ORIGINAL PAPER

ROLE OF THE RADIONUCLIDE METROLOGY IN QUALITY ASSURANCE OF THE ENVIRONMENTAL PHYSICS MEASUREMENTS

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Abstract. *This paper presents some aspects of the technical support offered by the Radionuclide Metrology laboratory from IFIN-HH in the field of environmental physics measurements.*

-The development of the basis of installations and methods for absolute standardization of radionuclides;

-The assurance of the equivalence between our primary activity standard and other national standards;

-The continuity of the traceability chain, assured by the development of secondary activity standards.

Keywords: *primary activity standard, international equivalence, secondary standards, national traceability.*

1. INTRODUCTION

The environment physics is tightly connected with precise radioactivity measurements, both directly and indirectly. The direct measurements refer to the control of the radioactive content of various environmental samples. The indirect measurements are connected with some nuclear methods used in the elemental analysis of samples, such as the instrumental neutron activation analysis (INAA), or use of the radioactive tracers for the characterization of environment, such as the measurement of ^{137}Cs migration. The laboratories involved in such measurements develop a large variety of measurement equipment and methods used in research, or in reporting these results to the national authorities and to the international organisations. The common feature of all these measuremens and reports is the necessity to assure their reliability via a continuus metrological traceability chain, up to the higher, primary standard level of activity [1].

The Radionuclide Metrology Laboratoy (RML) from IFIN-HH, the owner of the primary Romanian standard of activity, assures the entire technical support to the Romanian laboratories involved in the measurement of activity and cooperates successfully with all the world primary laboratories. The duties of RML are defined in the following terms: definition of the traceability chain and implementation of the quality management system.

- Traceability levels, according to the vocabulary of international metrology (VIM) [2]:

Level 1- Primary standard and international equivalence

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Realisation of the Activity primary standard by the development of methods and installations for standardization, participation at international comparisons, implementation of a quality system.

Levels 2 and 3 – National traceability

Level 2 consists in the realisation of the secondary standard system, and level 3 basically assures the national dissemination of the activity standards.

- The system for quality management and obtaining of the national accreditations for the RML.

2. THE INTERNATIONAL RML EQUIVALENCE PRESENTATION

2.1. REALISATION OF THE PRIMARY STANDARD

Basically, two methods are used: methods based on the 4πsr detection geometry for radionuclides emitting a single type of radiations, and coincidence methods for the radionuclides emitting coincident radiations.

The new 4πPC-γ coincidence methods and installations of RML.

The updated system contains: a proportional counter and two NaI(Tl) detectors, new electronical modules type ORTEC, or built in IFIN-HH, allowing for the semiautomatical operation.

An X,*γ* -X,*γ* coincidence system provided with two thinNaI(Tl) for standardization of ^{125}I [3] was built and validated. And original method for standardization of ^{99m}Tc was developed [4].

The liquid scintillation - triple to double coincidence ratio (LSC-TDCR).

The installation was realized with the support of LNHB-France and was recently validated in international comparisons [5]. A new detection block, consisting from a new optical chmber and 6 Channel Photomultipliers (CPM) was constructed [6, 7].

The primary Romanian radon standard.

It is the result of a collaboration with ICSI, Rm. Valcea and ISP, Timisoara, within the PNCDI II, contract 71-102. The system assures the traceability of ^{222}Rn measurements for the entire chain, from the primary standardization by LSC, up to the distribution of working certified standards [8, 9].

2.2 RECOGNITION OF THE INTERNATIONAL EQUIVALENCE OF THE PRIMARY STANDARD

2.2.1 The International System

The Authority of the World Metrology is the General Conference on Weights and Measures - GCWM (CGPM) having under authority the International Committee for Weights and Measures-CIPM. The Bureau International des Poids et Mesures-BIPM, Sèvres, France, has the custody of the international standards.CIPM coordinates the metrology branches through the Consultative Committees, such as the Comité Consultatif des Rayonnements Ionisants – CCRI. CCRI is divided in three sections. The Section II, CCRI(II) - Measurement of Radionuclides, coordinates the Radionuclide Metrology. The equivalence of the primary standards is assured at this level only by absolute methods of standardization. At the regional level, the Regional Metrology Organizations -MRO are operational. In Europe there exists the EURAMET: European Association of National Metrology Institutes and COOMET: Euro-Asian Cooperation of National Metrology Institutes. The connection between BIPM and

MROs is assured by the Joint Committee of the Regional Metrology Organizations and the BIPM - JCRB. At this level one assures the traceability also by using secondary (relative) standardization methods. The CIPM-MRA, Mutual Recognition Arrangement [10] defines the recognition of the calibration and measurement certificates issued by the National Metrology Institutes. Romania is part of CIPM-MRA, and the signatory is BRML-INM. The document contains four annexes: Annex A - List of signatories; Annex B - Key Comparison Data Base (KCDB) per Institute; Annex C- Calibration and Measurement Capabilities (CMC); Annex D- Key Comparison Data Base, summarizing all the key comparisons. The condition for the applicability of MRA by the signatory countries is the demonstration of the equivalence of the primary standards, or of the traceability for the others.

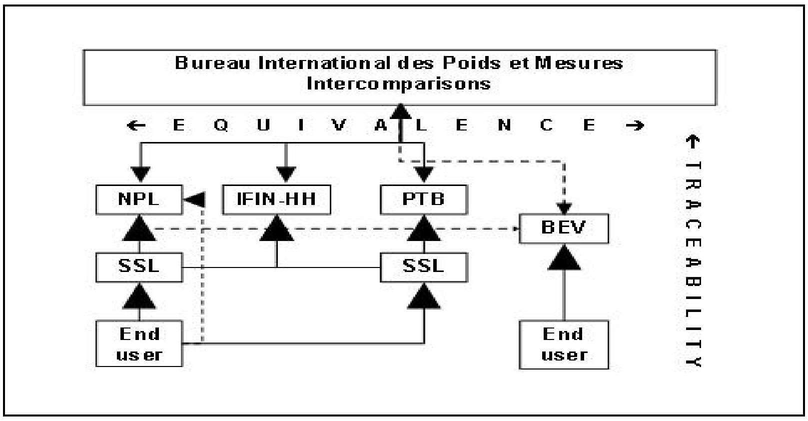

Fig. 1. Equivalence – traceability general scheme [11].

The equivalence is demonstrated only by the participation at international comparisons. IFIN-HH, RML, has participated at international comparisons since 1962.

The Key Comparison Data Base (KCDB) is included *in Annex B of the CIPM-MRA:* http://kcdb.bipm.org/AppendixB/KCDB_ApB_search.asp .

The Protocol 2351/18.03.2005, between BRML-INM and IFIN-HH establishes that IFIN-HH is designed by the BRML, as the owner of the primary activity (Becquerel) and derived quantities standard, with national and international responsibilities in the field.

- Institute designed as responsible for participation in the CIPM-MRA, included in *Annex A of the CIPM-MRA, List of signatories.*

- Institute responsible for EURAMET Technical Committee for Ionizing Radiations, TC-IR

Due to its scientific authority, IFIN-HH became:

-Member of the International Committee for Radionuclide Metrology (ICRM) http://www.physics.nist.gov/icrm

-Member of the Consultative Committee [CCRI(II)-CIPM],
http://www.bipm.org/en/committees/cc/ccri/CCRIsection2/members_ccrisection.html

2.2.2 The key Comparisons of the CIPM-BIPM

Definition of the Key Comparisons: Selected comparisons taken formally into account for the establishment of equivalence. CCRI(II) may approve supplementary comparisons, organized by other organizations, to be used for equivalence such as the EURAMET and COOMET. Relative methods may be used at this level, in order to extend the applicability of the CIPM-MRA, Annex C. *Comparisons: » CCRI(II)-K2. Radionuclide (Ex. Am-241)"*

These comparisons are organized at the worldwide scale. A single batch of radioactive solution is used for the preparation of all the samples sent to the laboratories for standardization. The laboratories standardize it and send to the BIPM the filled-in Reporting Form. The reports are evaluated at BIPM and the Draft reports are drawn up. The B Draft is submitted to the approval of the CCRI(II) and published. RML is registered with such comparisons in the KCDB with the radionuclides: ^{241}Am, ^{109}Cd, ^{139}Ce, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{55}Fe, ^{3}H, ^{125}I (2), ^{192}Ir, ^{177}Lu, ^{54}Mn, ^{32}P, ^{75}Se, ^{89}Sr, ^{204}Tl, ^{65}Zn.

- *Comparisons: "BIPM.RI (II)-K1.Radionuclide"*

BIPM disposes of two "Well type" ionization chambers, CENTRONIC IG11/20A, as the basis for creation of the International Reference System - SIR, for gamma-ray emitters [12]. The key comparison is carried out by the individual preparation and certification of an ampoule with standard solution, which is then sent to the BIPM with the Comparison filled-in Form. It is remeasured and the two results are compared. RML is registered with the radionuclides: ^{110m}Ag; ^{133}Ba; ^{60}Co (1983 and 2007); ^{134}Cs; ^{131}I; ^{57}Co; ^{137}Cs

- *Supplementary Comparisons*

Type: "EURAMET.RI(II)-S5. Radionuclide

These comparisons are deployed as Decay Data Evaluation Projects-DDEP. The first step of comparisons is similar to the CCRI(II)-K2. The second step consists in the measurement of the emission intensities for X and gamma-rays. RML is registered with: ^{169}Yb; ^{65}Zn; ^{124}Sb.

IAEA-CCRI(II).Matrix content. Radionuclides

These projects are organized within the frame of Coordinated Research Projects-CRPs and are approved by the CCRI(II); they refer to special types of samples and relative methods are accepted. The RML is registered as CCRI(II)-S6.Radionuclide for: ^{57}Co and ^{131}I, in nuclear medicine and as IAEA-CU-2007-06-CCRI(II) S5 – Determinaton of gamma emitting radionuclides TENORM in phosphogypsum.

2.2.3 Establishment of the SI Activity Unit (Becquerel) by key comparisons

The Activity Unit is established individually for each radionuclide, after the evaluation of the key comparison result which is expressed as the Key Comparison Reference Value – KCRV.

Four methods are used to calculate the KCRV: arithmetic and weighted arithmetic mean, median and weighted median. The most adequate variant is selected; in most cases, the arithmetic mean is preferred. The outliers are established after applying the exclusion criteria. The KCRV and its uncertainty are approved by the CCRI(II) and may be modified in time. The "outliers" are not eliminated from KCDB, but their difference from KCRV must be taken into account when reporting uncertainties.

Whenthe *SIR -BIPM.RI(II)-K1.Radionuclide* and the *CCRI(II)-K2. Radionuclide* results are registered for gamma-ray emitters, a link is established between the two types of comparisons, by the measurement in the BIPM ionization chamber of the ampoules sent for the K2 comparison.

Equivalence

An equivalence matrix is calculated. Two main quantities are reported and compared: Difference between the individual result and KCRV, (D_i) and its uncertainty (U_i), for k=2, [13]. The ratio of these quantities is the measure of degree of equivalence. The CCRI(II) approved "Draft B" is published in the KCDB and in *Metrologia, Technical Supplement*. IFIN-HH, RML, is registered in KCDB with 28 radionuclides.

Analysis of the ^{192}Ir key comparisons [14]

CCRI(II)-K2.Ir-192 was organized in 2002, with 17 participants, while BIPM.RI(II)-K1 comparisons include 10 participants. The KCRV was: a = (203.76±0.63) kBq g^{-1}, and the IFIN-HH result: a_{IFIN} = (204.81 ±0.33) kBq g^{-1} . SIR - KCRV is: A_{eSIR}= 19028(37) kBq, and the equivalent value of the K2 comparison: A_{eK2} = 19100(59) kBq. The combined KCRV = 19067(24) kBq and IFIN-HH equivalent: A_{eIFIN} = 19198(34). Consequently, it is reported that the final KCRV is based on the results of the following participants: K1-CMI-IIR, LNMR, OMH, IRA; K2-BARC, BIPM, BNM-LNHB, CIEMAT, CNEA, CSIR-NMI, IFIN-HH, IRMM, KRISS, NMIJ, NPL, PTB.

IFIN-HH equivalence matrix: D_i =0.13 MBq ; U_i = 0.07 MBq, k=2

Equivalence validity

A 20 years validity interval was adopted by consensus. Consequently, IFIN-HH, participated in 1983 and in 2007 at K1.Co-60 comparisons. As for ^{134}Cs, the participations were: K2 in 1978 and K1 in 2006. For ^{137}Cs, the participations were: K2 in 1982 and K1 in 2009.

2.2.4 SI Activity Unit (Becquerel) for other radionuclides

A matrix of radionuclides, codified by colours, according to the standardization difficulty by method, was established by the CIPM-CCRI(II) Key Comparison Working Group (KCWG), within the document Grouping Criteria Radionuclides for Supporting CMCs. It can be used as follows. A red coded radionuclide measured by LSC, such as ^{3}H, supports the radionuclides ^{89}Sr, ^{90}(Sr+Y), LSC measured, and green coded

2.3 IMPLEMENTAION AND INTERNATIONAL RECOGNITION OF THE QUALITY MANAGEMENT SYSTEM. APPROVAL OF CMC FILES.

An international agreement for the practical use of the results obtained by the National Metrology Institutes in assurance of the traceability chain and recognition of measurements and certificates was concluded through the document: "Joint statement by the CIPM and ILAC on the roles and responsibilities of national metrology institutes and national recognized accreditation bodies". The International Laboratory Accreditation Cooperation - ILAC has a regional organization scheme. European Cooperation for Accreditation – EAL is acting in Europe.

At the level of EURAMET, the implementation of the Quality Management System (QMS) is monitored by the EURAMET, Technical Committee – Quality, TC-Q.

The national counterparts of EAL are the national accreditation bodies, in our case RENAR.

At the level of CIPM - MRA, the statement is practically applied by the use of the Annex C of the CIPM MRA, Calibration and Measurement Capability files – CMCs. The approval and publication of CMCs for a NMI is the result of two types of evaluation components for the international recognition:

(i) Approval of the international equivalence for primary and of the traceability for secondary standards

(ii) Implementation of the QMS and recognition by the MRO's TC-Q.

2.3.1 Approval of equivalence and traceability.

The NMI, member of one MRO, draws up the primary or secondary CMC files. The statement is confronted with the KCDB for primary, or in our case EURAMET, for secondary standards. The reviewers verify the conformity of declared CMCs and approves them within the EURAMET, TC-IR. Afterwards, the CMCs drafts are sent for notification to all the world MROs (COOMET, Asia-Pacific, America, Africa, etc). After the peer review process, they are submitted for the approval of the JCRB.

2.3.2 Implementation and approval of the QMS.

The EN/ISO/CEI 17205:2005, referential “General requirements for the competence of testing and calibration laboratories” is applied. A complete documentation and its implementation are presented at the annual meetings of the EURAMET, TC-Q. The technical experts monitor the documents and after the first approval, the QMS is annually reconfirmed. This is a perequisite condition to maintain the CMCs in the published in Annex C of MRA.

2.3.3 IFIN-HH, RML situation.

34 CMC files, Radioactivity standards, passed the peer review process; the QMS was approved in 2007. All our CMCs were approved by the JCBR in April 2008 and published in CIPM-MRA, Annex C. http://kcdb.bipm.org/AppendixC/default.asp The consequences for RML:

(i) RML got the agreement of the BIPM Director to use the BIPM-logo in its Calibration Certificates

(ii) The RML Calibration Certificates are recognized everywhere throughout the world.

3. REALISATION OF THE ACTIVITY SECONDARY STANDARDS

They are transfer instruments of the activity unit from the primary to the lower order, working standards, for the end users, as standard (reference) products or calibration services. They consist from installations for the secondary (relative) standardization; their calibration is done with sources and solutions absolutely standardized with the primary standards.

3.1 SECONDARY STANDARD FOR HIGH ACTIVITY SOLUTIONS

The RML well type ionization chamber CENTRONIC IG12/20A was described in [15]. Recently the electrometric system was replaced with an electrometer Keithley E6517A and the calibration factor is expressed as pA/MBq. The validations were done by comparison with PTB-Braunschweig-Germany and with the RML results obtained in international comparisons, respectively the value of the KCRV [16]. The calibration factors are determined for the radionuclides: ^{241}Am, ^{57}Co, ^{99m}Tc, ^{186}Re, ^{188}Re, ^{153}Sm, ^{177}Lu, ^{75}Se, ^{169}Yb, ^{131}I, ^{133}Ba, ^{51}Cr, ^{192}Ir, ^{134}Cs, ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co, ^{152}Eu and three geometries:

(i) Solid sources;

(ii) ^{131}I capsules;

(iii) Solutions 2 mL, 3.6 mL, 5 mL.

3.2 LARGE AREA PROPORTIONAL COUNTER FOR THE CALIBRATION OF THE ALPHA AND BETA SOURCES IN 2ΠSR PARTICLE EMISSION

The counter is a multiwire, de 250 x 250 mm, proportional one. It was calibrated in terms of (number of impulses)/(number of emitted particles in 2πsr), by using sources standardized in the windowless 4πsr counter of the coincidence system. The calibration factors are established for: alpha sources - ^{241}Am; beta sources - ^{14}C, ^{147}Pm, ^{204}Tl, ^{90}(Sr-Y), maximum dimensions 250 x 250 mm.

3.3 NaI(Tl) DETECTOR FOR THE CALIBRATION OF GAMMA-RAY SOURCES

The NaI(Tl) chain of the coincidence installation is used for relative measurements of various types of gamma-ray emitter sources. The standardization is done for each type of source separately.

3.4 HIGH RESOLUTION SPECTROMETRIC SYSTEMS

RML disposes of a system containing: Alpha particle chain – vacuum chamber with Si-surface barrier. X and gamma rays chain - Si(Li) detector. High resolution GeHP detector: Relative efficiency 29%; Energy interval 35 keV – 3 MeV; Resolution: 0.85 keV (122 keV) and 1.74 keV (1.332 MeV). An efficient shield, consisting from: 10 cm old lead; 1 mm tin; 2 mm electrolytical cooper, assures an integral background rate on the whole energy interval: 1.4 s^{-1}.

4. THE THIRD TRACEABILITY LEVEL

The continuity of traceability chain is assured by the development of radioactive standards (reference materials), equipment calibration methods and organization of national comparisons in the field of environmental radioactivity measurements. This level of traceability for the environmental physics measurements will be presented in future paper.

5. CONCLUSIONS

- The Radionuclide Metrology Laboratory assures the representation of IFIN-HH, designed National Metrology Institute.
- The RML primary activity (Becquerel) standards are internationally equivalent.
- The RML transmits the activity unit on the whole national traceability chain.

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