at the LLNL In Vivo Measurement Facility. The hardware is designed to be portable and is occasionally deployed to respond to the LLNL Health Services facility or local hospitals for examination of personnel that may have radioactive materials within a wound.

The typical detection levels in using the LLNL portable wound counter in a low background area is 0.4 nCi to 0.6 nCi assuming a near zero mass source.

This paper documents the systematic errors associated with in vivo measurement of radioactive materials buried within wounds using the LLNL portable wound measurement system. These errors are divided into two basic categories, calibration errors and in vivo wound measurement errors. Within these categories, there are errors associated with particle self-absorption of photons, overlying tissue thickness, source distribution within the wound, and count errors. These errors have been examined and can cause significant issues when interpreting the measurement data.

## Calibration Errors:

Calibration errors include the following areas: calibration source photon self-absorption, errors in overlying tissue calibration (e.g., tissue composition and thickness), errors associated with the calibration source, and measurement error. Source photon self-absorption is most significant for dense materials with low specific activity, such as U-238.

### Errors Associated with Calibration Source Self-Absorption

The calibration of the LLNL wound counter utilizes near zero-mass point sources except for the $^{238}U$ calibration source. Uranium-238 calibration requires a large mass source to obtain sufficient counts for calibration. If the source material contained in a wound is significantly different from a near zero-mass point source, significant errors due to self-absorption are introduced (See the Wound Measurement Errors section of this paper for a more detailed discussion). Likewise, since a significant mass of uranium is required to calibrate the wound counting system for measuring uranium-containing wounds, the effect of a smaller/thinner source of uranium contained within a wound also introduces errors in the calibration of the wound counter.

Calibration of the LLNL wound counter for natural uranium typically uses a thin plate (0.3 mm) of $U_3O_8$ metal with a diameter of 2.5 cm. Operational experience has demonstrated that a source of this size is required for calibration in order to accumulate a sufficient count rate to minimize calibration count error. Monte Carlo simulations of 16.3 and 63 keV photons depositing in a 2.5 cm diameter detector from a 2.5 cm diameter disk of uranium were performed to determine the effect of self-absorption by the uranium calibration source.

Based on the estimated detection limited for uranium using the LLNL wound counting system, the minimum thickness of a detectable $^{238}U$ mass would be 0.5 mm. The change in the 63 keV photon efficiency from 0.05 mm thick disk source up to a 1 mm thick disk is shown in Fig2.

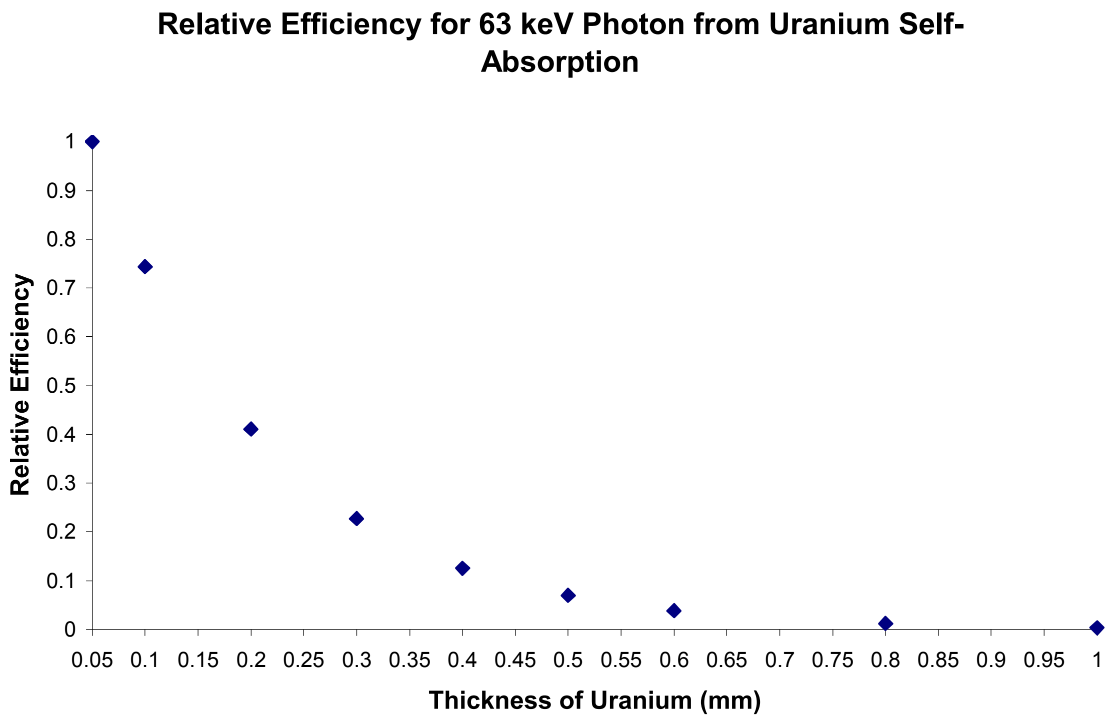


Fig. 2. The change in efficiency relative to a 0.05 mm thick source as a function of source thickness.

The error due to self-attenuation of the 63 keV photon through the 0.3 mm thick uranium disk used to calibrate the wound counting system will be approximately 77% relative to a 0.05 mm thin source. Similar errors can exist for other radionuclides such as plutonium and americium, however these radionuclides have a higher specific activity than uranium and measurable calibration sources are more likely to be near zero-mass sources. However if a wound were to have a significant mass of plutonium (e.g., a splinter of $^{239}$Pu), the near-zero mass calibration would present significant errors in the determination of activity in the wound.

## Errors associated with Calibration for Overlying Tissue Attenuation

A calibration correction is used when there is overlying tissue associated with a wound. The correction value is determined using known thickness of muscle equivalent material in conjunction with the calibration source. The error associated with the calibration correction for overlying tissue, assuming accurate determination of the source depth in the actual wound, typically ranges from 1% (@ 1mm depth) to 5% (@ 15 mm depth) for the 60 keV photon of $^{241}$Am.

The error associated with an unknown tissue composition can also introduce additional error for the lower energy photons. For the 16 - 17 keV photons, a 50/50 adipose to muscle ratio can introduce 2% error in the calibration efficiency at a nominal 1 mm tissue depth, 10% error at a nominal 5 mm depth, and 20% error at 15 mm depth.

The combined error in the efficiency for overlying tissue attenuation can be as high as 21% for the measurement of plutonium x-rays. For the measurement of 60 to 63 keV photons (such as Am-241 and U-238 via Th-234) the error in the efficiency is approximately 5%.

### Calibration Source Errors

The sources used for calibration of the wound counter are traceable to the National Institute of Standards. The errors associated with the certificate values of these sources are 1% for $^{238}$U, 1.3% for Plutonium, and 4% for $^{241}$Am.

### Calibration Count Errors

Calibration sources with sufficient activity to minimize count errors are used for routine calibration of the LLNL wound counting system. A measurement of 10,000 counts from the calibration source has an error of approximately 1%. For calibration sources used at LLNL the count error is typically less than 1.3%.

## Wound Measurement Errors

### Overlying Tissue Attenuation

Attenuation through overlying tissue can be significant; especially for the 17 keV photon associated with direct plutonium wound measurements. When calculating the activity in a wound, an estimate of the source depth is made. The more accurate the estimate of the source depth, the better the estimate of activity. The attenuation error associated with incorrect estimation of source depth is summarized in the following table (Table 1).

Table 1. Attenuation errors associated with inaccurate determination of overlying tissue thickness.

| Percent Error Associated with Tissue Attenuation | | | | | |
|---|---|---|---|---|---|
| **mm of under or over estimated tissue thickness** | **16 keV** | **17 keV** | **60 keV** | **63 keV** | **92 keV** |
| 0 | 0% | 0% | 0% | 0% | 0% |
| 1 | 12% | 10% | 2% | 2% | 2% |
| 2 | 23% | 20% | 4% | 4% | 3% |
| 3 | 32% | 28% | 6% | 6% | 5% |
| 4 | 40% | 35% | 7% | 7% | 7% |
| 5 | 48% | 42% | 9% | 9% | 8% |
| 6 | 54% | 48% | 11% | 11% | 10% |
| 7 | 59% | 53% | 13% | 12% | 11% |
| 8 | 64% | 58% | 14% | 14% | 13% |
| 9 | 69% | 63% | 16% | 16% | 14% |
| 10 | 72% | 66% | 17% | 17% | 16% |

## Geometry & Source Distribution

The calibration of the LLNL wound counter is based on a point source calibration (except $^{238}U$, which requires a relatively large source mass for calibration). If the source distribution in the wound is significantly different from a near zero-mass point source, then there are errors in the estimate of activity in the wound. The degree of error has been evaluated for a line source (such that the source distributed is along a vertical entry path of a wound) and as compared to a point source (Hickman, et. al., 1994). The possible errors are summarized in the table below (Table 2).

Table 2. Possible error associated with an unknown geometry.

| Maximum Penetration (mm) | Line-source to Point Source Ratio | % Error |
|---|---|---|
| 0 | 1.00 | 0 |
| 5 | 1.13 | 13 |
| 10 | 1.50 | 50 |
| 15 | 1.95 | 95 |
| 20 | 2.46 | 246 |
| 25 | 3.05 | 305 |
| 30 | 3.77 | 377 |

Wounds at 30mm are rare and most wound depths are much closer to the surface of the skin. However, the study by Hickman, et. al. was limited to just two basic distributions. Other distributions within a wound or at a wound site may introduce far greater errors than those determined for the two basic distributions. Therefore, it is reasonable to assume that distribution could introduce as much as 377% error in any measurement.

## Self Attenuation of Wound Deposited Material

When measuring uranium or other high-density radionuclides in a wound, an inherent assumption exists that the material in the wound is a zero-mass source. However when the specific activity is very small (i.e., the half-life of the radioisotope is extremely long), such as for uranium, the assumption that the source inside a wound has zero-mass can be in error. Density of the source dictates the degree of error that can be encountered when the source in the wound has mass. A high-density radionuclide such as uranium, when measurable, will typically have sufficient mass to affect the measurement of $^{238}$U in a wound. This mass of $^{238}$U can cause significant self-absorption and error in the estimated quantity of $^{238}$U in the wound. Particle self-absorption for $^{238}$U and other high-density materials can be significant. The following table summarizes the degree of error encountered for a $^{238}$U source (with $^{234}$Th in equilibrium with the $^{238}$U) of various diameters (i.e., mass).

Table 3. Self-attenuation errors associated with Uranium for an unknown source diameter/thickness.

| Diameter (mm) | Transmission | *%Error (from near zero mass source)* |
|---|---|---|
| **16 KeV** | | |
| 0 | 1.00E+00 | 0% |
| 0.05 | 7.65E-01 | 23% |
| 0.1 | 5.86E-01 | 41% |
| 0.5 | 6.89E-02 | 93% |
| 1 | 4.75E-03 | 211% |
| 1.5 | 3.27E-04 | 3000% |
| 2 | 2.25E-05 | >4000% |
| | | |
| **63 keV** | | |
| 0 | 1.00E+00 | 0% |
| 0.05 | 9.86E-01 | 1% |
| 0.1 | 9.72E-01 | 3% |
| 0.5 | 8.68E-01 | 13% |
| 1 | 7.53E-01 | 25% |
| 1.5 | 6.53E-01 | 35% |
| 2 | 5.67E-01 | 43% |
| | | |
| **92 keV** | | |
| 0 | 1.00E+00 | 0% |
| 0.05 | 9.95E-01 | 1% |
| 0.1 | 9.90E-01 | 1% |
| 0.5 | 9.49E-01 | 5% |
| 1 | 9.00E-01 | 10% |
| 1.5 | 8.54E-01 | 15% |
| 2 | 8.10E-01 | 19% |

The significance of the error associated with particle self-absorption becomes apparent when evaluating the volume, mass, and $^{238}U$ activity of a sphere of $U_3O_8$. Table 2 provides a comparison of the $^{238}U$ activity as a function of particle size.

The typical uranium detection level for the LLNL wound monitoring system is 0.4 nCi to 0.6 nCi assuming a zero mass source and no or little overlying tissue. Table 4 demonstrates that a sphere with a diameter less than approximately 0.7 mm is not detected using the portable wound monitoring system.

Table 4. $^{238}U$ activity as a function of the size, volume, and mass of a $U_3O_8$ particle.

| Diameter (mm) | Volume (cc) | Mass (g) | Activity (nCi) |
|---|---|---|---|
| 0.05 | 6.54E-08 | 1.23E-06 | 4.14E-04 |
| 0.1 | 5.24E-07 | 9.84E-06 | 3.31E-03 |
| 0.5 | 6.54E-05 | 1.23E-03 | 4.14E-01 |
| 0.7 | 1.45E-04 | 1.20E-03 | 4.00E-01 |
| 1 | 5.24E-04 | 9.84E-03 | 3.31E+00 |
| 1.5 | 1.77E-03 | 3.32E-02 | 1.12E+01 |
| 2 | 4.19E-03 | 7.87E-02 | 2.65E+01 |

Using the 63 keV photon from $^{234}Th$ in equilibrium with $^{238}U$, the errors associated with the measurement of $^{238}U$ in a wound can be 13% to greater than 43% due to self-absorption. If the determination of $^{238}U$ activity is purely based on the 16 keV x-ray from uranium, then the error due to self-attenuation at the detection level can be 93%.

Similar self absorption errors can be encountered with the direct measurement of the 17 keV photon emitted from Plutonium and the 59.5 keV photon of $^{241}Am$ when the source in the wound has significant mass (e.g., a splinter of material embedded inside the wound). The higher specific activities of Pu-239 and Am-241 provide lower particle mass and lower self-absorption than for uranium. Therefore, a maximum self-absorption at 59.5 keV is approximately 1.4% for Americium metal with an activity of 0.4 nCi. The 17 keV photon from Plutonium isotopes is easily attenuated and can be readily self-absorbed by a small mass of Plutonium metal. The self-absorption for pure Plutonium is evaluated to be 27% at a detected activity of 0.4 nCi.

## Count Error

The calculation of activity for the wound counter currently propagates the count error and efficiency count errors. The efficiency count errors are typically 0.5% to 3%. For count rates near background the count error is typically 10% to 15% depending on the extraneous background that is present in the count area. As the activity increases, the relative count error decreases, thereby making the count error less than 10% to 15% at activity levels above background.

## Total Errors Associate with Wound Counting:

The calculations for wound measurements at LLNL only incorporate the source count errors, wound count errors, and the wound depth calibration errors.

Table 5 summarizes the expected range of errors that may be encountered when performing a wound measurement where the material is detectable and deposited at 0 to 5 mm deep within a wound. As the depth increases, the errors will significantly increase. Without precise determination the source distribution and depth within the wound, the degree to which the following errors impact a wound measurement is largely unknown.

Table 5. Summary of errors associated with wound counting.

| Source of Error | Pu-239 | Am-241 | U-238 |
|---|---|---|---|
| *Calibration:* | | | |
| Calibration Source Self-Absorption Errors | <1% | <1% | 77% |
| Correction for Overlying Tissue[1] | 21% | 5% | 5% |
| Calibration Source Certification Error | 1.3% | 4% | 1% |
| Calibration Source Count Error | 1% | 1% | 1% |
| Total Calibration Error: | 21% | 6.5% | 77.2% |
| *Wound Measurement:* | | | |
| Overlying Tissue Absorption | 66% | 17% | 72% |
| Particle Self Absorption[2] | 27% | 1.4% | 93% |
| Geometry | 377% | 377% | 377% |
| Count Error | 15% | 15% | 15% |
| **Total Error (all sources):** | **385%** | **378%** | **403%** |

## Alternatives and Method Improvement

The largest sources of error in any wound measurement are (1) counting geometry/source distribution, (2) particle self-absorption, and (3) overlying tissue depth. Knowing the source distribution within the wound, the amount of overlying tissue, and the size/composition of the particle in the wound would allow for significantly improved calibrations, using empirical or Monte Carlo calibration methods. However, these three parameters are the most difficult parameters to elucidate while the material is resident in the wound. Removal of the wound activity with subsequent dissection and incremental analysis retrospectively allows for improved activity estimates.

Modest improvement in wound measurement is obtainable under appropriate circumstances. The use of a high-resolution detector can provide the ability to assess ratios of individual photons that are not otherwise resolved using a NaI detector. These ratios, coupled with Monte Carlo simulation, could be used to assess the depth of radioactive material in a wound. However, the accuracy of this type of assessment will

[1] Empirically determined using 15 mm of tissue overlay
[2] The true error can exceed these error determinations for large particles of low specific activity

depend on the accurate simulation of the unknown source density (self-absorption) and source distribution relative to the detector (geometry) within the wound. Thus, the degree of improvement would be minor since particle self-absorption and geometry will still provide large sources of error.

Using a high-resolution detector (such as High Purity Germanium) would allow for a more simplified calibration using a multi-gamma near-zero mass calibration source. However, calibration verification measurements will disagree when performing verification measurements on low specific activity/high density materials such as uranium. Uranium verification sources will have significant self-absorption. Likewise, the use of adequate high-resolution technologies with sufficient surface area to reduce geometry errors will be cumbersome to maintain and transport so long as portability is required.

The sensitivity of the detection can be improved by performing wound counting in a well shielded (low background area), or by adding shielding to the existing counter. However, the improvement is realized more in the detectability of radionuclides in the wound rather than significant improvement in the errors associated with wound counting. Another factor with large amounts of shielding is that additional shielding makes the wound counting system less portable.

## Conclusions

There are significant systematic errors when performing wound measurement. Under certain circumstances, these errors can be orders of magnitude greater than the 'typical' errors generated in Table 5. Under ideal and well-known circumstances, direct wound measurement may provide reasonably accurate results suitable for dosimetric determinations; however direct wound measurement is an extremely limited tool. Given the errors associated with wound measurement, a wound measurement system is better suited for qualitative determinations rather than quantitative measurements. Even with a non-portable, high resolution, shielded wound measurement system, a 'detect' versus 'non-detect' determination of radioactivity in the wound is much more appropriate, than the actual determination of activity in a wound.

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