Contract No. and Disclaimer:

This manuscript has been authored by Savannah River Nuclear Solutions, LLC under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting this article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for United States Government purposes.

Ruminations on NDA Measurement Uncertainty Compared to DA Uncertainty

Saleem Salaymeh[1], Stephen Croft[2], Steve Goldberg[3], Lynne Preston[4], Peter Santi[2], James Sprinkle[2], Ram Venkataraman[5], Glenn Pfennigwerth[6], Raymond Dewberry[1], Aaron Clare[1]
[1]Savannah River National Laboratory, [2]Los Alamos National Laboratory,
[3]JDOE- New Brunswick Laboratory, [4]DOE-HS-82, [5]CANBERRA, [6]Y-12

ABSTRACT

It is difficult to overestimate the importance that physical measurements performed with nondestructive assay instruments play throughout the nuclear fuel cycle. They underpin decision making in many areas and support: criticality safety, radiation protection, process control, safeguards, facility compliance, and waste measurements. No physical measurement is complete or indeed meaningful, without a defensible and appropriate accompanying statement of uncertainties and how they combine to define the confidence in the results. The uncertainty budget should also be broken down in sufficient detail suitable for subsequent uses to which the nondestructive assay (NDA) results will be applied.

Creating an uncertainty budget and estimating the total measurement uncertainty can often be an involved process, especially for non routine situations. This is because data interpretation often involves complex algorithms and logic combined in a highly intertwined way. The methods often call on a multitude of input data subject to human oversight.

These characteristics can be confusing and pose a barrier to developing and understanding between experts and data consumers. ASTM subcommittee C26-10 recognized this problem in the context of how to summarize and express precision and bias performance across the range of standards and guides it maintains. In order to create a unified approach consistent with modern practice and embracing the continuous improvement philosophy a consensus arose to prepare a procedure covering the estimation and reporting of uncertainties in non destructive assay of nuclear materials. This paper outlines the needs analysis, objectives and on-going development efforts. In addition to emphasizing some of the unique challenges and opportunities facing the NDA community we hope this article will encourage dialog and sharing of best practice and furthermore motivate developers to revisit the treatment of measurement uncertainty.

INTODUCTION

Every measurement has an uncertainty associated with it, resulting from errors arising in the various stages of sampling and analysis and from imperfect knowledge of factors affecting the result. An uncertainty is therefore an assessment of the quality of a measurement and is necessary when comparing results, particularly among different laboratories or among different techniques. Total measurement uncertainty (TMU) is the property of a specific value or result generated by a particular method of analysis. In contrast, bias and precision are properties of the measurement method. Uncertainty depends on many factor including; the repeatability of the instrument, on the reproducibility of the result over time, on the number of measurements in the test result, and on all sources of random and systematic error that could contribute to a deviation from the true value. Different customers have had different statistical tools which they favor. For example some customers separate the calibration error or long term bias for separate treatment that is they specify not to include it in the reported measurement uncertainty for a single measurement result.

Although a measurement process attempts to determine the value of a physical or chemical property, the actual measurement value is only an estimate of the true value. Measurement uncertainty characterizes the range of values within which the true value is expected to lie, with a specified level of confidence. Because uncertainty is intimately associated with a measurement or measurement process, it should be considered to be an integral part of the measurement result. A stated value from a measurement has limited basis of reliability without a statement of its associated uncertainty.

Given the global nature of commerce and technology and their dependence on measurements, a method of estimating uncertainty should be universally adopted and applied. The *Guide to the Expression of Uncertainty in Measurement* (GUM), published by the International Organization for Standardization (ISO) and adopted by the American National Standards Institute as an American National Standard (ANSI), provides the current international consensus method for estimating measurement uncertainty. This guide, for the most part seems to address and serve the destructive analysis (DA) community. An important advantage of GUM is that it provides a common framework to compare measurement results from different laboratories or different analytical methods. Because uncertainties that follow GUM principles are in general transparent and comparable, an assessment of different results can be made on the basis of the two values agreeing within the uncertainty of their measurements. The appropriate use of GUM could help to reconcile stockpile accountancy or shipper-receiver differences in measurements. It establishes general rules for evaluating and expressing uncertainty for a wide range of measurements, is applicable to calibration and test results, and forms the basis for accreditation requirements relating to measurement uncertainty estimation.
The advent of adopting this guide by ANSI has lead to many discussions among the Non Destructive Assay (NDA) professionals. In particular, among the members of the ASTM C26.10, Subcommittee for Developing Standards on NDA. As a result of these discussions the NDA User's Group decided to hold a workshop in conjunction with the C26.10 subcommittee in San Antonio, TX, January 25th - 28th, 2010. The workshop discussions were focused on aspects of expression of uncertainty for NDA Measurements. The workshop consisted of three major parts; the first was presentations, the second was interactive breakout sessions, and third was for the breakout session leaders to meet and prepare a joint statement. The topics of the presentations included the following titles;

- Summary of GUM,
- Statistical issues in the expression of NDA uncertainty,

- "TMU: It's more than statistics", and
- Linear Calibration

The workshop had four interactive breakout sessions listed below;

- Comparison of Measurement Uncertainty for DA and NDA,
- Lessons Learned from applying Measurement Uncertainty to NDA and DA,
- Compile a list of special situations (geometries, matrixes,…etc.) for which Uncertainty is difficult to assign, and
- Do we need an ASTM Standard and Why?

The purpose of the breakout session meeting was to prepare a joint statement on the results of the workshop. The workshop was well attended by representatives from; national laboratories, private industry, regulators and users.

This paper summarizes the workshop and suggests a path forward for the NDA User's Group and ASTM to move in the direction of developing a Standard on Expression of Uncertainties in NDA measurements.

Workshop Summary

In this section we will provide a detailed summary of the workshop. The first part of the workshop contained four presentations that intended to get the attendees to think about the GUM approach and its applicability to both NDA and DA. Following are the highlights of these presentations:

1. The first presentation was entitled "Summary of GUM" by Steve Goldberg, NBL. Topics covered in the presentation included:
    - ISO Guide (1995)
    - ISO 17025: Laboratory Accreditation
    - Example: interdiction of material in several different countries. Is it the same source?
    - Eurachem/CITAC Guide – quantifying uncertainty in analytical measurements.
    - Metrodata GmbH software workbench.
    - How NDA is may be different from DA
2. The second presentation was entitled "Statistical Issues in the Expression of NDA Uncertainty" by Tom Burr, LANL. Topics covered in the presentation included:
    - Taught the course "Statistics in NDA" at LANL
    - "Item specific bias": how to estimate?
    - Dave Beddingfield – DSTA- source reduction algorithm.
    - Unmodelled effects will affect the result in one way for "A" set of items, and in another way for "B" set of items!
    - Many questions raised on how to evaluate/or reduce uncertainties….and how to convince "customers" that you really

need the budget that you've requested to accomplish the evaluation.

- The need for regular PDP for safeguards measurements.

3. The third presentation was entitled "TMU: It's more than statistics by John Kirkpatrick, CANBERRA. Topics covered in the presentation included:
    - Example: source distribution uncertainty (point sources in a drum)
    - Calibration functions – normally neglect covariation matrix.
    - We need to be careful with the covariances. Should we go to Bayesian ISO 11929, how to deal with asymmetry of the distributions (non-gaussian).
    - Construction of a thorough, justifiable error budget is crucial.
    - How do we calculate TMU in the region near the lower limit of detection (LLD), minimum detectable activity (MDA)?
4. The fourth presentation was entitled "Linear Calibration" by Brian Young, CANBERRA. Topics covered in the presentation included:
    - Things the Deming code and MATLAB would not solve
    - Uncertainties in both x and y
    - York—1966—fitting a straight line, exactly.
    - Use Excel Solver for "black box" optimization

The second part of the workshop contained four interactive breakout sessions with the objective of compiling ideas and thoughts related to how we can provide personnel involved in performing, analyzing, and managing NDA measurements and users of NDA results (safeguards, MCA, criticality, waste management and certification, process control) a general understanding of the main components that contribute to uncertainty in NDA and DA measurements, and discuss the similarities and differences on how uncertainties are propagated in NDA and DA measurement techniques. The workshop was marked by vigorous discussions that included many examples and points of views that can not be covered in detail below. The following are highlights of these discussions:

I. Need for ASTM Standard Practice on Calculating and Propagating Uncertainty in NDA
    A. Definition of Practice
    B. Reasons for Standard
        i. First place for NDA user/expert to determine uncertainties
        ii. Communicate to NDA professionals and users the appropriate techniques of assigning uncertainties to measurements.
        iii. Provide understanding of GUM principles to NDA audience.

iv. GUM provides necessary foundation, but not necessarily sufficient for all NDA uncertainties
    1. doesn't cover issues that are the largest difficulties with determining TMU (no replicate measurements, limited representative standards, difficult to maintain traceability to certified standards)
    2. "GUM+" (Modeling guide to help provide information on uncertainties for modeling)

v. Standard would be means to update best measurement practice, and to unify and clarify different approaches to uncertainty calculations that are seen in the various C26.10 and N15 Guides and Standards, as well as in the various facilities and vendors.

vi. Provide a defensible and streamlined approach to TMU

C. Possible Scopes of New Standard;
   i. Pointer to literature,
   ii. Very detailed standard that would be applicable to all most of NDA techniques, and
   iii. Presentation of a good scientific approach to propagating and documenting uncertainty using GUM.

II. Comparison of Uncertainties in DA and NDA

A. The differences between uncertainties in DA and NDA are primarily due to the fact that in DA the sample is modified to match the analysis technique allowing for more control or measure of the measurement conditions, while in NDA the analysis technique is modified to match the item and measurement conditions.

B. Standards used in DA are often more similer to the sample being measured than NDA. This is due to the fact that it is not feasible to prepare a set of standards (isotopics, matrix, packaging, etc) to fit all NDA measurement regimes. The envelope of operation for a given method/instrument has to be determined through performance testing and evaluation. (Validation for DA, method qualification for NDA)

C. Address all known uncertainties for the measurement methodology in a manner that is consistent with the principles of GUM.

D. Uniform requirements for documenting all measurement results are desirable, independent of the method (NDA or DA) or instrument. The reporting should include a description of the methods used, assumptions that are made, treatment of the sample, standards used for calibration and measurement control, etc. (eg. Specifying whether error is one sigma, two sigma, etc)

E. To present a general comparison of expression of uncertainty between DA and NDA, we need to make sure that the comparison is between similar quantities. (apples to apples). For example, when samples are collected and DA results are reported, sampling uncertainty is often not carried through the entire calculation. In other cases, NDA uncertainties may not be adequately studied and evaluated to propagate through to the total measurement uncertainty. (e.g. uncertainties due to gamma-ray measurements which only sample the surface of an item.)
F. NDA requires process knowledge in order to determine certain components of the uncertainty while DA requires process knowledge in order to prepare and measure the sample.
G. DA has interlaboratory comparison programs to help provide defensible justification for the uncertainties associated with their measurement technique, NDA in general does not have such an exchange. (Exceptions being CALEX program operated by NBL and Waste Isolation Pilot Plant Performance Demonstration Project program which have limited activity and for Pu only)
H. A case study is included to demonstrate handling uncertainties in detail. The case study compares NaI-based Uranium Enrichment meter vs Thermal Isolation Mass Spectroscopy (TIMS).
    i. Similarities
        1. Both techniques are calibrated using traceable standards
        2. Multi-point linear calibration
        3. Both assume sample/item homogeneous composition
    ii. Differences (not a list of pros and cons)
        1. Standards not necessarily indicative of actual measured material for the enrichment meter technique.
        2. TIMS involves sample prep, NDA involves no Sample prep
        3. Number of assumptions for NDA methods is greater than for DA methods.
        4. TIMS, major portion of uncertainty is from standards. For NDA, the major portion of uncertainty is from factors other than the standards (statistics for instance)
        5. TIMS doesn’t care what the original material configuration was, so the uncertainty estimate is assumed to be the same between samples. NDA

has various factors to include into the uncertainty estimate. Uncertainties “may be” significantly different between measurements.

I. Similar comparisons can be done using the Handbook of Nuclear Safeguards Measurement Methods NUREG/CR-**2078** which has a table of various methods and statements of uncertainties. This table can be used as a springboard for intercomparison of specific “widely-used” methods.

III. Cases where it is difficult to assign uncertainties in NDA

A. Un recognized Human error and poor practices causes problems in both DA and NDA
B. Mathematically complex models that don’t allow analytical uncertainty analysis (measured inputs into reported results)
C. Geometry not well defined
    - Source location
    - Passive attenuation (drum walls, variations and thickness or corrosion)
    - Source self-attenuation (lumps, especially U)
    - Matrix voids/concentrations
    - Fill height
D. Correlations, interferences – nuclear data & fingerprints
E. Sources of “signal” other than the material you want to measure
F. Methods of determination of background
G. Quantification of fluctuating background
H. Blind spots in the instrument/process example is the field of view during hold up measurements
I. Spatial control (access, control)
J. Unique items or differences within/between calibration materials, measured items
K. Changes to measurement systems not identified by QA checks
    i. Crystal dead layer migration
    ii. Instrumental drift
L. Uncertainties reported by isotopic codes are often approximations valid in a limited range
M. Age of item
    i. Secular equilibrium
N. Mixed items (SNM or matrices)
O. Non homogenous isotopic composition
P. MOX as a new fuel type may challenge the traditional methods
Q. Propagation of error
    i. Gamma Efficiency calibration has strong input data associated e.g. two nuclides -> 14 lines and same sources -> 5 densities
R. Modeling geometry of holdup

S. Any non-automated (custom, one-of-a-kind) measurement situation – 99% of holdup and D&D measurements

CONCLUSIONS

The uncertainty is intimately associated with a measurement or measurement process, and it should be considered to be an integral part of the measurement result. A stated value from a measurement has limited basis of reliability without a statement of its uncertainty.

The differences between uncertainties in DA and NDA are primarily due to the fact that in DA the sample is modified to match the analysis technique allowing for more control or measure of the measurement conditions, while in NDA the analysis technique is modified to match the item and measurement conditions.

Standards used in DA are much closer to the sample being measured than NDA. This is due to the fact that it is not feasible to prepare a set of standards (isotopics, matrix, packaging, etc) to fit all NDA measurement regimes. The envelope of operation for a given method/instrument has to be determined through performance testing and evaluation. (Validation for DA, method qualification for NDA)

The NDA user's group workshop held in January 2010 was very successful in accomplishing the objectives established for it. The over all objective of this workshop was to facilitate providing personnel involved in performing, analyzing, and managing NDA measurements and users of NDA results (safeguards, MCA, criticality, waste management and certification, process control) a general understanding of the main components that contribute to uncertainty in NDA and DA measurements, and discuss the similarities and differences on how uncertainties are propagated in NDA and DA measurement techniques. The following are the main conclusions of the workshop:

- Established the need for ASTM Standard Practice on Calculating and Propagating Uncertainty in NDA,
- Provided a good discussion and considerations for Comparison of Uncertainties in DA and NDA,
- Provided a case study that demonstrates handling uncertainties in detail in NDA and DA, and
- Provide a list of special cases where it is difficult or not possible to assign uncertainty in NDA,
- Established the need for NDA interlaboratory comparison program,
- Established the need for NDA training and qualifications, and
- Theoretical approaches are necessary but must be tied to real world experience.

The authors are passionate about maintaining excellence and enhancing the state of the practice in NDA measurements and would like input from readers with ideas on how the proposed ASTM Standard Guide might best serve the NDA community.

AKNOWLEDGEMENTS
The authors would like to thank all the attendees of the workshop for their invaluable discussions and contributions. John Kirkpatrick, CANBERRA, Cynthia Gunn, Y-12, Thomas Sampson, LANL, David Dolin, SRNS, Tom Burr, LANL, Jeff Chapman, ORNL, Markku Koskelo, Aquila, Brian Young, CANBERRA, Brent McGinnis, ORNL, David Bracken, LANL, Richard Mayer, DOE, and Larry Berg, DOE-CNS.

REFERENCES

1. M. Franklin and J. Larisse, "Propagation of non-constant bias corrections in nuclear accounting.", Proc. 24th Annual Meeting of the INMM (Vol.XII), Vail, Colorado, USA, July 10-13, 1983.
2. Handbook of Nuclear Safeguards Measurement Methods NUREG/CR-**2078**
3. ANSI 15.36
4. ANSI 15.41
5. http://www.bipm.org/utils/common/documents/jcgm/JCGM_100_2008.pdf