BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
06-02-A (2018) 01-14

Radionuclide metrology: traceability and response to a radiological accident

da Cruz[a] P. A. L., Tauhata[a] L., da Silva[a] C. J., Di Prinzio[a] M. A., Delgado[a] J. U., Oliveira[a] A. E., Oliveira[a] E. M., Poledna[a] R., Loureiro[a] J. S., Ferreira Filho[a] A. L., da Silva[a] R. L., Filho[a] O. L. T., Veras[a] E. V., Rangel[a] J. A., Gomes[a] R.S., Dantas[a] V. B., Quadros[a] A. L. L., Almeida[a] M. C. M., de Souza[a] P. S., Ruzzarin[a] A., Conceição[a] D. A., Araújo[a] M. T. F., dos Santos[a] A. R. L., Iwahara[a] A.

[a] Instituto de Radioproteção e Dosimetria (IRD)/Comissão Nacional de Energia Nuclear (CNEN)
Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI)
palcruz@ird.gov.br

ABSTRACT

In case of radiological accident there are characteristic phases: discovery and initial assistance with first aid; the triage and monitoring of the affected population; release of affected people; forward the victims to medical care; as well as the preparation of report on the accident. In addition, studies and associated researches performed in the later period. Monitors, dosimeters and measurement systems should be calibrated by contaminating radionuclide standards. The radioactive sources used must be of metrological reliable. In Brazil, this is performed by LNMRI/IRD/CNEN, designated by INMETRO, which Radionuclide Metrology Laboratory is responsible by standardization and supplying of radioactive sources in different geometries and matrices. This laboratory has radionuclide solutions stock with controlled environmental variables for the sources preparation, which are calibrated and standardized by primary and secondary methods. It is also responsible by dissemination of standards and, in order to establish metrological traceability of national standards, participates in international key-comparisons promoted by BIPM and regional metrology organizations. Internally, it promotes the National Comparison Programs for laboratories that analyze environmental samples and the traceability for radiopharmaceuticals producing centers and nuclear medicine services in

Acepted: 2018-06-11

the country. The paper presents significant results for the main radionuclides standardized by the Radionuclide Metrology Laboratory by international key-comparisons and national comparisons to provide metrological traceability. With the obtained results, the LNMRI integrates the international metrology BIPM network and fulfills its function of supplying, with about a hundred of radioactive standards, the country's needs in different applications, such as environmental analysis, radiopharmaceutical and attendance to radiological accident.

Keywords: radionuclide metrology, radioactive standard sources, metrological traceability, response to a radiological accident; ^{137}Cs, Goiania.

---

1. INTRODUCTION

Radionuclide metrology is the area of the scientific knowledge that studies the radionuclides in their respective decay-schemes, aiming the elaboration of methods and techniques capable of performing measurements of their radioactivity with the greatest possible precision and accuracy degree. Thus, metrology brings together all the theoretical and practical points of view of the measurements, regardless the uncertainty measurement and the application field, guaranteeing reliability.

The main physical quantity of interest linked to radioactivity is the activity or decay rate. According to National Council on Radiation Protection [1], the activity of a radioactive source containing a radionuclide, in a given specific energy state and moment, is the expected value, at that moment, of the spontaneous nuclear transitions number, in time unit, at that energy state.

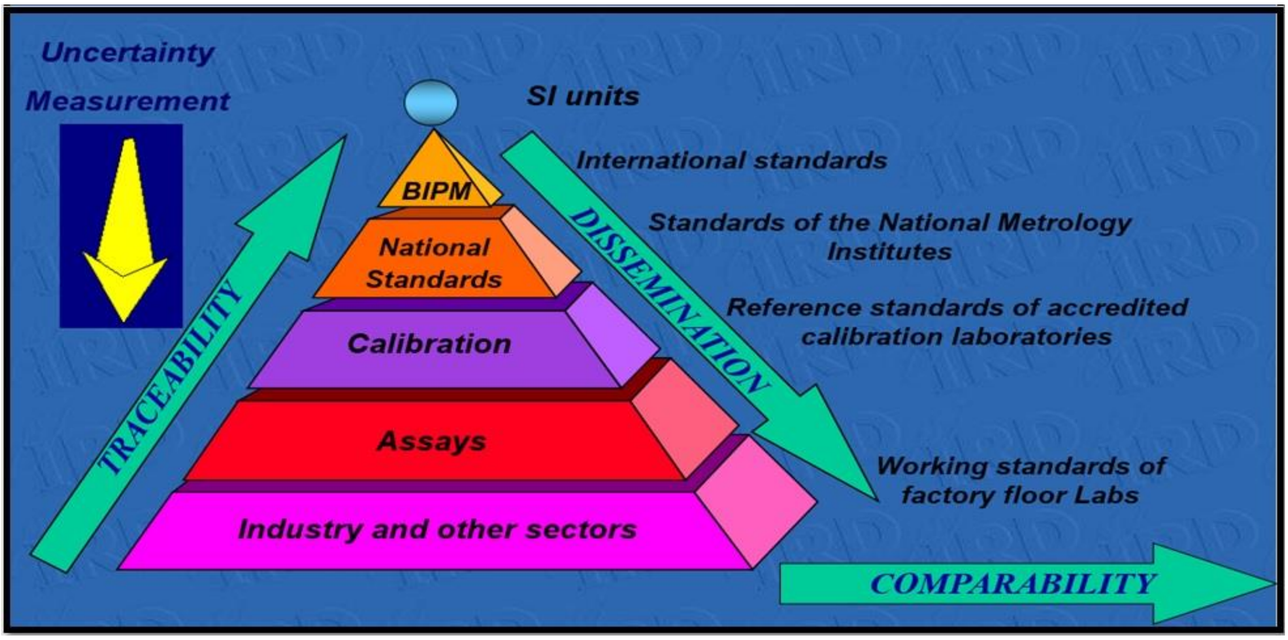
To standardize a radionuclide by a primary method means to measure directly all nuclear transitions occurring per unit of time. A complex task, since it involves several parameters such as decay, type of radiation, probabilities and differentiated energy lines. Thus, each radionuclide needs of different measurement techniques and approaches. The metrological culture suggests that the greater the number of standardization methods in a laboratory, better is the result.

As responsible by dissemination and custody of radioactive standards, the LNMRI must have the greatest number of standardization techniques and methodologies, and thus, ensuring the robustness of the results.

The LNMRI is the national metrology laboratory for ionizing radiation, designated by National Metrology Institute of Quality and Technology (INMETRO) since 1989, and it is responsible by dissemination and custody of radioactive standards. In this sense, the laboratory must have the greatest number of standardization techniques and methodologies, and thus, ensuring the robustness of the results.

It should be noted that for a device to be considered as an appropriate instrumentation it is necessary that, in addition to being suitable for the measurement, it should present in its measurement sequences some characteristics, such as: repeatability, defined by the agreement degree of the results obtained under the same measurement conditions; reproducibility, defined by the agreement degree of the results obtained under different measurement conditions; stability, defined as the instrument's ability to keep its measurement characteristics constant over time; accuracy, related to the agreement degree of the results with the "true value" or "reference value" to be determined; precision, related to the agreement degree of the results between them, usually expressed by the standard deviation in relation to the mean; sensitivity, which is the ratio between the response of an instrument and the corresponding stimulus variation, and; efficiency, which concerns the ability to convert the received stimuli into measurement signals [2].

Figure 1: *Metrological traceability: hierarchy of metrological system.*



The international metrology vocabulary [3] conceptualizes metrological traceability as the property of a result of measurement or value of a standard to be related to the established references, generally to national or international standards, through a continuous comparison chain, all having established uncertainties. Therefore, a continuous comparison chain is called the traceability chain.

The figure 1 illustrates the international hierarchy of the chain metrological to get the traceability, in which the Bureau International des Poids et Mesures (BIPM) is at the top of the international metrological chain, so that the measurement capacity of each National Metrology Institute (NMI) is compiled in a database maintained by BIPM, based on performance obtained in the key-comparisons.

2. MATERIALS AND METHODS

The traceability of the measurements for different radionuclides in primary systems in the LNMRI is obtained by participation in BIPM key-comparisons of the international metrological chain, European Metrology region (EURAMET) and the Asian Pacific Metrology Program (AMPM).

The LNMRI has primary methods $4\pi\beta$(PC)-γ(NaI) coincidence based on gas flow proportional counter [4]; $4\pi\beta$(LS)-γ(NaI) anticoincidence [5], CIEMAT/NIST and TDCR based on liquid scintillation [6]; peak-sum coincidence based on gamma-spectrometry [7]. In addition, secondary systems measurements: two ionization chambers, IG11 and IG12 [8], to keep the memory activity values obtained in the primary standardization from key-comparisons promoted by Bureau International des Poids et Mesures (BIPM) of the international metrological chain. It also provides methods for calibration based on sodium iodide and germanium for gamma spectrometry [9] and radionuclide calibrators for radiopharmaceuticals [10].

LNMRI has a large stock of radionuclide standards to supply national and international users. The radioactive sources in any geometry are prepared by differential weighing in balance of six decimal scales, therefore with a high precision degree and with the guarantee of traceability.

In order to establish the traceability and quality control of the activity measurement in the Brazil, LNMRI has been promoted national comparison programs with participants from environmental analysis laboratories and for the nuclear medicine. LNMRI also has implemented a management quality control based on NBR ISO/IEC 17025 to attend the requirements of national and international metrological chain.

3. RESULTS AND DISCUSSION

In order to guarantee the traceability of its measurements, the LNMRI has been participated in key- comparisons promoted by the BIPM, as showed in table 1 and 2.

The LNMRI was successful in these comparisons, with its measurements within of the established limits obtained by the average value of the results obtained by participating laboratories, and only in the of ^{59}Fe comparison the LNMRI result was outlier. Based on their key-comparisons and in maintaining the quality management system, the LNMRI already has 97 Capable Measurement Calibration (CMCs) lines registered at Key-Comparison Data Base (KCDB) of the BIPM, which demonstrates their capability to measure the activity quantity.

In the KCDB, a CMC is described by the measured quantity and its range, and is characterized by an uncertainty generally given at a 95 % level of confidence, together with the method or instrument used, the values of the influence parameters if any, and any other relevant information.

It means that LNMRI has a high metrological performance because for a national metrology laboratory publishing CMCs in KCDB, it should stablishing its traceability route by a primary realization, in which case traceability must be declared to its own demonstrable realization of the International System (SI). The figure 2 illustrates LNMRI performance in the BIPM key-comparison for ^{99m}Tc, the main radionuclide used in nuclear medicine.

The national intercomparison program (PNI) was implemented by LNMRI with the participation of regional environmental laboratories in Brazil [11]. The comparisons for environmental of spiked samples in matrices water, soil, air filter, vegetation and milk were computed from 1991 to 2016 and the number of participants increased from 11 to 29 per year.

A total of 32 radionuclides were analyzed, which 16 belonging to the natural U and Th series, plus ^{3}H and ^{40}K. At the beginning of the PNI, 11 laboratories analyzed ^{137}Cs sample and in 2016 this number increase to 15 one, to verify the performance of this radionuclide in this period.

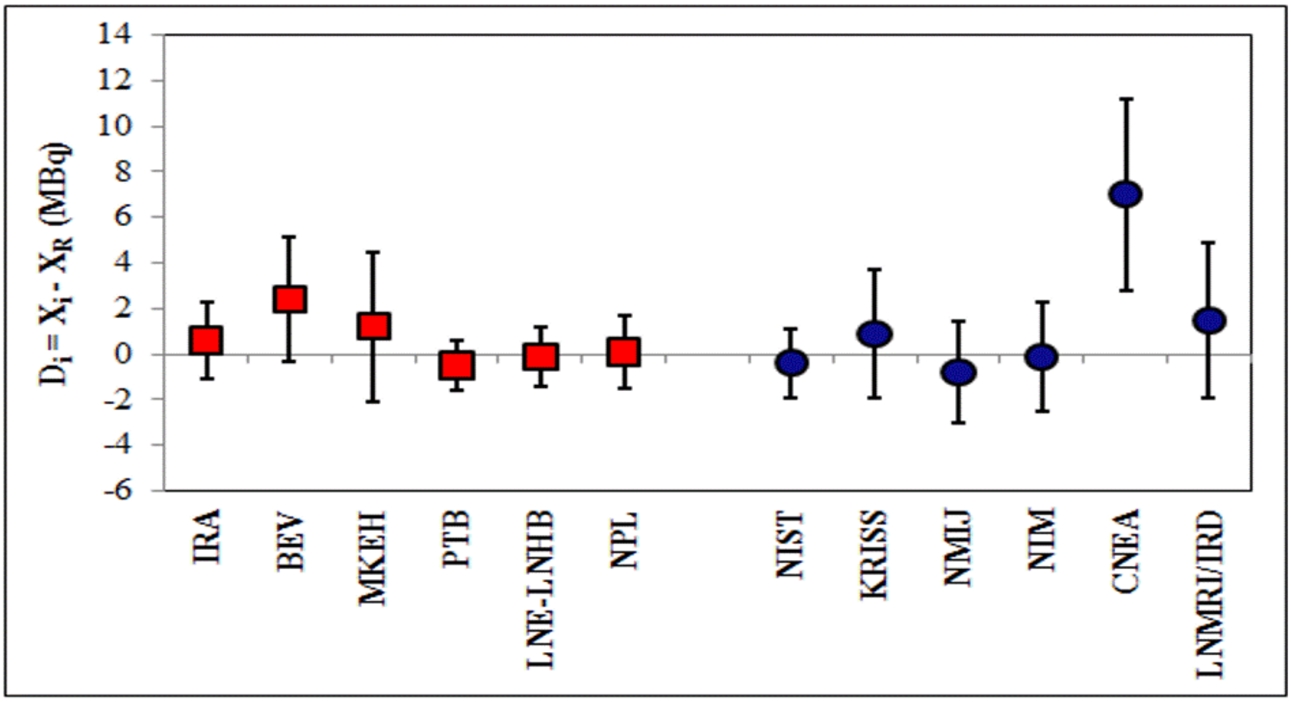
Table 1: Traceability of LNMRI measurements for radionuclide activity in BIPM key-comparisons.

Radionuclíde	Date	BIPM key-comparison
^{109}Cd	1986	CCRI(II)-K2.Cd-109
^{125}I	1988	CCRI(II)-K2.I-125
^{75}Se	1992	CCRI(II)-K2.Se-75
^{169}Yb	1997	EUROMET.RI(II)-K2.Yb-169
^{152}Eu	1999	CCRI(II)-K2.Eu-152
^{166m}Ho	1999	APMP.CCRI(II)-K2.Ho-166m
^{88}Y	2000	APMP.CCRI(II)-K2.Y-88
^{58}Co	2000	APMP.CCRI(II)-K2.Co-58
^{89}Sr	2000	CCRI(II)-K2.Sr-89
^{192}Ir	2002	CCRI(II)-K2.Ir-192
^{65}Zn	2002	CCRI(II)-K2.Zn-65
^{32}P	2002	CCRI(II)-K2.P-32
^{204}Tl	2002	CCRI(II)-K2.Tl-204
^{125}I	2004	CCRI(II)-K2.I-125
^{241}Am	2006	CCRI.RI(II)-K2.Am-241
^{55}Fe	2005	CCRI(II)-K2.Fe-55
^{131}I	2006	CCRI(II)-S6.I-131
^{124}Sb	2007	EUROMET.RI(II)-K2.Sb-124
^{57}Co	2008	CCRI(II)-S6.Co-57
^{67}Ga	2008	CCRI(II)-S2.Ga-67
^{177}Lu	2009	CCRI(II)-K2.Lu-177
^{99m}Tc	2013	BIPM.RI(II)-K4.Tc-99m
^{59}Fe	2014	APMP.RI(II).K2.Fe-59
^{68}Ge/^{68}Ga	2015	CCRI(II)-K2.Ge-68

Table 2: Traceability of LNMRI measurements for radionuclide activity in SIR/BIPM key-comparisons.

Radionuclíde	Date	SIR/BIPM key-comparison
^{60}Co	1985	CCRI(II)-K1.Co-60
^{134}Cs	1987	CCRI(II)-K1.Cs-134
^{57}Co	1991	CCRI(II)-K1.Co-57
^{152}Eu	1995	CCRI(II)-K1.Eu-152
^{133}Ba	1995	CCRI(II)-K1.Ba-133
^{139}Ce	1997	CCRI(II)-K1.Ce-139
^{131}I	1999	CCRI(II)-K1.I-131
^{192}Ir	1999	CCRI(II)-K1.Ir-192
^{51}Cr	2006	CCRI(II)-K1.Cr-51
^{134}Cs	2014	CCRI(II)-K1.Cs-134

Figure 2: *Equivalence degree of the result obtained by LNMRI compared to the KCRV/BIPM for BIPM.RI(II)_K4.Tc-99m (2013).*



The figures 3a, 3b and 4 present radionuclide assays and performance in the comparisons. In general, the performance of the participating laboratories increased significantly during this period, according two statistical criteria: performance D [12, 13].

Figure 3a: *Radionuclides analyzed by participant laboratories for PNI from 1991 to 2016.*

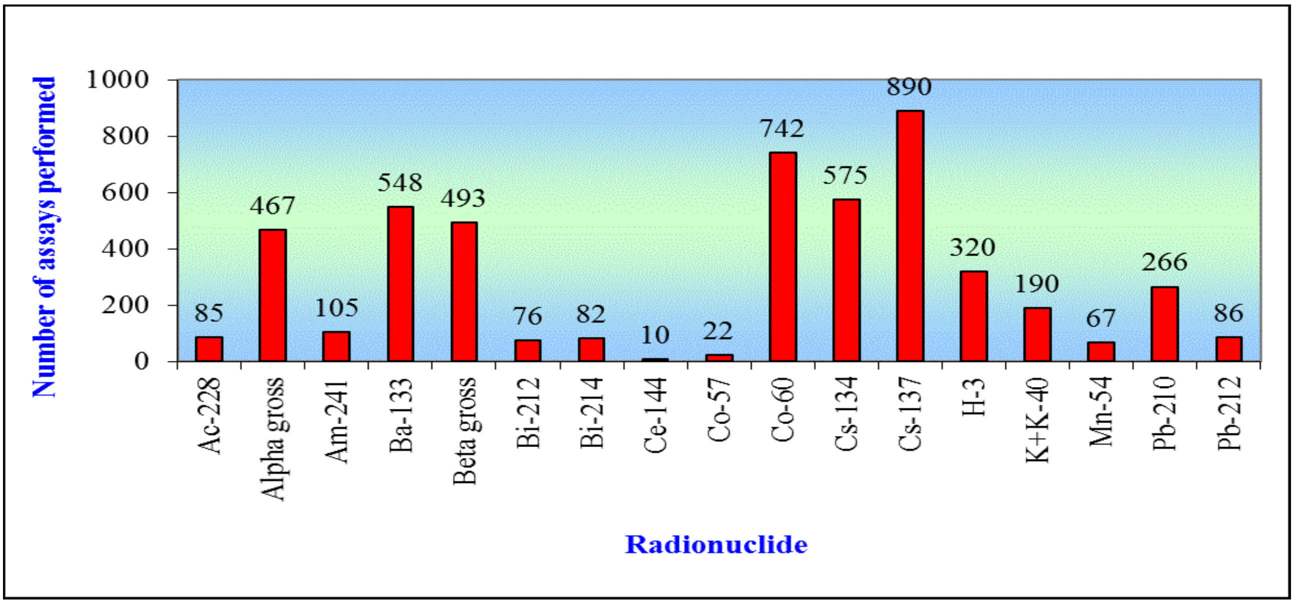


Figure 3b: *Radionuclides analyzed by participant laboratories for PNI from 1991 to 2016.*

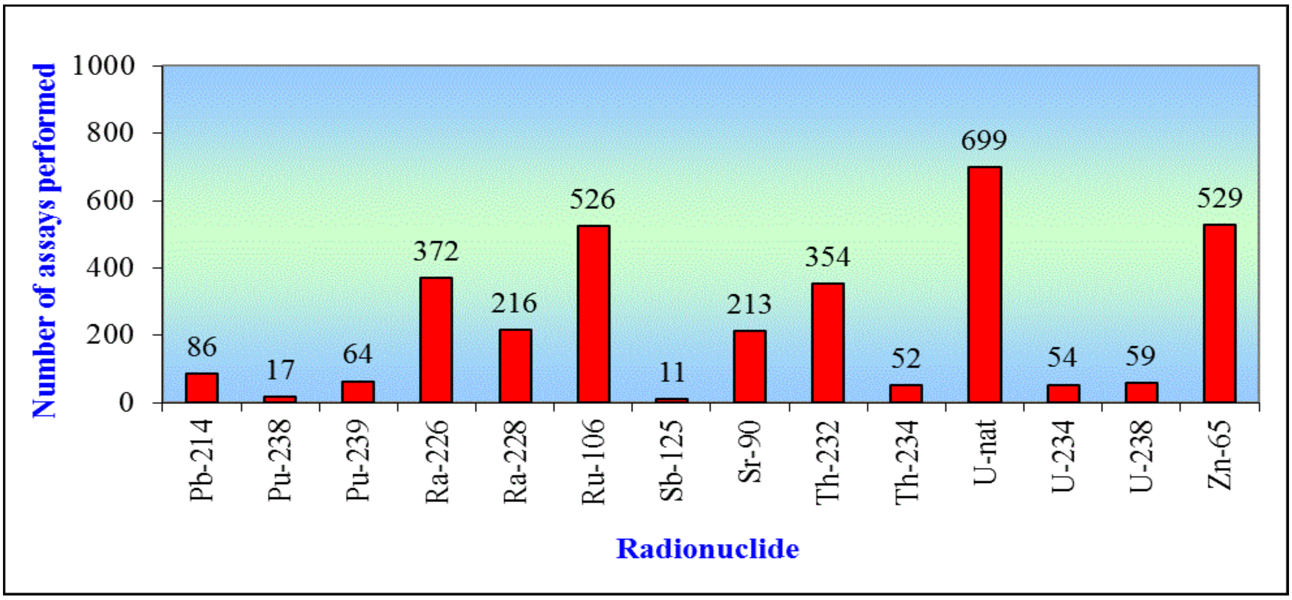
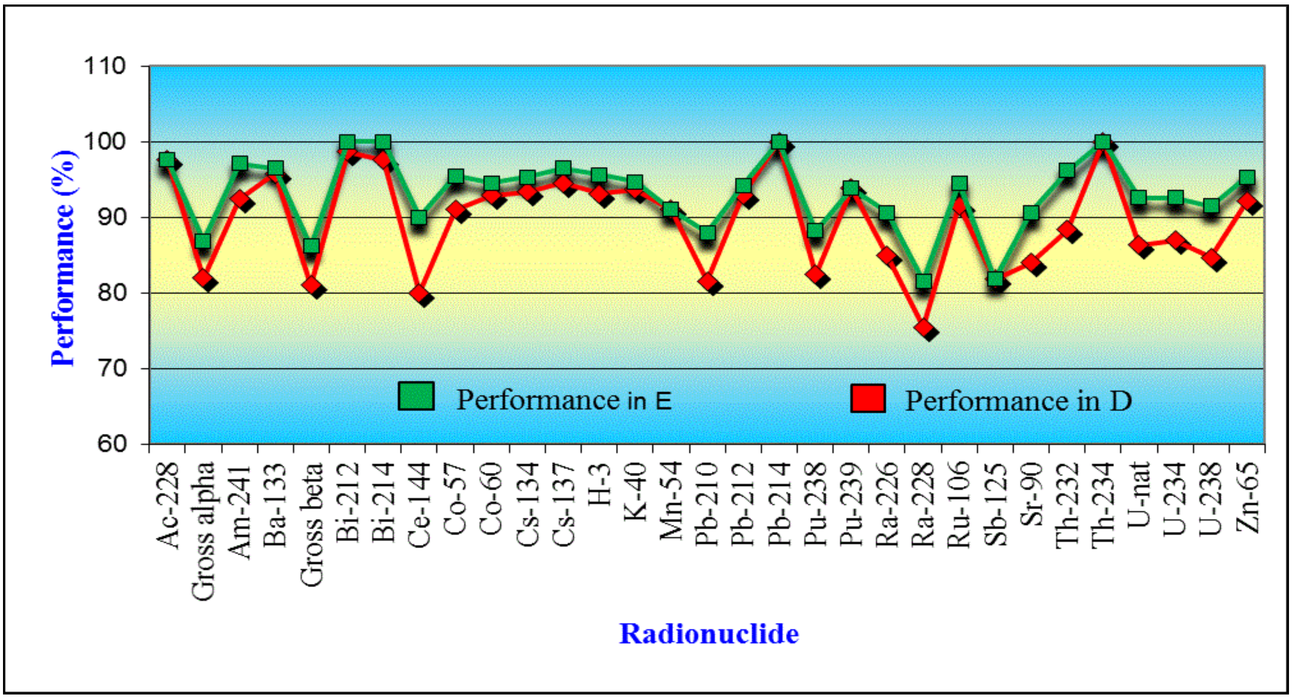


Figure 4. *Performance to radionuclides analyzed by participant laboratories for PNI form 1991 to 2016.*

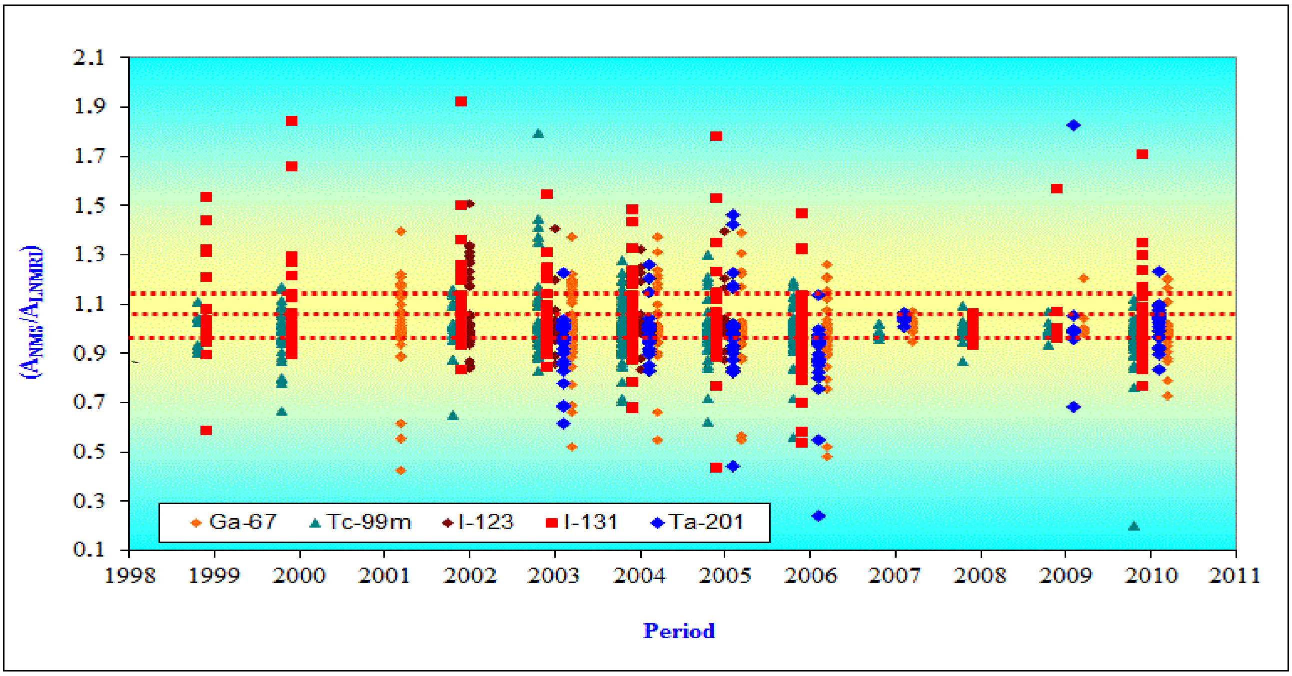


In the 1990s, the LNMRI started a comparison program with hospitals and clinics to provide the traceability of the measurements performed on the radionuclide calibrators, aiming at the optimization of the radioactive doses applied to the patients submitted to the medical procedures of therapy and diagnostic [14,15]. The figure 5 shows the comparisons performed in the period from 1990 to 2010 with ^{67}Ga, ^{99m}Tc, ^{123}I, ^{131}I and ^{201}Tl with the participation of 204 nuclear medicine services (NMS) in many regions of the Brazil.

The criterion for performance evaluation was the value of ratio of the activity measurements made by NMS and LNMRI. The good performance was $0.9 < R < 1.1$, according to the national standard CNEN-NN-3.05 and the performance obtained were: ^{67}Ga (72.8 %), ^{99m}Tc (80.3 %), ^{123}I (73.0 %), ^{131}I (74.8 %), ^{201}Tl (84.4 %).

Due to the high number of nuclear medicine services in Brazil, the national comparison program was modified recently and directed to the radiopharmaceutical producing centers, which could supply traceable radiopharmaceutical solutions to the nuclear medicine services, so that they could also obtain traceability to the LNMRI [16].

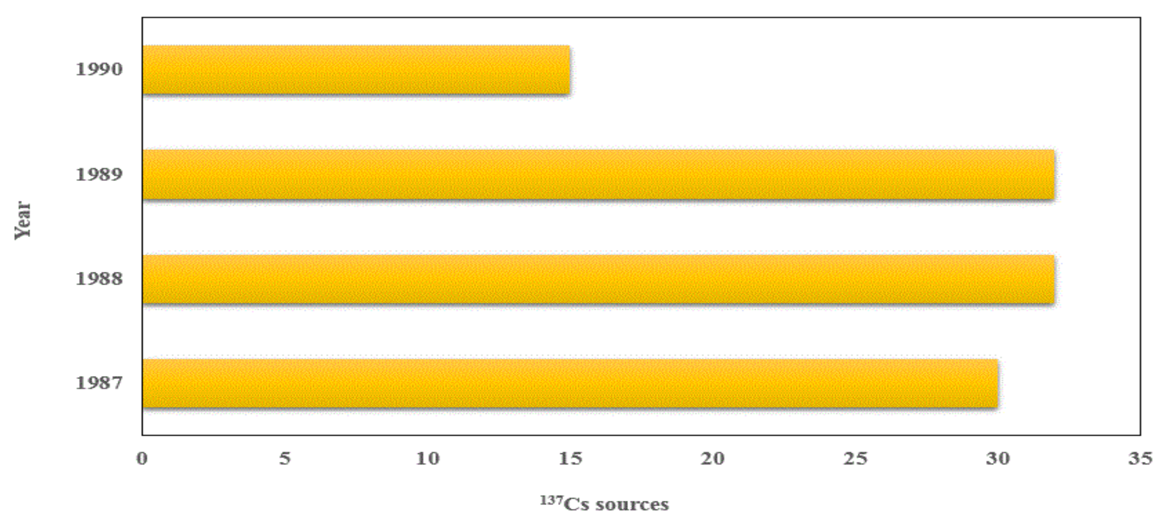
Figure 5: *Performance obtained by NMS/Brazil in LNMRI comparisons for different radionuclides in the period from 1990 to 2010.*



In order to establish traceability of activity measurements and assist in quality control in nuclear medicine in the country, LNMRI promotes quality control programs for Radiopharmaceutical Producer Centers (RPC) and Nuclear Medicine Services (NMS). As a benefit of this program, the RPCs are being certified for the activity measurements of the sources provided to the SMNs. Thus, they comply with National Health Agency (ANVISA) requirement RDC38. They have already been certified for ^{18}F, ^{67}Ga, ^{99m}Tc, ^{123}I, ^{131}I, and ^{201}Tl. NMS have traceability to these radionuclides, except ^{18}F due half-life. As a positron emitter, NMS traceability for this energy must be checked trough ^{68}(Ge+Ga) source that had activity successfully standardized in the frame of an international key-comparison sponsored by BIPM in 2014. Currently, the LNMRI has already emitted certified of calibration and its respective traceability report of the ^{18}F measurements to four radiopharmaceutical producing centers.

LNMRI has been supplied traceable radionuclide standard sources to national and international customers in different geometries for a wide application field, such as researches, industry, environment, agriculture and radiological emergency. The most significant example, it was in the face of the ^{137}Cs accident in Goiania/Brazil, 1987, with the LNMRI prompt response to the various requests for solid and liquid sources of this radionuclide in its multiple geometries for the calibration monitors, individual and environmental monitoring and the carrying out researches. The figure 6 shows the ^{137}Cs sources supplied by LNMRI in the period from 1987 to 1990 regarding the ^{137}Cs accident in Goiania/Brazil to detector calibration, monitoring and research programs.

Figure 6*: LNMRI response to the* ^{137}*Cs accident in Goiânia/Brazil.*



The LNMRI is designated by National Metrology Institute (INMETRO) for activity quantity in ionizing radiation as well as others laboratories for quantities in the fields of mechanical, electricity, thermal, optical, acoustics, time and frequency. Brazil is also a signatory of the Mutual Recognition Arrangement (MRA) under the BIPM, which was signed in 1999 by NMIs of thirty-eight member states of the Meter Convention. It is an arrangement between national metrology institutes which specifies terms for the mutual recognition of national measurement standards and for recognition of the validity of calibration and measurement certificates issued by NMIs [17].

The MRA by means of key-comparisons, demanding that each laboratory operates a quality system applied to the calibration services, which establishes the metrological equivalence among standards and certificates. The NMI's must adopt the requirements of ISO/IEC 17025 standard as the main document. LNMRI has implemented a quality program [18] and since the beginning of 2000 has submitted and approved 2004, 2009 and 2014 its quality management system based on ISO/IEC NBR 17025 competent compliance. This evaluation conducted by international experts has always been confirmed by the Quality System Task Force of the Inter-American Metrology System (QSTF/SIM) and has been performed through peer-review evaluation.

4. CONCLUSION

The LNMRI as designed Brazil's national metrology laboratory is qualified to supply the country's demands in various activity fields, including radiological and nuclear emergency situations, with a high precision and accuracy degree and with the guarantee of the metrological traceability. It also provides its metrological reliability to the quality control of the environmental laboratories and nuclear medicine services in country.

The implementation of the management quality program by LNMRI ensures that the results of its radionuclide standardization, traceable by BIPM key-comparisons, are accepted by the national metrology institutes of the international metrological chain.

REFERÊNCIAS

[1] NCRP - National Council on Radiation Protection and Measurements. **A Handbook of Radioactivity Measurements**. 2nd Edition. Report 58, Bethesda: NCRP, p.592, 1985.

[2] TAUHATA, L.; SALATI, I.; DI PRINZIO, R.; DI PRINZIO, M. A. R. R. **Radioproteção e Dosimetria: Fundamentos**. 9ª. Ed. Rio de Janeiro/Brasil: Instituto de Radioproteção e Dosimetria, 2013.

[3] BIPM-JCGM – Bureau International des Poids et Mesures-Joint Committee for Guides in Metrology. **International vocabulary of metrology: basic and general concepts and associated terms (VIM)**. 3rd edition 2012.

[4] IWAHARA, A.; ALMEIDA, I. P. S. ; TAUHATA, L. ; SILVA, C. J. ; POLEDNA, R. Influences from Delayed Events on the Activity Determination of Se-75 by the Coincidence Counting Method. **Nuclear Instruments and Methods in Physics Research**, v. A, n.339, p. 381-385, 1994.

[5] DA SILVA, C. J.; IWAHARA, A.; POLEDNA, R.; DE OLIVEIRA, E.M.; DI PRINZIO, M.A.R.R.; DELGADO, J. U.; LOPES, R. T. Lopes. Standardization of ^{241}Am, ^{124}Sb and ^{131}I by live-timed anti-coincidence counting with extending dead time. **Applied Radiation and Isotopes,** v. 66, p. 886–889, 2008.

[6] DA CRUZ, P. A. L.; DA SILVA, C. J.; IWAHARA, A.; LOUREIRO, J. S.; DE OLIVEIRA, A. E.; TAUHATA, L.; LOPES, R. T. TDCR and CIEMAT/NIST Liquid Scintillation Methods applied to the Radionuclide Metrology. **IOP Publishing Journal of Physics**. Conference Series 733 012099, 2016.

[7] DE OLIVEIRA, E. M.; IWAHARA, A.; POLEDNA, R.; DELGADO, J. U.; DA SILVA, C. J.; DA SILVA, R. L.; LOPES, R.T. Standardization of ^{65}Zn by sum-peak method. **Applied Radiation and Isotopes**, v. 242, p. 595-598, 2012.

[8] DA SILVA, C. J.; OLIVEIRA, E. M.; IWAHARA, A.; DELGADO, J. U.; POLEDNA, R.; OLIVEIRA, A. E.; MOREIRA, D. S.; R. L. da SILVA; GOMES, R. S.; De VERAS, E. V. Calibration of Ionization Chamber for ^{18}F and ^{68}Ga. **Applied Radiation and Isotopes**, v. 87, p. 188-191, 2014.

[9] DELGADO, J. U.; IWAHARA, A.; SILVA, C. J.; POLEDNA, R.; LOPES, R. T. Absolute measurements of gamma-ray emission probabilities of Yb169. **Nuclear Instruments & Methods in Physics Research A**, v. 455, p. 607-611, 2000.

[10] SANTOS, J. A.; IWAHARA, A.; DE OLIVEIRA, A. E.; SILVA, M. A. L.; TAUHATA, L.; LOPES, R. T.; DA SILVA, C. J. National Intercomparison Program for radiopharmaceutical activity measurements. **Applied Radiation and Isotopes**, v. 60, n.2, p. 523-527, 2004.

[11] TAUHATA, L.; VIANNA, M. E. M. ; OLIVEIRA, A. E.; FERREIRA, A. C. ; BRAGANÇA, M. J. C. S. ; CLAIN, A. F.; FARIA, R. Q. The Brazilian National Intercomparison Program

(ONI/IRD/CNEN): evaluation of 15 years of data. **Journal of Environmental Radioactivity**, v. 86, p. 384-390, 2006.

[12] JARVIS, A. N.; SIU, L. **Environmental Radioactivity Laboratory Intercomparison Studies Program**. Environmental Protection Agency (EPA)-600/4-81-004, EMSL-U.S. EPA, 1981.

[13] BEISSNER, K., 2002. On a measure of consistency in comparison measurements. **Metrologia**, v. 39, p.59 – 63, 2002.

[14] IWAHARA, A.; DE OLIVEIRA, A. E. ; TAUHATA, L.; DA SILVA, C. J.; BRAGUIROLLI, A. M.; LOPES, R. T. Performance of dose calibrators in Brazilian hospitals for activity measurements. **Applied Radiation and Isotopes**, v. 56, n.1-2, p. 361-363, 2002.

[15] TAUHATA, L.; IWAHARA, A.; DE OLIVEIRA, A. E.; REZENDE, E. A.; DELGADO, J. U.; DA SILVA, C. J.; DOS SANTOS, J. A.; NICOLID, I. G.; ALABARSE, F. G.; XAVIER, A. M. Proficiency tests in the determination of activity of radionuclides in radiopharmaceutical products measured by nuclear medicine services in 8 years of comparison programs in Brazil. **Applied Radiation and Isotopes**, v. 66, p. 981–987, 2008.

[16] OLIVEIRA, A. E.; IWAHARA, A.; SILVA, C. J.; DA CRUZ, P. A. L.; POLEDNA, R.; SILVA, R. L.; LARANJEIRA, A. S.; DELGADO, J. U.; TAUHATA, L.; LOUREIRO, J. S.; TOLEDO, B. C.; BRAGHIROLLI, A. M. S.; ANDRADE, E. A. L.; SILVA, J. L.; HERNANDES, H. O. K.; VALENTE, E. S.; DALLE, H. M.; ALMEIDA, V. M.; SILVA, T. G.; FRAGOSO, M. C. F.; OLIVEIRA, M. L.; NASCIMENTO, E. S. S.; DE OLIVEIRA, E. M.; HERRERIAS, R.; SOUZA, A. A. Traceability from governmental producers of radiopharmaceuticals in measuring ^{18}F in Brazil. **Applied Radiation and Isotopes**, v. 109, p. 236-241, 2015.

[17] MRA: Mutual recognition of national measurement standard and of calibration and certificates issued by national metrology institutes, **International Commitee for Weights and Mesures**, BIPM, 1999.

[18] ARAÚJO, M. M.; RODRIGUES, L. N. Implementation of the Quality System at LNMRI/IRD. **Proceedings of the II Brazilian Congress of Metrology**, Generalist Papers, pp 276-282, São Paulo, Brazil, 2000.