Introduction to Uncertainty Calculations in Radioactive Sources

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Objectives

- To understand the origin of the components that result in the uncertainty value associated with a calibrated source’s activity and/or output value.
- To understand the magnitude of these components and thus their relative contributions to the overall uncertainty value(s).
  - a source can have more than one calibrated characteristic and thus more than one uncertainty value associated with it
- To understand how to numerically combine these components’ values to calculate an overall uncertainty value.
  - not here to learn math
  - plenty of examples in the external literature

A Thought to Begin

There are two types of problems, those that are impossible and those that are trivial. When a solution is found for an impossible problem, the problem becomes trivial.

Outline

- The meaning of uncertainty
- Measurement components leading to uncertainty
- Which components apply to which source types
- Which components apply to which instruments
- Which components apply to which calibration methods
- Example calculation
- Final thoughts
- References

What Does an Uncertainty Value Mean?

- No radioactivity measurement is without uncertainty
- An uncertainty value is only an estimate
- An uncertainty value is of little worth without an indication of the likelihood the correct value falls within the activity/output range indicated by the uncertainty value (e.g., 10 ± 1 means a range of 9 -11)
  - a confidence level must be stated with an uncertainty value, typically 95% or greater

Relative Expanded Uncertainty

- Confidence level
  - k value
    - 0.6745 = 50%
    - 1.0000 = 68.3%
    - 1.6449 = 90.0%
    - 1.9600 = 95.0%
    - 2.0000 = 95.5%
    - 2.5758 = 99.0%
    - 3.0000 = 99.7%
  - most common values are 2 and 2.58
    - most national labs use k = 2 (“~95%”)
    - Eckert & Ziegler Isotope Products uses k = 2.58 (“99%”)
    - 10.0 ± 2.0 at 95% and 10.0 ± 2.6 at 99% are equivalent

Uncertainty Value Starting Point for Sources

- A national metrology institute (NMI) such as NIST must produce a source activity measurement with a properly determined uncertainty value using a primary measurement technique (e.g., 4πβ-γ) to produce a primary standard.
- An entity traceable to the NMI such as Eckert & Ziegler Isotope Products (EZIP) uses the primary measurement result to calibrate an instrument EZIP will use to make future measurements or a source it has manufactured (direct and comparator measurements, respectively) to produce a secondary standard.
- The secondary standard’s uncertainty value can’t be less than the uncertainty value of the primary standard but it can theoretically be statistically insignificantly larger than the primary standard’s uncertainty value, albeit highly unlikely.
- Both primary and secondary standards can be used to calibrate instrumentation in the laboratory or field.

Uncertainty Components

- Some of the components that go into a source’s activity uncertainty value are
  - radioactive decay
  - activity decay correction
  - background and “signal-to-noise” ratio
  - instrument stability
  - source configuration, including variation
  - source position relative to the detector, when applicable
  - duration of count, when applicable
  - weighing of active matrix (e.g., powder or solution)
    - NIST-traceable balance(s) necessary

Assessing Magnitude of Uncertainty Values

- Historical data
  - typical difference from known values
- Repeatability measurements
  - multiple source measurements using consecutive counts
- Reproducibility measurements
  - multiple source measurements over a longer period of time
    - typically involves multiple days or longer
    - involves placing and replacing the source (reproducing count geometry)
- Intercomparisons with one or more national metrology institutes
  - requirement to be formally traceable to NIST, etc.

Assessing Magnitude of Uncertainty Values-cont.

- Known instrument limitations
  - readability of output
  - technical specifications
  - geometric configuration
- Equipment and accessory variation
  - source holder placement
    - counting stands for gamma ray spectrometers
  - component movement
    - drawer on gas flow proportional counter
- Location effects on instrumentation/equipment
  - analytical balance in a gloveless box

Traceability to NIST

- Clearly declared uncertainty values required for intercomparison tests with an NMI such as NIST
  - ANSI N42.22:1995 has six categories for source traceability
    - alpha particle sources for total alpha activity
    - alpha particle sources used for high-resolution alpha spectrometry
    - beta particle emission sources with $E_{avg} < 100$ keV
    - beta particle emission sources with $E_{avg} > 100$ keV
    - gamma-ray emission sources with energies < 250 keV
    - gamma-ray emission sources with energies >250 keV
  - each intercomparison test requires an uncertainty value specific to the test
    - uncertainty value specified for each test is the minimum uncertainty value allowed for sources supplied by the source provider

Radioactive Decay

- Poisson distribution
  - standard deviation = (events)^{½}
    - 4 times the events leads to doubling the precision (i.e., “halving” the uncertainty)
  - precision improvement reaches a “point of diminishing returns”
    - source calibration is often a balance between throughput and precision
  - counting statistics for a standard source ideally does not significantly statistically increase the uncertainty value for a production source calibration
    - standard sources typically should not have activity levels at the lower limits of an instrument’s capabilities

Decay Correction

- Two factors
  - uncertainty value for half-life
    - strictly dependent on nuclear data parameter measurements
  - amount of correction
    - typically not a large contributor to the overall uncertainty value
  - large relative half life value uncertainty combined with significant decay correction leads to statistically significant increase in total uncertainty value
    - rarely a significant contributor to the overall uncertainty value

Background/Noise

- Signal-to-Noise ratio influences multiple aspects
  - source strength
    - source activity should be sufficiently high relative to the background level
  - source-to-detector distance
    - typically photon measurements only
    - larger source-to-detector distances reduce uncertainty in terms of source placement reproducibility but decrease counting statistics which increases relative uncertainty values unless longer count times are used
  - count duration
    - greater benefit for short duration measurements but short duration counts have larger relative uncertainty values in terms of count rate but not a major factor in most cases

Source Configuration

- Point sources
  - placement of active material (i.e., reproducibility of placement within holder)
  - density of support matrix
- Planar sources
  - shape (i.e., round vs. square vs. rectangular)
  - backing material
- Large volume sources
  - applies almost exclusively to gamma emitting sources
  - matrix density affects low-energy measurements most
  - container material increasingly important with higher Z
- Matching standard source configuration to unknown source configuration reduces total uncertainty value

Instruments Used for Measurements

- Photon measurements
  - reentrant ionization chambers (current mode instrument)
    - gamma rays
  - germanium detectors (e.g., HPGe) and NaI(Tl) detectors
    - gamma rays
  - Si(Li) detectors
    - x-rays
- Particle measurements
  - gas flow proportional counter
    - alpha and beta particle emission sources
  - liquid scintillation counter
    - alpha and beta contained activity sources
  - surface barrier detector
    - alpha particle emission sources only, energy and activity value

Source Position

- Source dimensions vs. detector dimensions
  - solid angle subtended
  - edge effects
  - depth of activity relative to source surface
- Often a significant contributor to contained activity value uncertainty for environment samples due to small source-to-detector distance
  - Marinelli beakers
  - filter papers

Instrument Stability

- Temperature
  - NaI(Tl) typically most affected instrument
  - thermal equilibrium is important for all instruments
- Humidity
  - particle emission source measurements most affected
- Power supply
  - voltage bias stability important for plateaus

Weighing

- Absolute weighing
  - – not typically used because active matrix must be contained in some object if for no other reason than to transfer the activity to final container
- Weighing by difference
  - – relative difference in container mass vs. active matrix mass
    - weighing a pipette tip full and then empty typically leads to less uncertainty than weighing solution in pipette by adding solution directly to container
    - typical pipette tip is < 1g but some containers are >50g

Particle Counting

- Branching Ratio
  - strictly dependent on nuclear data parameter measurements
  - detector efficiency for detecting emitted particle
- Source efficiency for emitting particles from source surface
  - typically an experimentally determined value
- Contained activity value and surface emission rate value are typically linearly correlated for alpha sources but independent for beta sources
  - surface emission rate for solid alpha sources used to determine contained activity
  - activity gravimetrically deposited for beta sources while surface emission rate is measured directly

Photon Counting

- Detector efficiency
  - direct, energy point for energy point calibration of detector
  - curve fitting for a set of efficiency points
    - often one of the biggest components of uncertainty in photon measurement
- Branching ratio
  - strictly dependent on nuclear data parameter measurements
    - use of different nuclear data parameter sets without proper correction can lead to additional uncertainty, which is unnecessary
- Use of comparator method eliminates some uncertainty components (e.g., detector efficiency and branching ratio) but is costly and time consuming
  - matching standard needed for each source configuration and radionuclide

Uncertainty Propagation

- Total uncertainty value only increases
- Formal definition

  $(\sigma_u)^2 = (\delta_u/\delta_x)^2(\sigma_x)^2 + (\delta_u/\delta_y)^2(\sigma_y)^2 + (\delta_u/\delta_z)^2(\sigma_z)^2 + \ldots$
- Addition/subtraction

  $(A \pm a) +/- (B \pm b) = A +/- B \pm (a^2 + b^2)^{½}$
- Multiplication/division

  $(A \pm a) * (B \pm b) = A*B \pm A*B*[(a/A)^2 + (b/B)^2]^{½}$

  $(A \pm a) / (B \pm b) = A/B \pm A/B*[(a/A)^2 + (b/B)^2]^{½}$
- Exponentiation

  $e^{\lambda A} = e^{\lambda A}\lambda\sigma_A$ (used when half life precisely known)

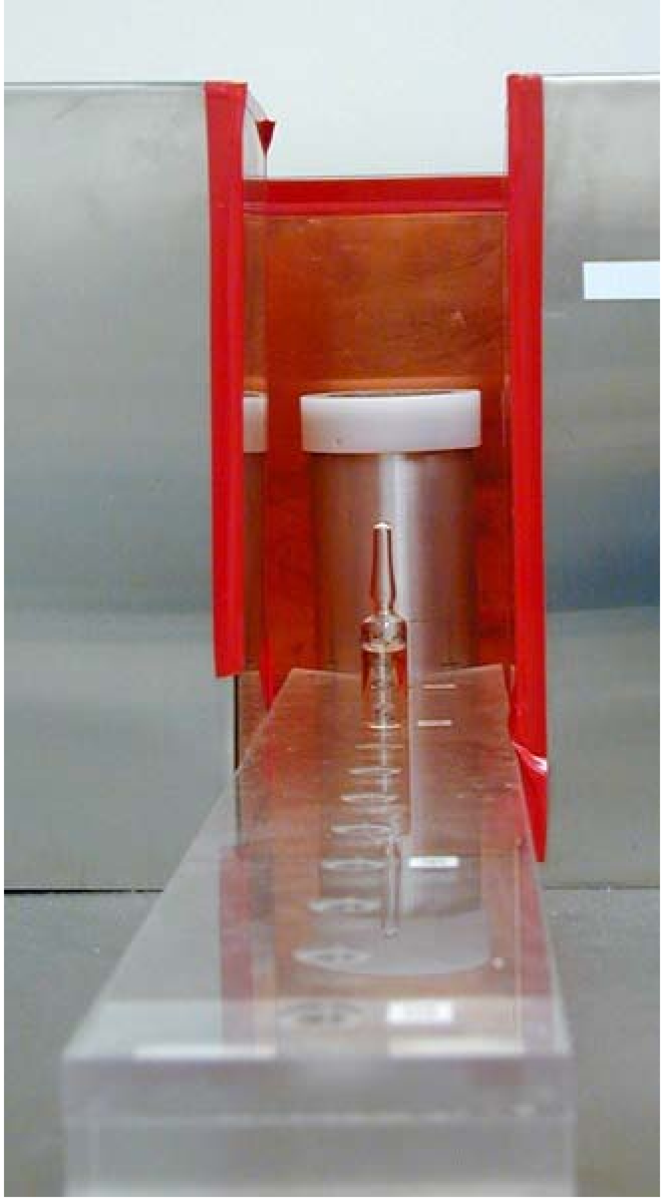
Combined Statistical and Relative Expanded Uncertainty

- The result of the propagation of uncertainty is the combined statistical uncertainty (CSU)
  - combined statistical uncertainty is given at 1σ
- The relative expanded uncertainty (REU), the final uncertainty value provided with source, is the product of the combined statistical uncertainty (CSU) times the coverage factor, k:

$$\mathbf{REU = k * CSU}$$

Example:
Gamma Spectrometry-Beaker

Example:
Gamma Spectrometry-Ampoule



Example:
Gamma Spectrometry-cont.

$$A_u = \{C_u F_u I P_u e^{-[\ln(2)]t/T}\}/\{EBD_u\}$$

where

A_u = activity of unknown
C_u = counts of unknown
F_u = fit of peak for unknown
I = instrument stability
P_u = position of unknown
t = time of decay (days)
T = half life for nuclide (days)
E = gamma ray detector efficiency
B = branching ratio
D_u = duration of unknown count

Distributions

- Normal (i.e., Gaussian)
  - variation probability from the true value follows the “bell curve”
- Rectangular
  - variation probability from the true value is equally likely regardless of variation magnitude
  - typically for measurement results at the limit of readability
    - mass determination is an example
- Triangular
  - special case of rectangular distribution
    - mass determination by weighing difference is an example

Example:
Gamma Spectrometry-cont.

Qty	Definition	Value	% Unc (k=1)	Standard Uncertainty	Distribution	Sensitivity	Uncertainty Contribution
C_u	unknown counts	1214076	0.193	2343.167	Normal	0.0174	40.79318
E	detector efficiency	0.04133	1.0	0.00041	Normal	511404.9079	211.36365
t	decay (d)	321	-	0.1	Rectangular	1.71468	0.17147
T	half life (d)	4933	-	11	Normal	0.19326	2.12584
B	branching ratio	0.2657	-	0.0011	Normal	79549.73595	87.50471
D_u	unknown count duration	5000	0.01	0.5	Rectangular	2.44062	1.22031
F_u	unknown peak fit	1	1.0	0.010	Triangular	8628.8848	86.28885
I	instrument stability	1	0.1	0.001	Rectangular	12203.0859	12.20309
P_u	unknown position	1	0.1	0.001	Rectangular	12203.0859	12.20309
A_u	unknown activity	21136.36	-	-	-	-	496.97
						REU (k=2)	**2.35%**

Take Home Message

- Most of the time, the majority of a measurement uncertainty value is the result of only a few components.
- It is better to overestimate an uncertainty value than to underestimate it but one should make the best estimate possible given the objective evidence available.
- There is no substitute for experience when there is not enough data and estimations must be made.

References

- NIST Technical Note 1297-1994 Edition, “Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results”
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