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Activity standardization of ^{131}I at CENTIS-DMR and PTB within the scope of a bilateral comparison

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ABSTRACT

The activity of an ^{131}I solution was measured at the Cuban Institute, CENTIS-DMR, as well as at the German National Metrology Institute, PTB, within the scope of a bilateral comparison. In particular, the comparison is aimed at an investigation of the measurement capabilities of CENTIS-DMR which provides activity standards in Cuba and organizes national comparisons, placing a particular emphasis on radionuclides for nuclear medicine, such as ^{131}I. Both institutes applied liquid scintillation counting techniques with efficiency tracing as well as secondary standardization procedures by means of calibrated ionization chambers and gamma-ray spectrometers. The results were checked for consistency and a good agreement was found. Moreover, a virtual link of the Cuban result to the International Reference System (SIR) at the Bureau International des Poids et Mesures (BIPM) is discussed.

1. Introduction

Although ^{131}I is being increasingly replaced by shorter lived radionuclides such as ^{18}F, ^{99m}Tc and ^{123}I it is still frequently used in nuclear medicine, e.g. for the diagnosis of thyroid carcinoma. In Cuba, it is utilized in several hospitals which can obtain activity standards from the Radionuclide Metrology Department of the Center of Isotopes of Cuba (CENTIS-DMR). CENTIS-DMR also organizes national intercomparisons to control and improve the quality of activity measurements in the country (Oropesa et al., 2006).

The traceability of the Cuban activity standards to the International Standard (SI) is desirable to ensure a high quality of applications. However, a direct submission of an ampoule to the International Reference System (SIR) at the Bureau International des Poids et Mesures (BIPM) is not possible since Cuba is only an associate member of the Metre Convention.

In this work, a bilateral comparison between CENTIS-DMR and the German National Metrology Institute, the Physikalisch-Technische Bundesanstalt (PTB), is presented. Aliquots of a radioactive solution of ^{131}I were measured in both institutes, using liquid scintillation counting in combination with the CIEMAT/NIST efficiency tracing method. In addition, secondary activity standardization techniques by means of reentrant 4π ionization chambers and gamma-ray spectrometry were applied. The aim of the comparison was to check the results for consistency and to check the measurement capabilities of CENTIS-DMR.

In 2004, the PTB submitted two samples of known activity of ^{131}I to the SIR at the BIPM. Thus, the comparison presented in this work also allows the creation of a virtual link to the SIR for CENTIS-DMR. Another possibility to obtain this link would be given via the recent International Atomic Energy Agency (IAEA) comparison in which CENTIS-DMR participated (Zimerman et al., 2008). The solution used for that comparison was provided by QSA Global (Braunschweig, Germany) and is traceable to the activity standards of PTB. Two participants of the IAEA comparison also submitted ampoules to the SIR recently, which would allow a further link to the key comparison value (KCRV) of the SIR (see Ratel et al., 2008 and references therein).

2. Experimental details

2.1. ^{131}I solution

A solution of the radionuclide ^{131}I used for the comparison with a total volume of 34 mL was prepared at CENTIS-DMR. The approximate activity concentration was 3.5 MBq/g on the reference date of March 14, 2006, at 16:00 UTC. The chemical composition of the solution was Na^{131}I in 0.9% NaCl, 0.25% $Na_2S_2O_3 \times 5H_2O$, 0.26% KI, 0.1% NaH_2PO_4 and 0.44% $(NH_4)_2HPO_4$.

A weighed portion of about 4 g of the ^{131}I solution was transferred into a P6-type ampoule, which was then sent to PTB.

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Another part of the solution was used at CENTIS-DMR for the activity measurements. The solution was checked for potential radioactive impurities in both laboratories. Neither laboratory could detect any gamma-ray emitting impurity. The participants agreed to use the half-life as evaluated by Schötzig and Schrader (2000), i.e. $T_{1/2} = 8.0207(9)$ d.

2.2. Activity measurements at CENTIS-DMR

At CENTIS-DMR, secondary activity standardization was carried out by means of a high-purity Germanium (HPGe) gamma-ray spectrometer (endcap 80 mm ∅; Be entrance window; rel. efficiency 30%). The detector was calibrated with the aid of activity standards of the gamma-ray emitting nuclides ^{57}Co, ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu and ^{241}Am in a point-source geometry. The standards were purchased from the National Metrology Institute of Hungary (MKEH, formerly OMH). The results of the ^{131}I measurements were also used to establish a calibration factor for a Capintec CRC 35 R^{TM} ionization chamber. The stability of the latter system is monitored by means of a long-lived ^{137}Cs source. Details on the standard sources and the calibration procedures were described by Oropesa et al. (2002).

In addition, liquid scintillation counting with an LKB-Wallac Rackbeta 1209 counter was applied to determine the activity concentration. In this system, the quench indicating parameter SQP(E) is determined with an external ^{226}Ra source. Small portions of about 30 mg of a diluted solution prepared from the ^{131}I solution used for the comparison were put into 20-mL low-potassium glass vials containing 15 mL of HiSafe^{TM} III scintillation cocktail. Masses of all samples used for the ^{131}I standardization at CENTIS-DMR, as well as the dilution factors, were determined with a Sartorius MC 210 S balance traceable to the Cuban national mass standard. All in all, 12 samples plus one background sample were prepared and nitromethane (CH_3NO_2) was used as quenching agent. The counting efficiency was determined according to the CIEMAT/NIST method with ^{3}H as a tracer. (Grau Malonda, 1999; Broda et al., 2007). For the calibration measurements, a ^{3}H solution was used which had been standardized at the Laboratoire National Henri Becquerel (CEA-LNHB) in France. Two counting efficiency tables were computed. One of them was obtained using the PTB code described below. The second efficiency table was calculated with the SOBEGA code employing a kB parameter equal to 0.0075 cm/MeV (Garcia-Toraño, 2006). The tracer efficiency was varied over a range from about 39.87% to 27.28% which corresponds to ^{131}I efficiencies of about 97.81% and 96.92%, respectively. The LSC activity concentration reported by CENTIS-DMR is the mean of the two values obtained using these two counting efficiency tables.

Applying the three methods for determining the activity concentration, a, on the reference date of 14th March 2006 (16 h UTC), the following results were obtained at CENTIS-DMR:

Gamma-ray spectrometry :	$a = (3.481 \pm 0.045)$ MBq/g,
Ionization chamber measurements :	$a = (3.484 \pm 0.065)$ MBq/g,
Liquid scintillation counting :	$a = (3.479 \pm 0.026)$ MBq/g.

The stated uncertainties are standard uncertainties ($k = 1$). The final result $a = 3.479(26)$ MBq/g is the result obtained by liquid scintillation counting, which is—in the case of ^{131}I-considered to be a primary activity standardization technique.

2.3. Activity measurements at PTB

Weighed portions of about 2 g of the solution were transferred into two flame-sealed PTB-type glass ampoules and then measured against a long-lived ^{226}Ra source by means of a calibrated 4π-ionization chamber of type IG12/A20, Centronic 20th Century Electronics Ltd. The chamber has an iron entrance window of approximately 3 mm thickness and was filled with argon to a pressure of 2 MPa. The ionization current was measured by means of a commercially available Keithley electrometer model 6517A. The activity A in this standard geometry is given by

$$A = k_N \, m_{Ra\text{-}226} \, R_{I\text{-}131}/R_{Ra\text{-}226}, \quad (1)$$

with $R_{I\text{-}131}$ and $R_{Ra\text{-}226}$ being the averaged ionization chamber readings of the current measuring electronics, corrected for background and decay. A more comprehensive description of the methods has recently been presented by Schrader et al. (2007).

The particular radionuclide calibration factor $k_N = 1/\varepsilon_{I\text{-}131}$ was determined in 1980 by means of 4πβ–γ coincidence counting. In 2004, the calibration factor was confirmed by 4πβ–γ coincidence counting and liquid scintillation counting measurements within the scope of a comparison at the International Reference System, SIR (Ratel et al., 2005). The excellent agreement of the activity determined by means of the ionization chamber with the aid of the old calibration factor and the activity obtained by means of primary methods in 2004 demonstrates the outstanding long-term stability of the system. This stability has been achieved particularly since all current measurements are traced back to the current measurements of a long-lived ^{226}Ra reference source. This technique allows compensating deviations due to changes in the climate conditions, and even a small leakage of the counting gas would be acceptable.

The samples for liquid scintillation counting were prepared with 15 mL Ultima Gold^{TM} scintillator, about 0.97 mL of distilled water, and a weighed portion of about 30 mg of the diluted ^{131}I solution in 20-mL low-potassium borosilicate glass vials. The solution masses of all samples prepared at PTB, as well as the dilution factor, were determined gravimetrically using two Mettler balances traceable to the German national mass standard. Eight samples were prepared with the radioactive solution and one sample without active solution in order to measure the background counting rate, which was then subtracted. As a quenching agent, nitromethane (CH_3NO_2) was used. All samples were shaken and centrifuged and then measured in a Wallac 1414 Guardian^{TM} liquid scintillation spectrometer with two photomultiplier tubes. The quenching indicator $SQP(E)$ was measured by means of an external source of ^{152}Eu. The calibration curve, i.e. the counting efficiency of ^{3}H as a function of the quenching indicator $SQP(E)$, was measured with the aid of a PTB standard solution of ^{3}H, standardized by internal gas counting (Günther, 1993).

2.4. Efficiency calculations

The counting efficiencies were computed according to the CIEMAT/NIST method by means of a PTB code that comprises components of other programmes. The methods to compute the counting efficiency of a β branch with a couple of γ branches presented by Günther (1994) were improved (see Appendix). The β branches were calculated by means of modified subroutines of the EFFY4 code, which is an updated version of EFFY (Garcia-Toraño and Grau Malonda, 1985). Altogether, 6 β branches and 17 γ branches in 16 cascades (path from ^{131}I to the stable ^{131}Xe) were considered. A cascade comprises one β branch and up to three coincident γ branches. Decay data were taken from TdeR (2006).

The ionization quenching function was calculated by means of the procedures described in a previous article, taking into account the atomic composition of the samples (Kossert and Günther, 2004). The dependence on the ionization quenching factor kB was