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THE ROLE OF UNCERTAINTY ESTIMATION ON THE IMPROVEMENT OF MEASUREMENT PROCESSES AND RESULTS COMPARISON

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ABSTRACT

Nuclear material measurements for mass fraction and isotopic abundance are routinely conducted in safeguards analytical laboratories. New analytical methods, changes in experimental techniques, modern instrumentation, and new software packages for calculation of results and measurement uncertainties are introduced towards making constant improvements. Scientists and technicians must adapt to these changes, keep up with the advancement, and ensure obtaining analyses results with improved accuracy and precision with correct evaluation of uncertainties. Furthermore, it is important to communicate the experimentally determined values and the uncertainties in such a manner so that all information associated with the reported results are fully understood not only by its originator, but also by other users with a need to work with those results. Standard methods (according to ISO guidelines) for calculating and expressing measurement uncertainty are given in several publications such as the “Guide to Expression of Uncertainty in Measurement” (GUM) [1] and the “Quantifying Uncertainty in Analytical Measurements” (EURACHEM / CITAC Guide CG 4) [2]. Note that the evaluation of measurement uncertainty is also a central element in quality assurance programs. In this paper, we will discuss the importance of appropriate estimation of uncertainties with specific reference to improve measurement processes in nuclear material accountability measurements, and in evaluation of performance test results.

1. INTRODUCTION

An effective system for accounting and control of nuclear materials requires reliable and good quality measurements. The inventory of nuclear materials must be determined with appropriate precision and accuracy. Reliable conclusions about the disposition of the materials (material in hand, material transferred etc.) can be made only through obtaining adequate measurement results.

When a system of accounting and control of nuclear material is subject to verification, routine results from the facility operator and from independent verification measurements are compared. The personnel responsible for verifying the accuracy of the reported results need to assure that methods used in the comparisons are indeed reliable and defensible. The comparative studies must necessarily take into account uncertainty estimates in both routine and verification measurements. Inaccurate conclusions may lead to undesirable technical and political consequences.

The international scientific community active in the field of metrology, recognizing the need for uniformity in terminology and methodology, has published the "Guide to the Expression of Uncertainty in Measurement" [1]. This guide, known familiarly as “GUM”, is the most recent internationally-adopted convention in expressing and estimating measurement uncertainties. The standardization approach proposed by GUM intends to provide enough transparency to the process of uncertainty estimation and adequate tools to conduct an inter-comparison of measurement results. This comparison, as stated before, constitutes a central element in the process of quantifying and verifying nuclear material inventories at nuclear facilities. Therefore, it becomes essential that the GUM be considered in field of nuclear accountancy and safeguards.

2. THE INTERNATIONAL TARGET VALUES AND THE GUM

The concept of target values was first introduced in 1979 by the Working Group on Techniques and Standards for Destructive Analysis (WGDA) of the European Safeguards Research and Development Association (ESARDA) for measurement results from destructive analyses of nuclear materials. The objective was to establish international standards on expected uncertainty components for the operator´s measurements and for the independent inspectors’ verification measurements. The IAEA decided to adopt the idea and, since then, convened several advisory meetings to discuss and expand the concept leading to the publication of “International Target Values” (ITVs).

The latest version of the ITV´s, published in 2010 [3], is commonly referred to as ITV-2010. This document is extensively used by international and regional safeguards inspectorates (i.e. IAEA, ABACC and EURATOM), by analytical laboratories and in safeguards analytical inter-comparison programs. In this revision, international standards in estimating and expressing uncertainties have been considered while maintaining a format that allows comparison with the previous editions of the ITV´s. The ITV´s-2010 are expressed as a two component system – designated as random and systematic – that result in a single uncertainty estimate (ITV) for each material (U and Pu) in different forms, concentrations and isotopic compositions and methods of analyses for nuclear accountancy and verification purposes. ITV-2010 presents uncertainty values in tables, grouped as follows:

- Bulk and Density Measurements
- Sampling Uncertainties for Element Concentration and ^{235}U Abundance
- Uranium Element Concentration Measurements (DA)
- Plutonium Element Concentration Measurements (DA)
- ^{235}U Abundance Measurements (DA)
- ^{235}U Abundance Measurements (NDA)
- Plutonium Isotope Assay of Pu and U/Pu materials
- Total Mass of ^{235}U (direct NDA)
- Total Mass of Pu (direct NDA)

Similarly, the GUM approach yields a single value for the uncertainty, and recommends the preparation of a “budget” table that describes the relative contributions of all known sources that make up the total reported uncertainty. In other words, the GUM method includes uncertainties from the “traditional” random and systematic components, and in addition uncertainties from all other known sources (e.g., those associated with temperature, day-to-day and analyst-to-analyst variations). This detailed uncertainty expression makes it possible to conduct a consistent analysis of the reported result and appropriate pair comparison. It is important to note that previous versions of ITV´s are somewhat different in defining the target values; the two components (random and systematic) were considered to be separate in those publications.

3. ANALYSIS OF MEASUREMENT RESULTS IN INTER-LABORATORY EVALUATIONS

The main objectives of Measurement Evaluation Programs are performance evaluation of analytical measurement results, including several factors such as day-to-day variation, analyst-to-analyst variation, and instrument-to-instrument variation. Regulatory organizations have formally recognized the importance of measurement evaluation programs as a means to provide independent verification of the internal quality as practiced in safeguards measurement laboratories. Good quality control is essential for generating good quality analytical results.

In some of the measurement evaluation programs, as conducted until 2010, the test samples were evaluated for accuracy and precision with reference to the corresponding ITV´s-2000 [4] (previous version of the ITV´s) – i.e., in terms of "random and systematic components of uncertainties" only. Other sources of uncertainties were usually ignored; for example, uncertainties associated with characterized values for the test samples. In the GUM method, a comprehensive evaluation of uncertainties is possible. This difference between GUM based evaluation and the ITV-2000 method must be recognized. The difference is expected to be not appreciable if the uncertainty in the characterization of the test sample is small relative to uncertainties in the measurement results produced by the laboratory that is being tested.

In the field of nuclear safeguards, the analysis techniques have improved steadily due to advances in data analysis methods and through the use of “state-of-the-art” instruments. One possible consequence of these improvements is changes in the relative contributions of the various uncertainty sources that impact a specific measurement process. For example, in mass spectrometry measurements of fissile isotope abundances, the uncertainty contribution from reference materials (used to establish the traceability chain) is becoming an important contributor comparable to measurement uncertainties themselves. In consequence, the contribution of the reference material to the final uncertainty can no longer be ignored. The appropriate identification of the used reference material is also relevant due to possible correlation effects if different results are compared. This is one instance where the GUM method will yield a realistic estimation of uncertainty through inclusion of the reference value uncertainty contribution in the total.

4. UNCERTAINTY ESTIMATION AS A TOOL TO EVALUATE MEASUREMENT PROCESSES

The adequate estimation of the uncertainties associated with a measurement can provide relevant information about the process and support decisions aiming at its improvement and optimization. The following example illustrates it. Table 1 shows a typical uncertainty budget (a table showing the relative contributions of each input quantity to the final result) for the determination of total uranium content in a pure compound by destructive analysis using the titration technique [5]. In this method, the mass of uranium in one aliquot is determined based on the volume of titrant solution, with pre-established concentration, added to each the sample aliquot. Uranium reference material is used for bias correction and quality control. Relative standard uncertainties better of equal to 0.05% may be achieved.

Table 1. Typical Uncertainty Budget for Uranium Titration of Pure Compounds.

	Mass of Standard (4-place)	Purity of Standard	Repeatability of Standard Analysis	Mass of Sample (4-place)	Repeatability of Sample Analysis	Final Standard Uncertainty (%)
Contribution to Final Uncertainty (%)	3.0	1.5	18.2	16.6	60.7	0.024

Note: Buoyancy and U235 enrichment corrections assumed as insignificant contribution to the final uncertainty.

In this example, the most important uncertainty contributor is the repeatability of the sample analysis (typically 3-5 aliquots). However, supposing that a 3-place balance is used instead of 4, a new uncertainty budget shows up as in Table 2. One can notice a completely different distribution of uncertainty contributions and a significant increasing of the final relative uncertainty. It becomes clear that, in this example, the inappropriate selection of the balance may impose significant increment to the final uncertainty and the target uncertainty level of 0.05%rel. is not achieved anymore.

Table 2. Simulated Uncertainty Budget after Change in Balance.

	Mass of Standard (3-place)	Purity of Standard	Repeatability of Standard Analysis	Mass of Sample (3-place)	Repeatability of Sample Analysis	Final Standard Uncertainty (%)
Contribution to Final Uncertainty (%)	14.7	0.1	0.9	81.3	3.0	0.11

The estimation of the uncertainties associated with relevant input quantities, using standard statistical methods and carefully observing if correlations are present, is one of the basic recommendations of GUM. It also suggests the preparation of an uncertainty budget in order to clearly present the results and make it easier the evaluation process, the identification of significant influences and planning of improvements or optimizations.

4. CONCLUSIONS

A consistent evaluation of nuclear safeguards measurement results is possible only through consideration of the results along with the uncertainties. The steps involved in the calculation of the results and estimation of uncertainties must be “transparent” not only to the analyst responsible for producing the results but also to others with a need to work with those results (e.g., inspectors verifying the validity of the results, inter-laboratory measurement evaluation programs). The transparency is easily achieved by using standard methods and approved terminologies in expressing the results and associated uncertainties. It appears that uncertainties calculated using the GUM method will fulfill these requirements. Additional efforts must be made by safeguards laboratories to train their scientists and technicians in the GUM method for expressing nuclear measurement results and uncertainties. Laboratories and analysts must demonstrate the ability to produce measurement results with a comprehensive and correct estimation of uncertainties associated with those results. Inter-laboratory programs must be capable of providing evaluation outputs in compliance with GUM and advising laboratories and analysts in case of non-compliance. The changes incorporated into ITV´s-2010 reflect the GUM approach. We expect that, in the future, all nuclear accountancy laboratories and safeguards organizations work towards evaluating and reporting uncertainties in compliance with the guide.

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