FROM INTERNATIONAL EQUIVALENCE TO NATIONAL TRACEABILITY IN RADIONUCLIDE METROLOGY

Maria Sahagia, Anamaria Cristina Razdolescu, E.L.Grigorescu, A.Luca, C.Ivan

« Horia Hulubei » National Institute of R&D for Physics and Nuclear Engineering, POB MG-6, code 077125, Bucharest, Romania
*e-mail msahagia@ifin.nipne.ro

Abstract

The methods for absolute (direct) standardization, coincidence method for radionuclides emitting several simultaneous radiations and liquid scintillation counting for radionuclides emitting a single radiation, the obtained results, as the technical basis for a primary radioactivity standard recognition, are shortly reviewed. They were published in many scientific papers. The key and supplementary comparisons organized by the CIPM-CCRI(II), EUROMET, other supplementary IAEA comparisons, former COMECOM, as well as bilateral comparisons results, served as basis for the recognition of the international equivalence of the Romanian radioactivity standards. In order to assure the national traceability, our laboratory prepared and delivered radioactive standards, standardized radioactive sources, calibrated and checked radioactivity measurement equipment and organized national comparisons. The laboratory, attested by the BRML and notified by the CNCAN, is also on way to be accredited by the RENAR, as a calibration and testing laboratory, according to the RS EN ISO/IEC 17025:2005 standard requirements. Its QS was recognized by the Q-TC EUROMET

Key words

Equivalence, traceability, radioactivity

1 INTRODUCTION

The ionizing radiation metrology laboratory, including both branches, dosimetry and radionuclide metrology, of the Institute of Atomic Physics, was created during sixties, in the former section of applications of nuclear techniques based on the use of radioactive sources, as an immediate practical necessity to characterize the radioactive sources and to measure the irradiation doses, implied by the above mentioned applications. From the very beginning, it had a pioneering activity for development of measurement
equipment, elaboration of standardization methods, and mainly the validation, through the participation in international comparisons, of the Romanian newly developed standards. In the field of radionuclide metrology, the developed standardization systems and methods assured the absolute (direct) standardization, as basis for a primary radioactivity Romanian standard. In order to carry out standardization, adequate methods for preparation of radioactive sources were elaborated. The following steps in our activity were to assure their validation and recognition at the international scale. This aim was reached by the participation at international comparisons of standard sources and solutions. This experience started in 1962, when our laboratory participated at the International Commission for Radiation Units (ICRU) comparison for $^{60}$Co sources measurements; this responsibility passed in 1963 to the International Bureau for Weights and Measures, Bureau International des Poids et Mesures (BIPM). The laboratory participated constantly in such comparisons, registered as Key Comparisons in the Annex B, BIPM - Key Comparisons Data Base (KCDB). At the same time, we participated at International Reference System, Systeme International de Reference (SIR) comparisons, included also in the KCDB. Within the frame of the EUROMET system, a third series of key comparisons, included in the KCDB, is registered. At present, besides the classical key-comparisons, other supplementary comparisons, organized by the International Atomic Energy Agency (IAEA) are included also inside the KCDB. Our laboratory participated successfully at regional comparisons, organized by the former COMECOM and at bilateral or multilateral comparisons. All these comparison schemes are part of the recognition of the international equivalence of the primary Romanian radioactivity standard. The subsequent direction of the laboratory’s development was the assurance of the national traceability in radionuclide metrology. These tasks were accomplished by the following actions: (i) Development of the relative (indirect) standardization systems and methods for secondary calibration. (ii) Development of the methods for preparation of radioactive sources and solutions. (iii) Development of methods for the calibration and for the metrological check of equipment under the metrological control of the state. (iv) Organization of national comparisons. The implementation of a Quality System (QS) for Quality Assurance (QA), aimed both for the national, as well as for the international, Quality - Technical Committee (Q-TC) of EUROMET recognition, according to the EN ISO/IEC 17025:2005, and the final approval of the Calibration and Measurement Capability (CMC) documents. The paper presents some significant achievements of the laboratory, in the above mentioned activities, which assured the linkage between international equivalence and national traceability in radionuclide metrology.

2 ABSOLUTE STANDARDIZATION SYSTEMS AND METHODS

2.1 The $4\pi$PC-$\gamma$ coincidence method

A $4\pi$PC-$\gamma$ coincidence installation, containing a home made Proportional Counter (PC), flow type, and a NaI(Tl) detector, was set in the sixties. The electronics is NIM modular of types Nuclear Enterprises and Canberra. In the present configuration, it has been used for more than 30 years, with some replacements of modules. Recently, in the frame of the Excellence Research Program (CEEX), Module IV, 2006-2008, we deploy a contract having as subject the construction of a new coincidence installation, by using new modules and a semi-automatic operation system for command, acquisition and processing of measurement data. A photon-photon coincidence detection system,
provided with two thin NaI(Tl) detectors for the standardization of special radionuclides, is also operational. The original, internationally recognized contribution of the laboratory in the development of the $4\pi$PC-$\gamma$ coincidence method is published the papers [1-8].

2.2 The Liquid Scintillation - Triple to Double Coincidence Ratio method (LSC-TDCR)

A liquid scintillation counter, based on the principle of the Ratio between Triple and Double Coincidences (LSC-TDCR), realized with the contribution of the Laboratoire National Henri Becquerel (LNHB)-France and Radiochemical Centre (RC)-Poland, was put into operation in 2002; the attestation documentation was issued in 2004. The LSC-TDCR system and method were used for the measurement of nuclides for key and bilateral comparisons and for laboratory’s own necessities. The original contributions were published in the papers [9-11]. Adjacent to the standardization installations, a Mettler M5 micro analytical and analytical balances, traceable at the national mass standard of the National Institute of Metrology, are used for the quantitative preparation of sources for absolute standardization, as well as for other types of sealed sources.

3 INTERNATIONAL EQUIVALENCE

3.1 Key comparisons

These comparisons rely on the exclusive participation of the laboratories performing absolute standardization, as the primary objective is to avoid any mutual dependence of results. The Radionuclide Metrology Laboratory (RML) has participated in many such comparisons, as above mentioned, for a 45 year period. After the adoption if the CIPM-MRA, in 1999, with Romania as a signatory, the CIPM established the technical documents, as Appendices to the document. First step in our national official recognition was the valorisation of our long history of comparisons, by the Romanian Bureau of Legal Metrology (BRML). The “Horia Hulubei” National Institute of R&D for Physics and Nuclear Engineering, IFIN-HH, the official representative of the RML, became a designed laboratory, nominated on the Appendix A of CIPM-MRA. As a consequence of the recognized research and participation in comparisons, IFIN-HH became a member of the CIPM-Consultative Committee for Ionizing Radiations, Section II, Radionuclide Measurement, [CIPM-CCRI(II)] in 2004. The list of IFIN-HH, RML key comparisons, considered as basis for implementation of the MRA is registered in the Appendix B, Key Comparison Data Base (KCDB). Several types of such comparisons in a total number of 25 positions are present; they can be searched at the site: http://kcdb.bipm.org/AppendixB/KCDB_ApB_search_result.asp?sea... In all the cases, the degree of equivalence between participant laboratories, as well as between them and the Key Comparison Reference Value (KCRV), are presented.

3.1.1 Comparisons codified as: » CCRI(II)-K2. Radionuclide (Ex. Am-241)”

They refer to the participation at large scale comparisons, when a solution sent by the organizer is measured by the participants, and individual results, expressed as radioactive concentration of the solution, are reported to the BIPM where they are evaluated. They are under different stages of evaluation, such as presented in KCDB: (i) Approved for equivalence; (ii) Report in progress, Draft B; (iii) Report in progress,
Draft A; (iv) In progress. Our laboratory is registered with a total number of 15 radionuclides.

3.1.2 Comparisons codified as: “BIPM.RI(II)-K1.Radionuclide”

A system of two ionization chambers was installed at the BIPM in 1976, and served as basis for the creation of the SIR for gamma-ray emitting radionuclides. At present, the SIR is on way to be extended for alpha and beta particle emitters. These comparisons refer to radioactive solutions prepared and standardized, also exclusively by absolute methods, by laboratories themselves. They are sent to the BIPM, accompanied by the measurement document, where they are re-measured and the result is compared with the response equivalent of the other participants. The same types of equivalence degree are established and the same kinds of evaluation stages, as in the previous case, are presented on the KCDB. The process is a continuous one, each new participant being registered in the same position, starting with the first one. Our laboratory is registered with a total number of 6 positions.

3.1.3 Comparisons codified as: “EUROMET .RI(II)-K2. Radionuclide

These comparisons were deployed in the frame of EUROMET, Decay Data Evaluation Project (DDEP), and consist in the followings. A solution is sent to the participants, the radioactive concentration is determined absolutely, and evaluation of the results is similar as in the previous cases. The following step is the participation of the laboratory in projects regarding the determination of emission probabilities of various types of radiations in the radioactive decay.

3.2 Supplementary comparisons included in KCDB

These comparisons are organized by other bodies, such as the IAEA. They generally refer to measurements by relative methods, presented more in detail at point 4. We participated at a supplementary key comparison KCDB registered CCRI(II)-S6.I-131/2006 comparison [12], and we are planned for another supplementary, CCRI(II)-S5/2007 comparison regarding measurement of environmental samples.

3.3 Regional and bilateral comparisons

During the period 1975-1988, our laboratory participated successfully at regional comparisons, organized in the frame of the Commission for Atomic Energy of the former COMECOM. A total number of 8 comparisons were organised. What is worth to mention, is that for that period, the chosen radionuclides were difficult to standardize and some of them were only recently considered for CIPM key comparisons.

Another aspect regards the bilateral or multilateral comparisons, especially aimed to complete the validation process of the new liquid scintillation system and method. The radionuclides of interest were H-3, comparison with LNHB-France [11], and Ni-63, comparison with LNHB, RC-Poland, and CSIR- South Africa [13].

All the above presented comparison schemes are part of the recognition of the international equivalence of the primary Romanian radioactivity standard.
3.4 Elaboration of the Calibration and Measurement (CMC) documents

As a consequence of the internationally recognized equivalence, we elaborated, and transmitted to the EUROMET Ionizing Radiations - Technical Committee (IR-TC), a number of 21 CMCs, supported by comparisons, which are under analysis at the Joint Committee of the Regional Metrology Organizations and the BIPM (JCRB), and we expect to be approved, in connection with their coverage by the EUROMET Q-TC approved Quality System.

4 NATIONAL TRACEABILITY

Soon after its creation, our laboratory was implied in accomplishing all the duties connected with the assurance of the national traceability in radionuclide metrology. These requirements were responded by the following actions: (i) Development of the relative (indirect) standardization systems and methods for secondary calibration of radioactive sources and solutions and transfer of the radioactivity unit from the primary standard to these systems. (ii) Development of the methods for preparation of radioactive sources and solutions, adequate for practical applications; (iii) Development of methods for the calibration of radioactivity measurement equipment, as well as for the metrological check of the equipment under the metrological control of the state, belonging to various users; (iv) Organisation of national comparisons regarding the measurement of radioactive sources and solutions, known as proficiency tests.

The implementation of a Quality System (QS) for Quality Assurance (QA), aimed both for the national recognition, according to the EN ISO/IEC 17025:2005 Standard requirements, and for recognition of Institute’s QS by the Quality-Technical Committee (Q-TC) of EUROMET, had as a final result the attestation (BRML) and designation as notified calibration body (CNCAN). It is expected to get the final approval of the Calibration and Measurement Capability (CMC) documents and also to obtain the accreditation by the national body, RENAR.

4.1. Relative (indirect) standardization systems and methods

- A well-type ionization chamber, CENTRONIC IG12/20A, calibrated for gamma-ray emitters, is used for the measurement of vials containing radioactive solutions and solid sources. Recently, the system was upgraded by a high performance Electrometer. The system calibration was accomplished both by transfer of radioactivity unit from the primary standard, coincidence installation, verified in international comparisons, and by direct comparison with the Physikalisch-Technische Bundesanstalt (PTB), Germany [14].
- A large area multiwire, sealed proportional counter for surface alpha and beta sources calibration, transfers the radioactivity unit from the primary standards to the sources, and measures directly their alpha and beta particle emission rates [15].
- Two gamma spectrometry systems with HPGe and GeLi detectors, and adequate software’s, allow for carrying out relative standardization and spectrometric analyses. The new spectrometric system, based on a high efficiency HPGe detector, provided with adequate shielding and software for operation and for processing of data, also part of the above mentioned CEEX-contract, will allow further development of advanced studies, such as published already [17,18].
4.2 Preparation of radioactivity standards for practical applications

The calibration and metrological check of the equipment for measurement of the radioactivity in various fields of applications, imposed the development of technics for the preparation of a large variety of standard sources and solutions, to be used in house, or delivered to the external customers, performing measurement.

4.2.1 Preparation of radioactive solutions

A large variety of radioactive solutions, physico-chemically stable, adequate as radioactivity standards, were prepared [19, 20]. The majority of them, even for external delivery, are standardized absolutely, by the methods presented at point 2. After the transmission of radioactivity unit to the ionization chamber (see 4.1) the precise relative measurement is now possible for gamma-ray emitting radionuclides.

4.2.2 Preparation of point and large area beta sources

These sources are of immediate interest, both for calibrations, as well as for the effective measurement of the so called “Beta global” radioactive content of environmental, industrial and food chain samples. Their preparation and measurement of the particle emission rate in $2\pi$sr geometry are described in [15].

4.2.3 Preparation of point and volume gamma solid standard sources

Sets of point gamma–ray emitting sources, to be used for the calibration of $\gamma$-ray spectrometers, were prepared both from standard solutions and from concentrated solutions and then relatively standardized. Generally a number of 9-10 radionuclides were used for preparation of spectrometry sets [21]. The extensive national requirement of volume activity standards, necessary for the measurement of environmental, industrial, food chain volume samples, by the $\gamma$-ray spectrometry method, with matrices as similar as possible with the samples to be measured, resulted in realization of solid volume standards with matrices: water equivalent, soil, zeolite, [22, 23]

4.3 Methods for calibration and metrological check of measurement equipment

In this field, the laboratory was requested to calibrate complex equipment used at various IFIN-HH or external units. Some of them are the Tritium monitors, under operation at the Nuclear Power Plant (NPP) Cernavoda, for which an original calibration method was elaborated [24]. The Radioisotope Calibrators are the most used equipment for radioactivity measurement in Radioisotope Production Departments, Radiopharmaceutical Control Laboratories and Nuclear Medicine Units; they are under the metrological control of the state. The quality requirements in measurement and obtained results of calibration are presented in [25, 26]

4.4 Organisation of national comparisons

The most significant test, recognized as relevant in the evaluation of the capability of a laboratory, is the participation in proficiency tests, organized by a recognized metrology laboratory. This subject, regulated by the ISO Guides 43-1 and 43-2, is very debated, especially in connection with the significance of results, standards for acceptance, and so on. The new ISO 13528:2005” Statistical methods for use in proficiency testing by inter-laboratory comparisons” was not yet implemented. We
organized, in the past, four such national comparisons, first of all for a scientific output. However, the results, without any legal interpretations were communicated to the control bodies, CNCAN and Authority for Public Health, Bucharest. Two comparisons referred to measurement of radioactivity in environment [27,28] and two regarded measurement of solutions in nuclear medicine units [29,30]. The criteria for evaluation of results relied on the ratios between measured and conventionally true activities, R, and evaluation of the capability of measurement units of being situated within the limits 0.9 <R<1.0, rather than the uncertainties reported by participants, as these legal limits are critical, mainly in nuclear medicine measurements. On the other side, the comparisons were intended to reveal, at the national level, the necessity of applying corrections on the results.

At present time, a national comparison, under the supervision of the IAEA, regarding the measurement of I-131 in nuclear medicine units is on way.

4.5 Implementation of the Quality System

The implementation of a Quality System (QS) for Quality Assurance (QA), aimed both for the national recognition, according to the EN ISO/IEC 17025:2005 Standard requirements, and for recognition of Institute’s QS by the Quality- Technical Committee (Q-TC) of EUROMET.

The effort was focussed on several directions [31]: (i) The elaboration of the RML-Quality Documents, in agreement with the IFIN-HH general valid ones; the list contains the Quality Manual, Organization, System and Work Procedures and Instructions (ii) Primary implementation of the QS within the RML; (iii) Two internal audits, of the IFIN-HH audit team, were done. An external audit “ Pre-assessment evaluation of the accomplishment of the EN ISO/IEC 17025-2005 requirements in the laboratory” was made by Michael J Woods, from the Ionizing Radiations Metrology Consultants (IRMC) Ltd., UK and a detailed report was formulated (iv) Corrective actions, regarding the revision of documents and QS implementation. (v) Application at the national accreditation body RENAR. On the other side the RML obtained already the attestation and designation as notified calibration body from BRML and CNCAN. As a consequence of the EUROMET Q-TC body acceptance of the IFIN-HH QS, the final approval of the Calibration and Measurement Capability (CMC) documents and their inclusion in the CIPM-MRA, Appendix C, is expected.

5 CONCLUSIONS

- The Radionuclide Metrology Laboratory of IFIN-HH, as a primary radioactivity standard laboratory, fully demonstrated its equivalence on the international level, by its presence in the CIPM (BIPM)-KCDB and other international data bases.
- The laboratory assured the national traceability in radioactivity measurements, by: preparation and delivery of radioactive standards, calibration and metrological check of measurement equipment, organization on national comparisons.
- The implemented Quality System checked both at the national and international levels, is a premise for obtaining of the national RENAR accreditation.
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