

In vivo monitoring in radiological emergencies: a program to intercompare the physical quantity activity on portable detectors

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Abstract. One of the goals of metrology is to provide reliability in measurement systems. Recently, portable detectors, which ones are calibrated in the ambient dose equivalent rate or exposure rate, have been applied in "in vivo" dosimetry activities. Thus, there are several methodologies for calibrating, with the usage of different anthropomorphic physical simulators, resulting in efficiency-energy calibration curves, which cannot be directly applied in intercomparison exercises. The present work suggests an easily reproducible methodology to provide the intercomparison of the efficiencies. The MCNPX mathematical simulation code was used to determine an optimized geometry. A cylindrical physical simulator filled with water, with sealed sources of the ⁵⁷Co, ¹³³Ba and ¹³⁷Cs placed in the geometric centre, was submitted to a measurement of counts, where a NaI(Tl) detector was used to determine the efficiencies at 0.20 m distance. When the efficiencies of the ¹³⁷Cs point source and ¹³⁷Cs volumetric sources were compared, the standard deviation was 7.9%. It is expected the efficiencies reference value of the 2.7 x 10⁻⁴ CPS/Bq and an interval of $\pm 10\%$ of this value to intercomparison exercises and verifying the constancy of the equipment's response in its use in internal dosimetry activities.

1. Introduction

The Metrology is the science of measurement and its applications, which includes all theoretical and practical aspects of measurement, whatever the measurement uncertainty and field of application are [1]. One of its objectives is to provide reliability in measurement systems.

In the field of ionizing radiation, metrology provides a safe application of nuclear energy in industry, medicine, agriculture, electricity supply and in radiological and nuclear emergencies. The portable detectors equipped with the gamma radiation spectrometry function used in emergencies are factory standard calibrated generally in the physical quantities of ambient dose equivalent rate (μ Sv/h), exposure rate (mR/h) and count rate (CPS or CPM).

In another perspective of use, these detectors have been applied in internal monitoring, which aims to determine the activity of a radionuclide incorporated by an individual, based on measurements of the count number from his body. From this count, the committed effective dose can be determined using ICRP biokinetic models and dose coefficients available in the literature. The application of portable detectors is an additional alternative to the activities carried out by laboratories dedicated to internal



dosimetry, keeping in mind a possible high demand for this type of service in a real situation of radiological or nuclear emergency.

Regarding, several authors [2], [3], [4], [5] proposed calibrations in portable detectors, of the scintillator or semiconductor type, using anthropomorphic physical simulators available in their laboratories, filled with a radionuclide solution or sealed sources, in their own geometries, resulting in different efficiency-energy calibration curves. In a real radiological or nuclear emergency situation, these portable systems can be used, reproducing the same calibration geometry. Knowing the radionuclide, by spectrometry, and consequently the photopeak's energy, the efficiency-energy calibration curve can determine the counting efficiency, given in CPS/Bq. Then, the activity of the incorporated radionuclide can be determined from the measured count rate (CPS). With this activity data, using the biokinetic models available in the literature and the retention fraction m(t), the incorporation I (Bq) is determined for an adult individual, applying the dose coefficients (μ Sv/Bq) of this radionuclide.

With the evolution of the number of calibrations of these portable detectors, for use in internal monitoring, carrying out intercomparison exercises turns out to be extremely important. However, currently, there is no way to carry out a direct intercomparison of the efficiency-energy curves, precisely because the methodologies employed are different.

Thus, the present work proposes a new and simple methodology, which is easily reproducible, to provide the intercomparison of the efficiencies of portable detectors used in internal dosimetry, applied in radiological and nuclear emergencies, promoting the metrological traceability of physical quantity activity and the reliability of calibrated systems.

2. Materials and Methods

Initially, it was necessary to select a low-cost, easily reproducible physical simulator. A cylindrical acrylic tank, with a diameter of 0.30 m and a height of 0.35 m, with a thickness of 0.004 m, filled with water to a height of 0.30 m was selected to reproduce the scattering of gamma radiation in biological tissues. A portable NaI(Tl) detector (0.0762×0.0381) m², with 1024 channels and detection range in gamma photon energy from 25 keV to 3 MeV, for operational use in radiological and nuclear emergencies, was used. This detector operates in a temperature range of -20°C to 60°C and relative humidity up to 100% [6].

The radionuclides selected for calibration were ⁵⁷Co, ¹³³Ba and ¹³⁷Cs, as they emit gamma radiation in the range of 122-661 keV. Furthermore, ¹³⁷Cs has the following advantages: long half-life (30 years), well-known gamma photopeak energy (661.6 keV, 85.05%) [7] and be widely used in calibration work. Being a fission product in thermonuclear power plants, it is strongly likely to be an agent of internal contamination in real accidents situations.

In order to determine the number of ¹³⁷Cs point sources to be placed inside the cylindrical simulator and the distances between the sources and the detector, a mathematical simulation was performed with the MCNPX Monte Carlo code, version 2007 [8]. This process allowed an ideal geometry for the experiment. Six arrangements of ¹³⁷Cs point sources were simulated, the simplest being a source in the geometric center of the water volume of the simulator. The other arrangements were: 3 and 5 sources in line, on the horizontal axis, parallel to the base, passing through the geometric center of the water volume, spaced 0.05 m from each other; three cross-shaped arrangements of 5, 9 and 13 sources, in the vertical plane, passing through the geometric center of the volume of water, spaced 0.05 m from each other. In all arrangements, the aligned or cross-shaped, the sources planes were parallel to the face of the detector window.

With these arrangements of 137 Cs sources, distances from the detector window to the nearest external face of the simulator of 0.005 m, 0.010 m, 0.020 m, 0.030 m, 0.050 m, and 0.070 m were used. From 0.100 m to 0.500 m, the distances were spaced from 0.050 m to 0.050 m, and from 0.600 m to 1.000 m, from 0.100 m to 0.100 m, making up 20 distances, from 0.005 m to 1.000 m.



According to the results of the mathematical simulation, the experiment was set up with a sealed point source of ¹³⁷Cs, with activity of $(1.86 \times 10^5 \pm 5.58 \times 10^3)$ Bq, at the time of the experiment. The NaI(Tl) detector (0.0762 x 0.0381) m² was placed on a tripod, with the axial axis of the active volume coinciding with the geometric center of the volume of water, in which the source of ¹³⁷Cs was located (figure 1). The selected distance, based on the mathematical simulation, was 0.20 m. Background (BG) and ¹³⁷Cs source counts were measured for 86,400 s (24 h). Background radiation counts were obtained to be discounted from the source count number in the corresponding Region of Interest (ROI) on the ¹³⁷Cs photopeak.



Figure 1. Experimental setup.

Using Equation 1, the efficiencies from the simulation and the experiment were calculated, which could then be compared in order to validate the data obtained [9].

$$\varepsilon = (N_{ctg} \Delta t) / (A \times I\gamma)$$
⁽¹⁾

 N_{CTG} is the net count number, Δt is the count time, A is the activity on the date of measurements, and I γ is the gamma emission intensity with photopeak energy of the radionuclide.

With the proposal of an easily reproducible methodology, three sealed volumetric sources (57 Co, 133 Ba and 137 Cs) in an epoxy matrix in a 20 mL flask were used to determine the channel-energy calibration curve and the efficiency-energy curve, in the range from 122 keV to 661 keV. These sources are commonly used in quality control tests in nuclear medicine. The three sources were placed together at the geometric center of the volume of water in the cylindrical acrylic simulator and measurements of the number of counts were performed with the NaI(Tl) detector (0.0762 x 0.0381) m², at a distance of 0.20 m and counting time of 3,600 s. This process was repeated using only the 137 Cs volumetric source, in the center of the simulator. Thus, it was possible to compare the counting efficiencies when using the sealed point source and the volumetric sources.

3. Results and Discussion

About the mathematical simulation, the obtained count numbers per story were compared, at each distance, for all proposed sealed point source geometries. The distance that presented the lowest standard deviation, among all others, was 0.200 m (table 1), and therefore selected for the experiment.

lined up			cross-shaped			Standard
1 source	3 sources	5 sources	5 sources	9 sources	13 sources	deviation
2,3781 x 10 ⁻⁴	2,3899 x 10 ⁻⁴	2,3748 x 10 ⁻⁴	2,3437 x 10 ⁻⁴	2,3105 x 10 ⁻⁴	2,3047 x 10 ⁻⁴	1 55 0/
± 1,18 %	\pm 0,65 %	\pm 0,65 %	\pm 0,65 %	\pm 0,66 %	\pm 0,66 %	1,33 %

Table 1. Counting numbers per story – distance 0.20 m.



Considering, on the date of the experiment, the activity of the ¹³⁷Cs source (1.86 x $10^5 \pm 5.58$ x 10^3 Bq), the counting rate obtained (4.594 x $10^1 \pm 0.03$) CPS and the emission intensity I $\gamma = 0.8505$, the experimental efficiency resulted was $\mathcal{E} = (2.902 \text{ x } 10^{-4} \pm 8.710 \text{ x } 10^{-6})$ CPS/Bq.

The simulation efficiency was calculated from the number of counts per story (2.3781 x 10⁻⁴), considering the number of stories as the number of gamma radiation emissions, originating from the same sealed source of ¹³⁷Cs, for a time of 86,400 s (1.6079 x 10¹⁰). Thus, it resulted in a count rate of (4.425 x 10¹ ± 0.29) CPS. The simulation efficiency was then $\mathcal{E} = (2.796 \text{ x } 10^{-4} \pm 8.583 \text{ x } 10^{-6})$ CPS/Bq. The difference between experimental efficiency and simulation efficiency was 3.80%.

Considering that the uncertainties of the experimental and simulation efficiencies were, respectively, 3.00% and 3.07%, and comparing with the difference of 3.80% between these efficiencies, it appears that the values of the efficiencies were convergent, validating the experimental arrangement.

The channel-energy curve obtained with the use of ⁵⁷Co, ¹³³Ba and ¹³⁷Cs sources was linear, as expected (figure 2).



Figure 2. Channel-Energy Curve.

The table 2 shows the count number for each sealed source (⁵⁷Co, ¹³³Ba and ¹³⁷Cs), background radiation, count rate and efficiencies.

ROI (ch)	CTG	BG	CTG Net	CPS	Efficiency (CPS/Bq)
(26.42) 5700	5,71 x 10 ⁷	1,37 x 10⁵	5,70 x 10 ⁷	1,58 x 104	3,82 x 10 ⁻⁴
(50-42) - * C0	± 7,56 x 10 ³	± 3,70 x 10 ²	± 7,57 x 10 ³	± 2,10 x 10 ⁰	± 1,14 x 10 ⁻⁵
(109-131) -	9,00 x 10 ⁶	8,05 x 10 ⁴	8,92 x 10 ⁶	2,48 x 10 ³	4,90 x 10 ⁻⁴
¹³³ Ba	± 3,00 x 10 ³	± 2,84 x 10 ²	± 3,01 x 10 ³	± 8,37 x 10 ⁻¹	± 1,47 x 10 ⁻⁵
(105 247) ¹³⁷ Cc	3,82 x 10 ⁶	5,49 x 10 ⁴	3,77 x 10 ⁶	1,05 x 10 ³	2,51 x 10 ⁻⁴
(195-247)CS	± 1,96 x 10 ³	± 2,34 x 10 ²	± 1,97 x 10 ³	± 5,47 x 10 ⁻¹	± 7,52 x 10 ⁻⁶

Table 2. Efficiencies - ROI (122 keV, 356 keV, 661 keV).

The energy-efficiency curve obtained follows in figure 3, agreeing with the expected result for a NaI(Tl) scintillator type detector in this energy range [10].



Figure 3. Efficiency-Energy Curve.

The efficiency obtained with measurements carried out only with the ¹³⁷Cs source at the geometric center of the simulator was $(2.60 \times 10^{-4} \pm 7.81 \times 10^{-6})$ CPS/Bq.

Thus, comparing the efficiencies obtained in the experiments in the case of the sealed point source of 137 Cs (2.90 x $10^{-4} \pm 8.71$ x 10^{-6}) CPS/Bq, sealed volumetric source of 137 Cs of 20 mL (attached with the sources of 57 Co and 133 Ba) (2.51 x $10^{-4} \pm 7.52$ x 10^{-6}) CPS/Bq, and only the 20 mL volumetric sealed source of 137 Cs (2.60 x $10^{-4} \pm 7.81$ x 10^{-6}) CPS/Bq, the percentual deviation of these three values was of the order of 7.96%.

It is expected with the reproduction of the present methodology in a greater number of NaI(Tl) scintillator detectors that the results obtained of the efficiencies tend to a reference value of (2.67×10^{-4}) CPS/Bq. An interval of $\pm 10\%$ of this value could be applied for verifying the constancy of the equipment's response over time.

4. Conclusion

The methodology adopted was simple assembly, using sealed sources of 137 Cs, 57 Co and 133 Ba inserted in the geometric center of the water volume of the acrylic phantom, with the detector at 0.20 m of the phantom. At that distance, the efficiencies for the energy of 661 keV were within an average variation of 7.96%.

It is expected that, with the spreading of the methodology of this work, the efficiencies obtained, from several other portable NaI(Tl) scintillator detectors, are within a range of up to $\pm 10\%$ of a reference value.

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