National Standards for Radioactivity Measurements: Historical Overview and Current Status

R. Collé

Physics Laboratory, National Institute of Standards and Technology Gaithersburg, MD 20899-8462 USA rcolle@nist.gov

The National Institute of Standards and Technology (NIST), formerly the National Bureau of Standards (NBS), is the nation's metrology laboratory and is the highest authority within the USA for assuring the compatibility and quality of physical measurements [1]. The national basis for accurate radioactivity measurements is established by NIST through various mechanisms:

- (i) developing unique national standards and primary measurement methods;
- (ii) disseminating suitable transfer standards, e.g., the present suite of Standard Reference Materials (SRMs) [2] that are linked to national standards;
- (iii) establishing and providing calibration services;
- (iv) conducting measurement proficiency and traceability testing programs; and
- (v) participating in measurement comparisons with the national metrology institutes of other nations to ensure international consistency.

The NIST has been involved in performing radioactivity measurements and calibrations since 1914. Initially, this was done to support the international radium industry and market, inasmuch as ²²⁶Ra in the two decades following its discovery became the most expensive material on earth, with a value that peaked at about \$ 125 000 USD per gram [3]. Over the following years, NIST prepared, calibrated, and disseminated standards of nuclides in the three naturally-occurring series (headed by ²³⁸U, ²³⁵U, and ²³²Th) in the form of various materials, minerals, ores, and solutions.

In the late 1940s and early 1950s, the need for many other radioactivity standards became manifest with the advent of controlled nuclear fission and with the consequential production and widespread use of manmade radioactive materials from both reactor and accelerator operations [4]. The availability of suitable standards was essential for the increasingly important applications of radioactive materials in industry, medicine, and research. The first of these standards were for radionuclides that for the most part are the obvious and familiar ones: ³H, ¹⁴C, ²²Na, ²⁴Na, ³²P, ⁵⁵Fe, ⁶⁰Co, ⁶⁵Zn, ⁹⁰Sr, ¹³¹I, ¹³⁷Cs, ¹⁹⁸Au, ²⁰⁴Tl, and ²¹⁰Po. During this period, only two laboratories in the world, the National Physical Laboratory (NPL) in the UK and NIST in the USA, were producing and distributing radioactivity standards in quantity and in variety. These two laboratories, under the aegis of the International

Commission on Radiological Units (ICRU) and along with the Canadian National Research Council laboratory at Chalk River prepared the necessary samples and began to conduct informal measurement comparisons with the metrology laboratories of all interested countries [5], in which standardization activities soon proliferated. The success of the effort for over a decade led to recognition, in 1963, that the comparisons should be more formalized and that the proper repository for coordination of these international activities was the International Bureau of Weights and Measures (BIPM) in Sèvres, France. Interestingly, the uncertainty values provided on NIST standard certificates until some time in the 1960s was not based on detailed uncertainty component assessments, but rather was based on the "accuracy" reflected in measurement agreements obtained by comparisons with other laboratories.

A modern era for standards can be thought to have originated in the 1970s, following a number of systematic "needs" studies [6-8] that identified the increasing demand for standards in more and more rapidly growing areas of application -- particularly for radiopharmaceutical and environmental measurements. This resulted in collaborations between NIST and users in various measurement communities in establishing "traceable" measurement systems, involving secondary laboratories, measurement quality assurance (MOA) programs, proficiency testing, and the development of a wider array of SRMs. A joint program with the forerunners of the Nuclear Energy Institute (NEI) is the oldest and perhaps the most important of these [9,10] inasmuch as it became a NIST hallmark MQA program upon which many others were modeled. This program provides SRMs of very short-lived nuclides for nuclear medicine quality control that are not likely to be otherwise available. These radiopharmaceutical standards are distributed not only to NEI participants, but are also made available at select announced times to the general public. They (with their approximate half-lifes) include: ^{125}I (59 d); ^{131}I (8.0 d); ^{133}Xe (5.2 d); ^{67}Ga (3.3 d); ^{201}Tl (3.0 d); ^{111}In (2.8 d); ^{99}Mo (2.7 d); ^{90}Y (2.7 d); and $^{99}\text{m}\text{Tc}$ (0.25 d) [2].

The late 1970s also saw the emergence of another new and novel aspect of radioactivity standards, *viz.*, the development of natural matrix SRMs [11]. Based on an evident need identified by the worldwide low-level environmental measurement community, NIST initiated a

program to produce standards of radionuclide-bearing sediments, soils, and biological materials that were collected from natural sites. These natural matrix SRMs consist of ground, homogenized powders that are characterized for as many as 20 radionuclides at environmental activity levels. They are intended to be used as "real" samples that can evaluate radioanalytical methods and test a laboratory's capability of performing environmental radioactivity measurements in similar matrices at low levels. Uniquely, the calibration measurements leading to certification for these standards are obtained in cooperation with other highly experienced national and international environmental measurements laboratories. The natural matrix SRMs currently or soon to be available are: Columbia River Sediment; Human Lung; Human Liver; Rock Flats Soil #2; Freshwater Lake Sediment; Peruvian Soil; Ashed Bone; Ocean Sediment; Ocean Shellfish; and Seaweed [2].

At the present time, there are approximately 65 radioactivity SRMs available in a number of configurations, although stable solutions of single species of about 45 different alpha, beta, gamma and x-ray emitting nuclides, certified for massic activity, are the most common form. The certified relative expanded uncertainties (combined standard uncertainty multiplied by a coverage factor of k = 2) for most SRMs is typically less than 1 % and is almost never greater than 2 %.

A vigorous applied research program in fundamental radionuclidic metrology, supported by upwards of 40 unique instruments and methods [12], underlies the continued supply of SRMs and the development of new standards.

REFERENCES

- Although this laboratory was called the NBS from its founding in 1901 until 1988, all subsequent historical cites will refer to it as NIST. Cf., R.C. COCHRANE, Measures for Progress: A History of the National Bureau of Standards, U.S.. Dept. Commerce, Washington., DC (1966); E. PASSAGLIA, A Unique Institution. The National Bureau of Standards, 1950-1969, NIST Spec. Publ. 925, U. S. Govt. Printing Office, Washington, DC (1999); J. SCHOOLEY, Responding to National Needs: The National Bureau of Standards Becomes the National Institute of Standards and Technology (1969-1993), NIST Spec. Publ. 955, U. S. Govt. Printing Office, Washington, DC (2000).
- NATIONAL INSTITUTE OF STANDARDS AND TECHNOLOGY (NIST), Standard Reference Materials for Radioactivity, NIST, Gaithersburg, MD (June, 2006); Standard Reference Material Catalog, Spec. Publ. 260, NIST, Gaithersburg, MD (2007).

- 3. B.M. COURSEY, R. COLLÉ, J.S. COURSEY, "Standards of Radium-226: From Marie Curie to the International Committee for Radionuclide Metrology," *Appl. Radiat. Isot.* **56**, 5 (2002).
- 4. W.B. MANN, "The Preparation and Maintenance of Standards of Radioactivity," *Int. J. Appl. Radiat. Isot.* 1, 3 (1956).
- 5. A.V. ASTIN, E.C. BULLARD, W.B. LEWIS, "International Comparisons of Radioactivity Standards, *Science* **123**, 895 (1956).
- 6. NATIONAL ACADEMY OF SCIENCES (NAS), National Uses and Needs for Standard Radioactive Materials, NAS, Washington, DC (1970).
- 7. L.M. CAVALLO, B.M. COURSEY, S.B. GARFINKEL, ET AL., Needs for Radioactivity Standards and Measurements in Different Fields, *Nucl. Instrum. Meth.* **112**, 5 (1973).
- 8. R. COLLÉ, "Radon Measurements: National Needs and the Role of NBS," *Trans. Amer. Nucl. Soc.*, **39**, 84 (1981); *A Survey of Radon Measurement Needs and Activities in State Radiation Control Programs*, jointly publ. Conference of Radiation Control Program Directors, Inc. and NBS, U.S. Govt. Printing Office, Washington, DC (1982).
- 9. R. COLLÉ, "AIF-NBS Radioactivity Measurement Assurance Program for the Radiopharmaceutical Industry," *Measurements for the Safe Use of Radiation*, Natl. Bur. Stds. Spec. Publ. 456, p. 71, U.S. Govt. Printing Office, Washington, DC (1976).
- D.B. GOLAS, "NIST Radiopharmaceutical Standard Reference Materials and the NEI/NIST Radiopharmaceutical Measurement Assurance Program," Appl. Radiat. Isot. 49, 329 (1998).
- K.G.W. INN, Z. LIN, Z. WU, ET AL., The NIST Natural Matrix Radionuclide Standard Reference Material Program for Ocean Studies, *J. Radioanal.* Nucl. Chem. 248, 227 (2001).
- 12. D.D. HOPPES, "Basic Radionuclide Measurements at the U.S. National Bureau of Standards," *Environ. Int.* **10**, 99 (1984).

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