The Monte Carlo Method: Versatility Unbounded In A Dynamic Computing World
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MONTE CARLO MODEL OF IONIZATION CHAMBERS USED FOR ACTIVITY MEASUREMENTS

Veronika Olšovcová
Czech Metrology Institute
Radiova 1, 102 00 Prague, Czech Republic
volsovcova@cmi.cz

ABSTRACT

The quality assurance in nuclear medicine requires regular accuracy checks of devices used for the determination of the activity of radioactive pharmaceuticals administered to the patients in vivo. All the tested devices, well-type high pressure ionization chambers often called radionuclide calibrators, are in the Czech Republic checked and/or calibrated against secondary national standard of activity; i.e. 4πγ ionization chamber of the Czech Metrology Institute.

Recently, the demand for measuring activity of radionuclides that are not yet routinely used has been increasing. As a routine determination of calibration coefficient of the chamber for the particular radionuclide is not always economical, it is useful to be able to predict calibration coefficients from the knowledge of their decay scheme. Furthermore, it is practical to estimate suitable calibration coefficients or correction factors for different containers and volumes of the sample.

By means of the Monte Carlo MCNP4C code basic characteristics, such as the response function, sample position dependence and sample volume dependence, of the 4πγ ionization chamber were calculated. Similar model was created for a commercially available chamber Bqmeter, which is routinely used in nuclear medicine departments. The calculated and experimentally determined data were compared. On the whole, the results are satisfactory.

Key Words: radionuclide calibrator, comparison, response function

1 INTRODUCTION:

For the maximum safety of patients, the quality assurance in nuclear medicine requires regular accuracy checks of devices used for the determination of the activity of radioactive pharmaceuticals administered in vivo. In the Czech Republic annual accuracy checks, obligatory by law, lie in the responsibility of the Czech Metrology Institute (CMI).

The activity of radiopharmaceuticals is usually determined using so-called radionuclide calibrators or activimeters. Radionuclide calibrators are well-type ionization chambers, filled typically with argon to a pressure up to several MPa. The control unit of the device converts the current (pA) measured by an electrometer to units of activity (MBq) by means of the calibration coefficients, specific for each radionuclide. These are determined experimentally by direct comparison of the chamber response with the activity value determined by absolute methods or calculated from response function fitted to the measured data.

All calibrators are checked and/or calibrated against the national secondary standard of activity; i.e. CMI's 4πγ ionization chamber. Recently, the demand for measuring activity of radionuclides that are not yet routinely used has been increasing. Experimental determination of the calibration coefficients is not always economical, especially for rarely measured radionuclides. Therefore, it is useful to calculate the coefficients from the knowledge of their decay scheme. Even though this is also achievable through the traditional methods, a Monte Carlo model of the chamber is more universal. Monte Carlo approach enables to predict a suitable calibration coefficients or correction factors for different containers and volumes of the sample as well, which can be very helpful, as the reference conditions are in practice rarely met.

To start with, a model of the CMI's 4πγ ionization chamber, acting as a national secondary standard of activity, was created. Then one of the frequently used radionuclide calibrators in nuclear medicine departments in the Czech Republic, trade name Bqmeter (manufactured by Consortium BQM, 36% of all devices used in the Czech Republic in 2001), was modeled. Both models have been accomplished using MCNP4C code [1]. First, basic characteristics of the chambers, such as the response function, sample position dependence and sample volume dependence were calculated and the results were compared with measured data.

2 MONTE CARLO MODEL OF THE CHAMBERS:

2.1 Geometry arrangement

2.1.1 CMI 4πγ ionization chamber

The CMI 4πγ ionization chamber is a well-type high-pressure ionization chamber, with steel walls and aluminium electrodes. The chamber is filled with argon to a pressure of 1.4 MPa. The active volume of the chamber is formed by two concentric cylinders of 19.60 cm and 10.04 cm diameters and 49.40 cm in height. The chamber is shielded by a lead 10 cm thick. The experimentally determined reference point for the measurements lies at the cylinder axis, 24.7 cm above the active volume base. The dimensions for the model were taken from design drawings.

2.1.2 Bqmeter ionization chamber

The Bqmeter is one of the commercially available chambers for activity measurements in nuclear medicine departments in the Czech Republic. The construction materials of this chamber

a) b)

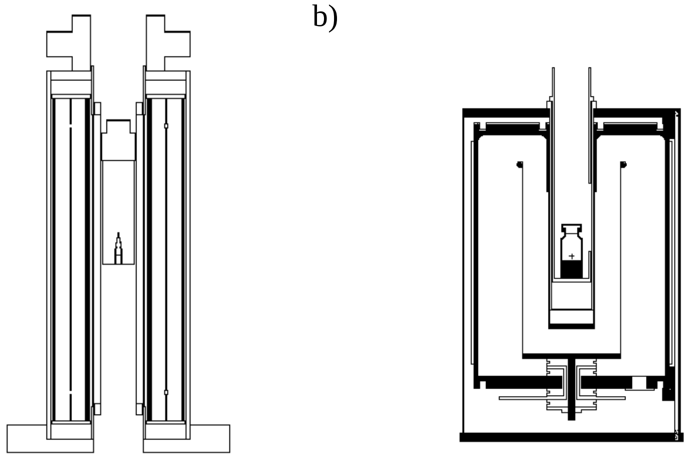

Figure 1. Geometry settings of a) CMI 4πγ ionization chamber with an ampoule and b) Bqmeter ionization chamber with a vial

are similar to the materials of the chamber described above. The chamber is filled with argon to a pressure of 0.35 MPa. The active volume of the chamber is formed by two concentric cylinders of 4.5 cm and 20.2 cm diameters and height 21.2 cm and 27.4 cm, respectively. The reference point for the measurements lies at the cylinder axis, 4.78 cm above bottom of the inner cylinder shaping the active volume. The model was created according to design drawings.

2.1.3 Containers

Two types of sodium glass containers, a 1 ml ampoule and a 10 ml vial, are used for calibration in CMI, while vials are used in nuclear medicine applications only. Reference volumes of the solutions in these containers are 1 ml and 5 ml, respectively.

2.2 Settings

2.2.1 Materials

Compositions of construction materials and their densities were taken from the tables [2]. The chemical compositions of the modeled radioactive samples correspond with the real ones. Where hypothetical monoenergetic sources were used, e.g. in computation of the response function, the material of the sample was assumed to be water.

2.2.2 Source

The source in the calculation model is a cylinder of height according to the container reference volume. For calculations of response as a function of the sample volume the cylinder height was changing with the volume.

The starting energy of the particles (photons) for each radionuclide was inserted into MCNP input file as a discrete distribution on the basis of the TORI data [3], except for hypothetical monoenergetic sources. All the particles start with a default statistical weight and directions uniformly distributed to 4π.

2.2.3 Parameters of the calculation

The detailed photon physics treatment was set up for the whole energy range. Coherent scattering occurs and all photon collisions can create electrons. TTB (Thick Target Brehmstrahlung) approximation for electron handling was used. The lower energy cutoff was set to 1 keV for both photons and electrons.

For photon interactions cross sections ZAID=ZZZ000.02P library supplied with MCNP code was used. This library uses ENDF data [4] and data of Storm and Israel [5]. Similarly, the electron interaction data tables are taken from ZZZ000.03e, which is derived from the ITS3.0 code system [6].

The number of histories was not set in advance: instead, the computation time was regulated to obtain results with a statistical uncertainty below 0.05%.

2.3 Calculations

For both ionization chambers and both standard containers, response function, responses for selected radionuclides, changes in the response with increasing volume of the source, and influence of a source displacement on the response were calculated.

The ionization current, *I*, produced by a source emitting *N* photons of a given energy per second was calculated by formula:

$$\mathbf{I} = \frac{\mathbf{E_d Ne}}{\mathbf{W_i}} \qquad (1)$$

where E_d is the mean energy deposited in the gas of the chamber active volume per emission of a photon, *e* is the electron charge and W_i is the average energy for creating an ion pair in argon.

The mean energy deposition (averaged over a cell) E_d was determined using the MCNP4C code, tally 6.

2.4 Average energy for creating an ion pair in argon

The numerators for the equation (1), calculated for 30 radionuclides measured in CMI 4πγ ionization chamber, were plotted against the experimentally determined chamber responses. By the use of the least square method, the most suitable value for W_i was determined. This was done for both the ampoule and vial geometry. The arithmetic average of the two obtained values yields W_i=25.94eV, which lies within the experimental values (23.8-26.4) eV cited for argon in [7].

3 RESULTS:

3.1 Response Function

When creating the model of the chamber, it is useful to study a response function of the chamber over the whole range of possible photon energies first. Its knowledge enables to identify the minimum/threshold energy necessary for a photon to contribute significantly to the chamber response. Then it is possible, when setting the radionuclide spectra into the input file, to omit photons below this energy threshold and thus simplify and speed up the calculation. The threshold energy is lower for ampoule geometry thanks to the lower self-absorption of smaller reference volume and thinner container walls.

3.1.1 CMI 4πγ ionization chamber

Response functions for the vial and ampoule geometry were calculated in steps of 20 keV up to 100 keV and in steps of 50 keV between 100 keV and 2 MeV. The calculated values were compared with the measured ones for several radionuclides emitting monoenergetic photons (^{109}Cd 88.04 keV, ^{99m}Tc 140.51 keV, ^{111}In 209.09 keV, ^{51}Cr 320.08 keV, ^{7}Be 477.59 keV, ^{137}Cs 661.65 keV, ^{54}Mn 834.83 keV and ^{60}Co 1252.88 keV), see Fig. 1. The shapes of the response functions for both containers are very similar. The threshold energy is 50keV for the vial and 45keV for the ampoule geometry.

There are more than 50 experimentally determined calibration coefficients for 4πγ ionization chamber. Nevertheless, the calculation of the chamber response was performed for the 30 radionuclides emitting gamma rays only, or having beta transitions with a maximum energy below 400 keV. Table 1 presents a comparison of the experimental and calculated calibration coefficients for both standard containers.

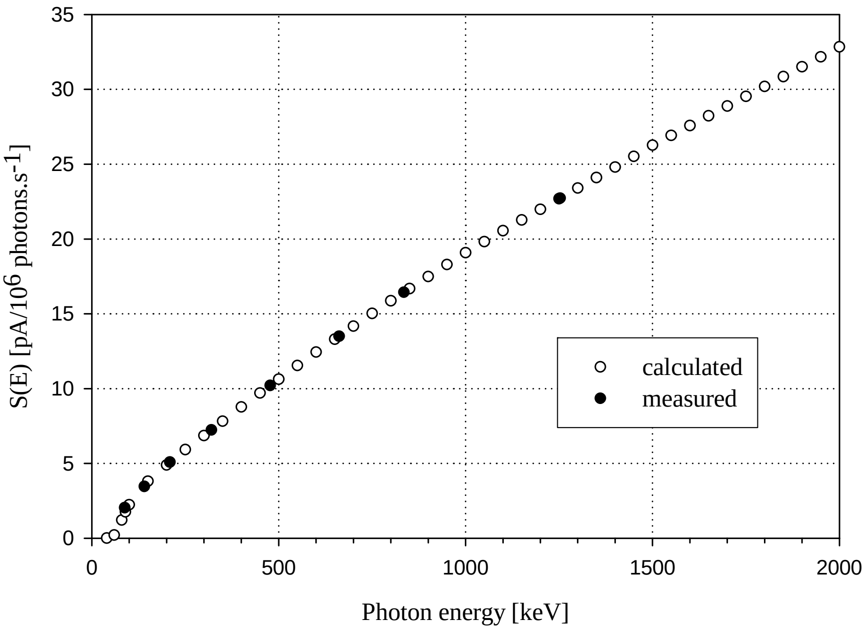

Figure 2. CMI 4πγ ionization chamber: The response function S(E) versus photon energy E[keV] for vial geometry.

Table I. CMI 4πγ ionization chamber: Relative deviations of the experimentally determined and calculated calibration factors for some radionuclide solutions

Deviation [%]	Radionuclide	
	ampoule	vial
0.0-0.5	^{22}Na, ^{51}Cr, ^{54}Mn, ^{56}Co, ^{58}Co, ^{82}Br, ^{85}Sr, ^{134}Cs, ^{137}Cs	^{46}Sc, ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{85}Sr, ^{133}Ba, ^{134}Cs, ^{137}Cs, ^{203}Hg
0.6-1.0	^{59}Fe, ^{60}Co, ^{95}Nb, ^{133}Ba, ^{203}Hg	^{22}Na, ^{65}Zn, ^{88}Y, ^{58}Co, ^{113}Sn
1.1-2.0	^{46}Sc, ^{65}Zn, ^{88}Y, ^{111}In	^{56}Co, ^{82}Br, ^{95}Nb, ^{111}In, ^{139}Ce
2.1-3.0	^{113}Sn	^{75}Se, ^{99m}Tc
3.1-4.0	^{7}Be, ^{75}Se, ^{99m}Tc, ^{123}I, ^{139}Ce	^{7}Be, ^{24}Na, ^{141}Ce
4.1-5.0	^{24}Na, ^{64}Cu, ^{103}Ru, ^{131}I	---
5.1-6.0	---	^{64}Cu, ^{103}Ru, ^{123}I, ^{131}I
6.1-7.0	^{67}Ga, ^{141}Ce	^{67}Ga

3.1.2 Bqmeter ionization chamber

Response functions for both ampoule and vial geometry were calculated in steps of 10 keV up to 100 keV and in steps of 50 keV between 100 keV and 1 MeV. The calculated values were compared with the measured ones for some radionuclides emitting monoenergetic photons (^{99m}Tc 140.51 keV, ^{51}Cr 320.08 keV and ^{137}Cs 661.65 keV); photons in narrow energy interval (^{125}I, mean energy 28.37 keV) and annihilation photons (^{18}F 511 keV) see Fig. 2. The threshold energies are 20 keV and 15 keV for the vial and ampoule geometries, respectively. The function forms a peak with the maximum at 50 keV and 45 keV for a vial and ampoule, respectively. Bqmeter ionization chamber is more sensitive to photons of lower energies than 4πγ ionization chamber, as its construction is less massive.

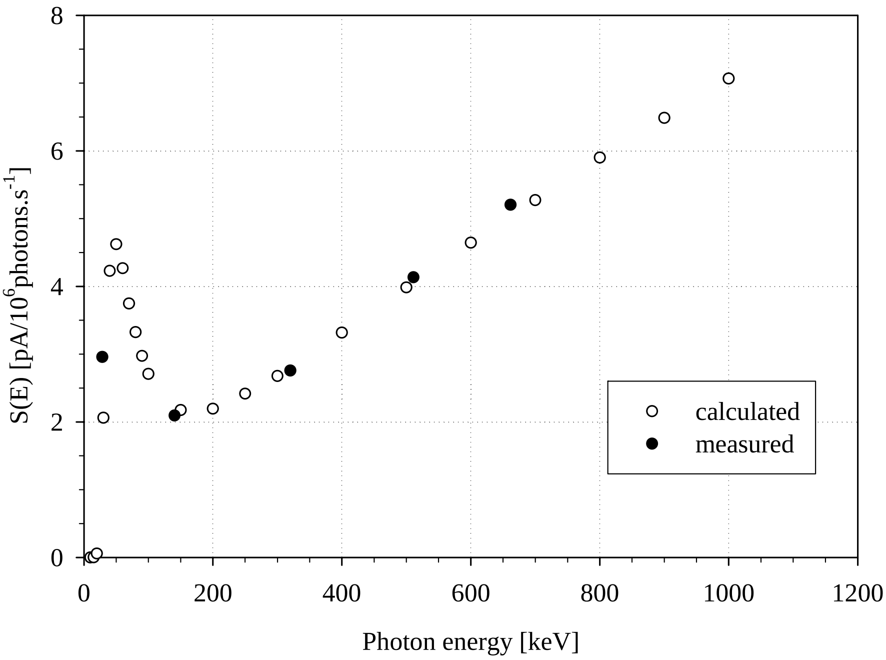

Figure 3. Bqmeter ionization chamber: The response function S(E) versus photon energy E[keV] for a vial geometry.

Table II. Bqmeter ionization chamber: Relative deviations of the experimentally determined and calculated calibration factors for some radionuclide solutions

Deviation [%]	Radionuclide	
	ampoule	vial
0.0-1.0	---	^{51}Cr, ^{57}Co, ^{99m}Tc, ^{153}Sm
1.1-2.0	^{57}Co, ^{131}I, ^{137}Cs	^{18}F, ^{137}Cs
2.1-4.0	---	^{131}I, ^{111}In
4.1-6.0	^{54}Mn, ^{241}Am	^{67}Ga, ^{123}I

Because this device is used at nuclear medicine departments only, the manufacturer provides calibration coefficients for 16 medically relevant radionuclides in vial geometry. However, the chamber response was calculated for only those radionuclides without significant beta component. Furthermore, calibration coefficients for 5 radionuclides in the ampoule geometry were determined experimentally and compared with the values predicted from the calculations. Table II. shows the discrepancies between calculated and experimental calibration coefficients.

3.2 Dependence on Sample Volume

Next, the changes in chamber response with different volume of the sample in the container were studied. All the results were normalized to the reference volume (1 ml in the ampoule, 5 ml in the vial). The calculated dependence of the chamber response on the sample volume of the same activity is presented for a vial only.

3.2.1 CMI 4πγ ionization chamber

Volume dependence of the chamber response was studied for ^{99m}Tc only, see Fig. 4. The effect is less than 1.5% over the whole vial capacity range.

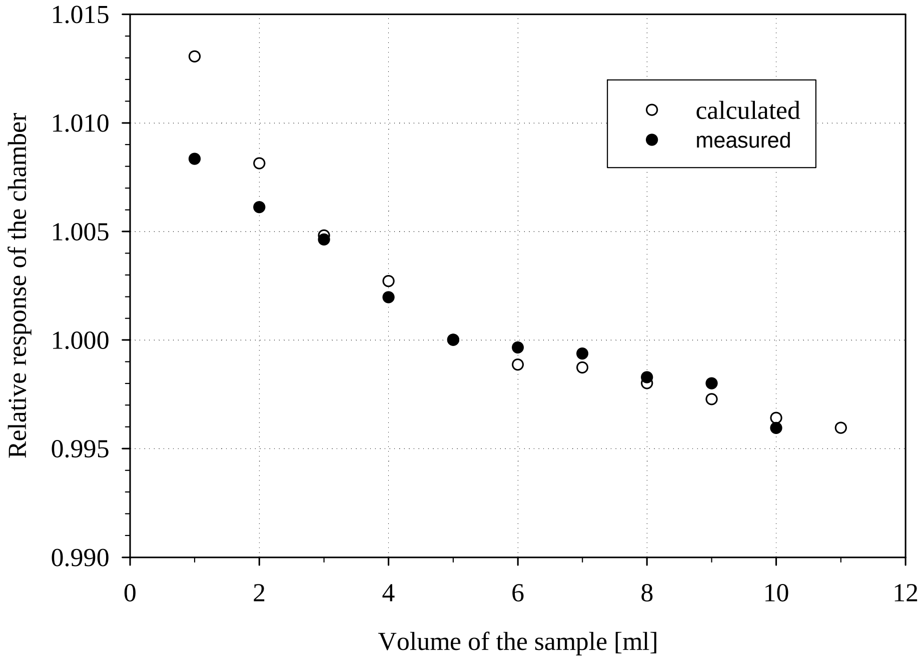

Figure 4. 4πγ ionization chamber: Relative response for 99mTc a vial as a function of the solution volume, normalized to 5 ml.

The volume dependences of the chambers' responses differ; compare Fig. 4 and Fig. 5. The relative response of the 4πγ ionization chamber is higher for smaller sample volumes due to the lower sample self-absorption, while solid angle remains nearly the same. The volume dependence is reverse for Bqmeter ionization chamber, due to its different construction. Here, the effect of higher self-absorption is predominated in bigger sample volumes by a significant increase of the solid angle.

3.2.2 Bqmeter ionization chamber

While in 4πγ ionization chamber the samples with reference volume are studied only, in nuclear medicine departments the activity of different volumes of the radioactive samples is often measured. Therefore, the volume dependence of the chamber response is more important for Bqmeter chamber and was studied in more detail.

Both the calculations and measurements performed for the three chosen radionuclides reflected well the different sensitivity of the chamber response to the sample volume, depending on the energy of the photons emitted by the radionuclide. Consequently, while the effect of having non-reference sample volume on the chamber response is less than 1% over the whole vial capacity for ^{99m}Tc solution, in case of ^{125}I, with considerably lower photon energies, the discrepancy can reach as much as 15%, see Fig. 5.

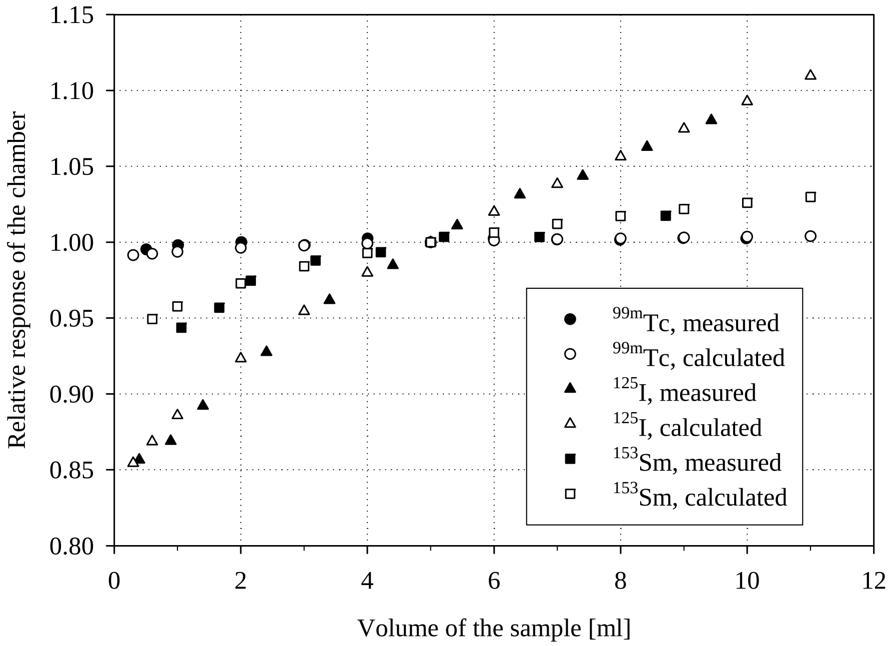

Figure 5. Bqmeter ionization chamber: Relative response for ^{99m}Tc, ^{125}I and ^{153}Sm in a vial as a function of the solution volume, normalized to 5 ml.

3.3 Dependence on the displacement of the source

Another factor influencing the correct response of the chamber is the displacement of the source from the reference position of the chamber. It can be either vertical (axial), when the source is moved along the chamber axis, or horizontal (radial), when the source is moved along the chamber radius. This feature was studied for both standard containers with reference volume of the sample.

The results for vertical dependence are available for the Bqmeter chamber only, since the calculated dependences for 4πγ chamber could not be compared with measured ones due to the technical difficulties. However, from the performed calculations it appears that the courses of the dependencies are similar to those presented for Bqmeter.

In order to demonstrate a different horizontal dependence for higher and lower photon energies, the chamber response to ^{241}Am (59.54 keV), ^{57}Co (122.06 keV, 136.47 keV) and ^{137}Cs (661.65 keV) was studied. Dependence of the relative responses on the radial displacement agrees with the results of Gostely and Laedermann [8]. For lower photon energies relative response rapidly decreases with distance from the reference position due to the increase of the effective absorber thickness, while for high energies this effect is suppressed by an increase of the solid angle. As a result, the relative response slightly increases with increasing distance from the centre.

3.3.1 CMI 4πγ ionization chamber

The dependence of chamber response on the horizontal distance of the sample from the reference position for ^{137}Cs and ^{57}Co in an ampoule is shown in Fig. 6. The results are normalized to the response obtained with sample in the chamber reference position.

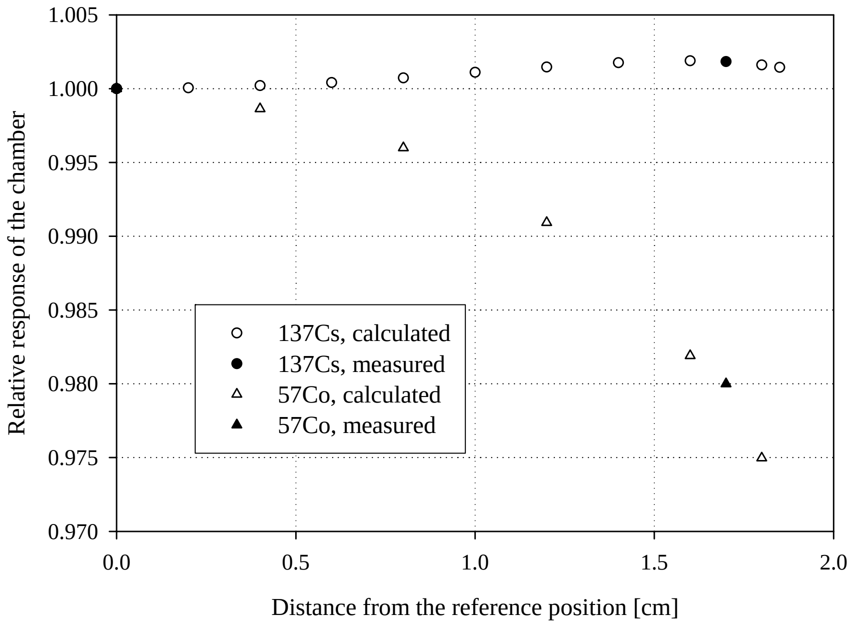

Figure 6. 4πγ ionization chamber: Relative response for ^{137}Cs and ^{57}Co in an ampoule as a function of the horizontal (radial) source displacement.

3.3.2 Bqmeter ionization chamber

Effect of horizontal displacement on the chamber response is less pronounced for Bqmeter than for 4πγ chamber, due to the thinner inner walls of the chamber. The dependence is presented in Fig. 7 for ^{241}Am, ^{57}Co and ^{137}Cs in an ampoule. The results for a vial geometry are similar.

The vertical source displacement was studied for ^{137}Cs and ^{99m}Tc, see Fig. 9. As the Bqmeter chamber is lower than 4πγ chamber, the vertical dependence is steeper. On the other hand, the construction of the chamber forces the user to measure in vertical direction in the reference position only.

All the results obtained from calculations were normalized to the chamber response for sample positioned in the reference position.

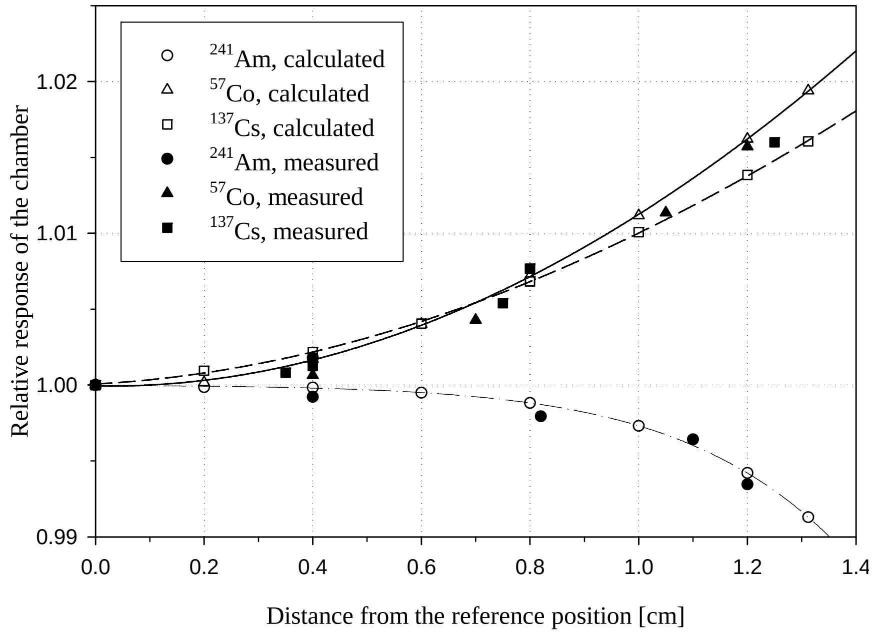

Figure 8. Bqmeter ionization chamber: Relative response for ^{57}Co, ^{137}Cs and ^{241}Am in an ampoule as a function of the radial source displacement.

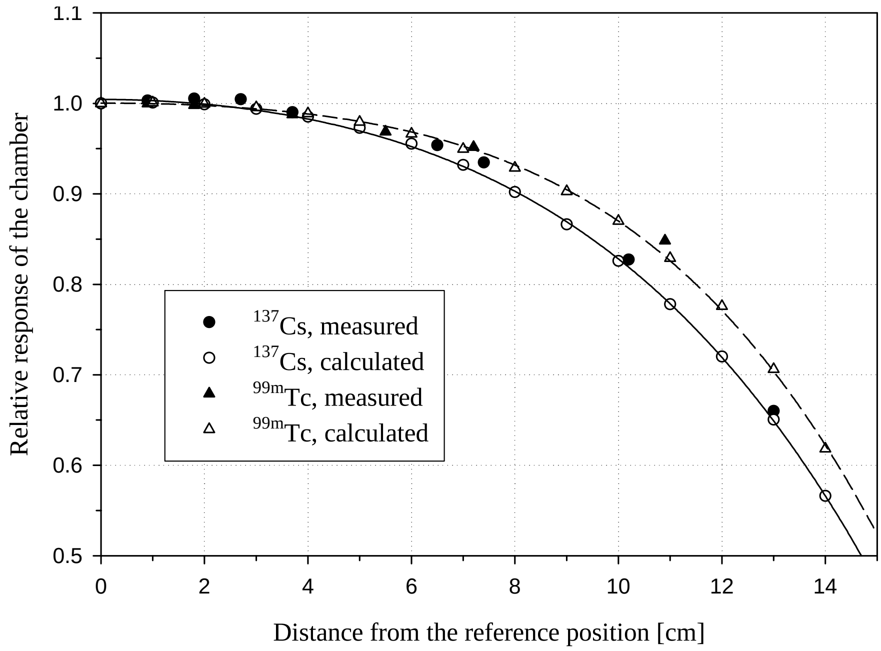

Figure 9. Bqmeter ionization chamber: Relative response for ^{137}Cs and ^{99m}Tc in a vial as a function of the vertical source displacement.

4 CONCLUSIONS:

Basic characteristics of the CMI 4πγ ionization chamber and the Bqmeter chamber were calculated and compared with measurements for two basic sample containers: vial and ampoule. The response function, sample volume and vertical dependences are presented for both chambers, together with comparison of calculated and experimentally determined calibration coefficients. The horizontal dependence is given for Bqmeter chamber only, due to the technical difficulties with 4πγ ionization chamber. The study of this parameter should be accomplished soon.

In the future, syringe geometry should be studied, since activity of the sample is measured prior to the application to the patient in various syringes at nuclear medicine departments. It would be helpful to examine the necessity of usage of geometry factors and determine their values, if needed. The performed studies of the activimeters confirm the achievability of this objective with acceptable uncertainties.

5 ACKNOWLEDGMENTS:

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