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Calculation of HPGe efficiency for environmental samples: comparison of EFFTRAN and GEANT4



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ABSTRACT

Determination of full energy peak efficiency is one of the most important tasks that have to be performed before gamma spectrometry of environmental samples. Many methods, including measurement of specific reference materials, Monte Carlo simulations, efficiency transfer and semi empirical calculations, were developed in order to complete this task.

Monte Carlo simulation, based on GEANT4 simulation package and EFFTRAN efficiency transfer software are applied for the efficiency calibration of three detectors, readily used in the Environment and Radiation Protection Laboratory of Institute for Nuclear Sciences Vinca, for measurement of environmental samples. Efficiencies were calculated for water, soil and aerosol samples. The aim of this paper is to perform efficiency calculations for HPGe detectors using both GEANT4 simulation and EFFTRAN efficiency transfer software and to compare obtained results with the experimental results. This comparison should show how the two methods agree with experimentally obtained efficiencies of our measurement system and in which part of the spectrum do the discrepancies appear. The detailed knowledge of accuracy and precision of both methods should enable us to choose an appropriate method for each situation that is presented in our and other laboratories on a daily basis.

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1. Introduction

One of the main problems in quantitative gamma-ray spectroscopy is the determination of detection efficiency, for different energies, different source-detector geometries and different composition of voluminous sources. In most cases the straightforward calibration methods are used, a number of semi empirical methods have also been developed, and finally the Monte Carlo simulations are occasionally used to generate the spectra and extract the desired efficiency [1]. This last method which generates the response function of the spectrometer, however, is unique when the deconvolution of continuous spectra is an object. Significant advance in this respect would represent the announced wide availability of response function of large Ge detectors for some standard source-detector geometries [2]. For non-standard cases one has to resort to some of the general simulation programs. The best known and the most sophisticated one is the CERN GEANT4

package developed to simulate the workings of complex particle detectors, which, when used for Ge detectors, needs adjustments for low energies and requires large computing resources, both in speed and memory [1].

Efficiency transfer has been a popular method of calculating full-energy- peak efficiencies of a sample of interest on the basis of an experimental efficiency curve measured on the same detector, but with a calibration source of different size, geometry, density or composition. The procedure saves time and resources, since sample-specific experimental calibration is avoided. It has proven especially useful in environmental measurements [3], where on one hand, an ultimate precision in calibration is usually not required and on the other, a variety of different sources might be measured by a laboratory carrying out a monitoring program around a nuclear power plant, for example [4].

The aim of this paper is to perform efficiency calculations for HPGe detectors using both GEANT4 simulation and EFFTRAN efficiency transfer software and to compare obtained results with the experimental results. This comparison should show how the two methods agree with experimentally obtained efficiencies of our measurement system and in which part of the spectrum do the discrepancies appear. The detailed knowledge of accuracy and

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precision of both methods should enable us to choose an appropriate method for each situation that is presented in our and other laboratories on a daily basis.

2. Materials and methods

Experimental efficiency calibration is readily performed in our laboratory. For the purpose of calibrating a measurement system for water, soil-like and aerosol samples, a set of secondary reference materials was produced by spiking the matrix with certified radioactive mixture solution ER X 9031-OL-426/12 issued by Czech Metrological Institute, Inspectorate for Ionizing Radiation. The radioactive solution contained following radionuclides: ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{203}Hg , ^{113}Sn , ^{85}Sr and ^{88}Y , with the energies that span from 59 to 1898 keV with total activity of 1342 Bq at reference date 31.08.2012. The content of the mixture was chosen in such a manner that, for the most part, excludes coincidence summing effects. The preparation of the secondary reference material was conducted as described in [5].

The secondary reference material with water matrix was placed in two polystyrene cylindrical bottles of 125 and 250 ml, while the spiked soil was placed in two polystyrene cylindrical containers of 100 and 200 ml. For the aerosol matrix, aerosol dust, collected using F&J Specialty Product constant air-flow sampler, was spiked and placed into a vial. The measuring time for all calibration sources was 60,000 s. The geometry of the detectors was defined as specified by the manufacturer, while the bulletization and depth and diameter of the central cavity were estimated based on the known dimensions of other detectors produced by the same manufacturer. The geometry of the sample was defined by the diameter of the container and sample filling height. The sample matrix was defined by its chemical formula and density. The same parameters were used for simulations and calculations.

Monte Carlo simulation, based on GEANT4 simulation package [6], has been developed to obtain the response of germanium detectors, with the aim to reproduce experimental spectra of detectors in wide range of applications in gamma spectroscopy measurements [7,8]. GEANT4 allows the description of an experimental setup represented by a structure of geometrical volumes filled by given materials and associated with tracking media. In the simulation, germanium detectors, together with the whole detector assembly, are constructed in great detail, according to manufacturer's data specifications. Sample dimensions are also measured with care, and sample volumes are constructed in the simulation code accordingly. Primary photons are generated in the sample volumes with uniformly random positions and momentum directions in full space (4π sr). Each photon undergoes interaction processes and deposits energy in the detector or it traverses the detector (or the whole system) without interaction. All types of relevant interactions of photons and electrons/positrons with matter are taken into account, using low-energy data packages

(dataset G4EMLOW6.32, model G4EmLivermorePhysics); tracking of the particles is performed down to below 10 keV. Distribution of photon energy deposited in the detector's active volume gives spectral response of the detector, i.e. simulated spectrum. From simulated spectrum one can obtain simulated full peak intensities, and from this, derive simulated detector efficiency for a given energy.

Many software packages were developed in order to perform efficiency transfer calculations and coincidence summing corrections with known set of parameters. One of such software is EFFTRAN, developed in Belgian Nuclear Research Centre in Mol. The efficiency transfer calculation in EFFTRAN is based on calculating a ratio of the efficiencies for the sample of interest and for the calibration sample and multiplying it with the measured efficiency of the calibration sample. A great advantage of the method, as pointed out and verified by its founders, is that in the calculated ratio, many inaccuracies in the detector model can be expected to cancel out to a large degree, making it possible to work directly with non-optimized detector data supplied by the manufacturer. The cancellation of differences also applies to the interaction cross-section data that different implementations of this method may use and to their physical models of particle interaction and tracking [4,9]. On the other hand, in order to calculate the unknown efficiency for cylindrical sample starting from known efficiency of the point source, program requires relatively precise information on geometry of the detector (crystal geometry, housing geometry and composition, active and inactive layers etc.). This data is not always known and cannot be precisely defined, thus contributing to the measurement uncertainty and that contribution has to be estimated as accurately as possible. However, the lack of precision and uncertainties rising from poor knowledge of the composition of the sample cannot be avoided.

In this paper, all three methods of efficiency calibration were applied. The detectors used for measurement, as well as for simulation and calculation, were the ones used in our laboratory for measuring the environmental samples. These are two p-type detectors with relative efficiencies of 18% (named Detector 1) and 50% (named Detector 3) and one n-type detector with relative efficiency of 20% (named Detector 2). The characteristics of the detectors are presented in Table 1. In order to perform the simulations, the detector was modeled taking into account the technical features obtained from the manufacturer, except for the central void, whose size parameters were estimated.

3. Results and discussion

For the purpose of calibrating our measurement system for environmental samples, a set of 5 secondary reference materials was produced, as explained in Section 2 and measured for 60,000 s in contact geometry. The full energy peak efficiency was calculated

Table 1
Detector characteristics, as provided by the manufacturer.

	Detector 1	Detector 2	Detector 3
Geometry and type of detector	Closed coaxial - p type Canberra	Closed reverzibile coaxial - n type Canberra	Closed coaxial - p type Canberra
Relative efficiency	20 % (certificate)	18 % (certificate)	50 % (certificate)
Resolution	0.850 keV (certificate)	0.759 keV (experimental)	1.00 keV (experimental)
on 122 keV	1.8 keV (certificate)	1.69 keV (experimental)	1.9 keV (experimental)
on 1332 keV	51:1 (certificate)	56.1:1 (experimental)	65:1 (certificate)
Peak/compton ratio	49.5 mm	48 mm	65 mm
Crystal diameter	56.5 mm	48.5 mm	67 mm
Crystal length	5.5 mm	5 mm	5 mm
Crystal to window distance	Al	Be	Be
Entry window	(+) 4500V	(-) 4000V	(+) 3000 V
Working voltage			

according to the equation:

$$\varepsilon = \frac{N}{t \cdot P_\gamma \cdot A} \quad (1)$$

where N represents the net count, t is counting time, P_γ is emission probability and A is source activity on the given energy, with the decay correction. Quantity N was not corrected for dead time losses, since the duration of the measurement was set on live time.

Combined measurement uncertainty for the experimental values was calculated according to the following equation:

$$u(\varepsilon) = \sqrt{(\delta A)^2 + (\delta N)^2 + (\delta M)^2} \quad (2)$$

where δA represents relative uncertainty of the radioactive solution given by the manufacturer, δN is the relative counting uncertainty, δM is the uncertainty introduced in the process of production of the secondary reference material and $\delta A + \delta M$ is estimated to be approximately 2–3%. The uncertainties introduced via t and P_γ are negligible. Relative measurement uncertainty $u(\varepsilon)$ for all energies did not exceed 5% at 2σ level of confidence.

The same setting was used for GEANT4 simulation. The geometry of the detectors was defined as specified by the manufacturer, while the bulletization and depth and diameter of the central cavity were estimated based on the known dimensions of other detectors produced by the same manufacturer and [9]. The geometry of the sample was defined by the diameter of the container and sample filling height. The material of the sample was defined by its chemical formula and density.

In case of GEANT4 simulation, combined uncertainty was estimated according to following consideration. The main variables that have been input into the simulation are 9 characteristic dimensions of the detector (crystal diameter and length, crystal cavity diameter and length, top and side dead layer, end cap diameter, window thickness and window to crystal gap) and 4 characteristics related to the sample (sample volume, sample and container material, namely density and chemical composition, and container to absorber gap). For these variables, except chemical composition of sample and container, the uncertainty can be estimated to be 1% for geometry of crystal and container to absorber gap and 10% for window thickness and window to crystal gap. In order to minimize the discrepancy between simulated and measured values, bulletization, dead layer and window to crystal gap were varied in the simulation, and the uncertainty was lowered to estimated 1%. The chemical composition of the container is well defined, but for the sample (except water) the situation is more complicated and poor knowledge of the chemical composition of soil and aerosol can be the source of larger uncertainty. This is estimated to be 10%. Since the uncertainty of the simulation, $u_{simulation}$ is calculated according to

$$u_{simulation} = \sqrt{\sum (\delta x_i)^2} \quad (3)$$

where δx_i represents relative uncertainty of the value x_i , overall combined uncertainty of the simulated results is estimated to be 5.3% for soil and aerosol samples and 3% for water samples.

When using EFFTRAN for efficiency transfer calculation, one has to have a known experimental calibration curve as a starting point. The most usual approach is to have the calibration for source with geometry and composition similar to the one of interest. In worst case scenario, a laboratory possesses only point sources but needs measurements of different bulk samples. In this paper, we tried to make use of point sources and calculate efficiency for the cylindrical sources. For a total of 6 energies, measurement was conducted using point sources ^{241}Am , ^{109}Cd , ^{137}Cs , ^{54}Mn and ^{60}Co produced by LMRI Coffret d'etalons gamma ECGS-2, placed at the detector cap ($x=0$ cm). The count was used

to calculate experimental efficiency, also according to Eq. (1), for point source [5], as a starting point for efficiency transfer calculation with EFFTRAN. Combined uncertainty for EFFTRAN calculation was calculated according to Eq. (2) with the δM defined as an uncertainty arising from the definition of the detector geometry, which was estimated at 5%, and δA equal to 2.5% as defined by the manufacturer of the point sources. Overall uncertainty of the calculated results is hence estimated to be 7.5%

Since the secondary reference materials contain elements with energies other than those of measured point sources, efficiencies for those energies had to be calculated. For that purpose, a point source calibration curve was obtained by fitting the measured values using following fitting function [10]:

$$\ln \varepsilon = \sum_{i=1}^5 a_i (\ln E)^{i-1} \quad (4)$$

where ε is efficiency, E is energy given in keV and a_i are fitting coefficients. It is established that for our detectors, the best calibration curve is achieved by using $i=5$. Point source efficiencies obtained using this calibration curve, were then used as a starting point for calculations via EFFTRAN software. The information on the detector geometry as well as the composition and geometry of the samples were set to be the same as for GEANT4 simulation.

The efficiencies obtained using these three different methods are compared and their relative differences presented in Figs. 1–3.

Fig. 1 represents relative differences of the efficiency for water matrix placed in two different bottles for all three detectors (due to the housing geometry of Detector 3, only 125 ml bottle was investigated). As it can be seen, both GEANT4 and EFFTRAN produce results that are in relative good accordance with the experimental ones. Majority results are within a 10% margin that falls within the uncertainty limits. This is the consequence of water being well defined, low absorbing matrix with simple chemical composition, so the accordance between results is expected. It is however noticeable, that EFFTRAN produces slightly higher results for Detector 2, which is an n-type detector with Be window.

Fig. 2 depicts relative differences of efficiency for the soil matrix placed in 200 and 100 ml cylindrical geometry. Since the preparation of soil sample for production of secondary reference material included drying at 105 °C, it is assumed that most of the organic and volatile material was lost. According to this, soil matrix was defined as a composition of 90% SiO_2 , 1% Ca, 4% K, 1% Fe and 4% C. The composition greatly varies with the type of soil and this definition is only an approximation. Fig. 2 shows that in case of soil matrix in cylindrical container, GEANT4 gave relatively satisfactory results with the discrepancy of 2.6–10.8% with respect to experimental results for Detector 1. The same is valid for Detector 2, with the discrepancy of 5.5–11.8% and Detector 3 with the discrepancy of 0.6–10.5% with respect to experimental results. EFFTRAN produces results that differ from the experimental ones in the mid-energy region for all detectors. Also, for p-type detectors, efficiency for the high energy part of the spectrum shows the greatest discrepancy. The efficiency is mainly underestimated for the p-type, with the largest discrepancy for the highest investigated energy and overestimated for the n-type detector. This can be explained as a consequence of the lack of point sources that cover that region of spectrum. Thus the starting point for EFFTRAN calculation is somewhat incomplete, which can be resolved by measuring more point sources that produce energies in the desired region (^{85}Sr and ^{113}Sn , or ^{152}Eu for example), that are unavailable at present.

Similar can be said for aerosol matrix, presented in Fig. 3. In general, GEANT4 produces acceptable results which are within

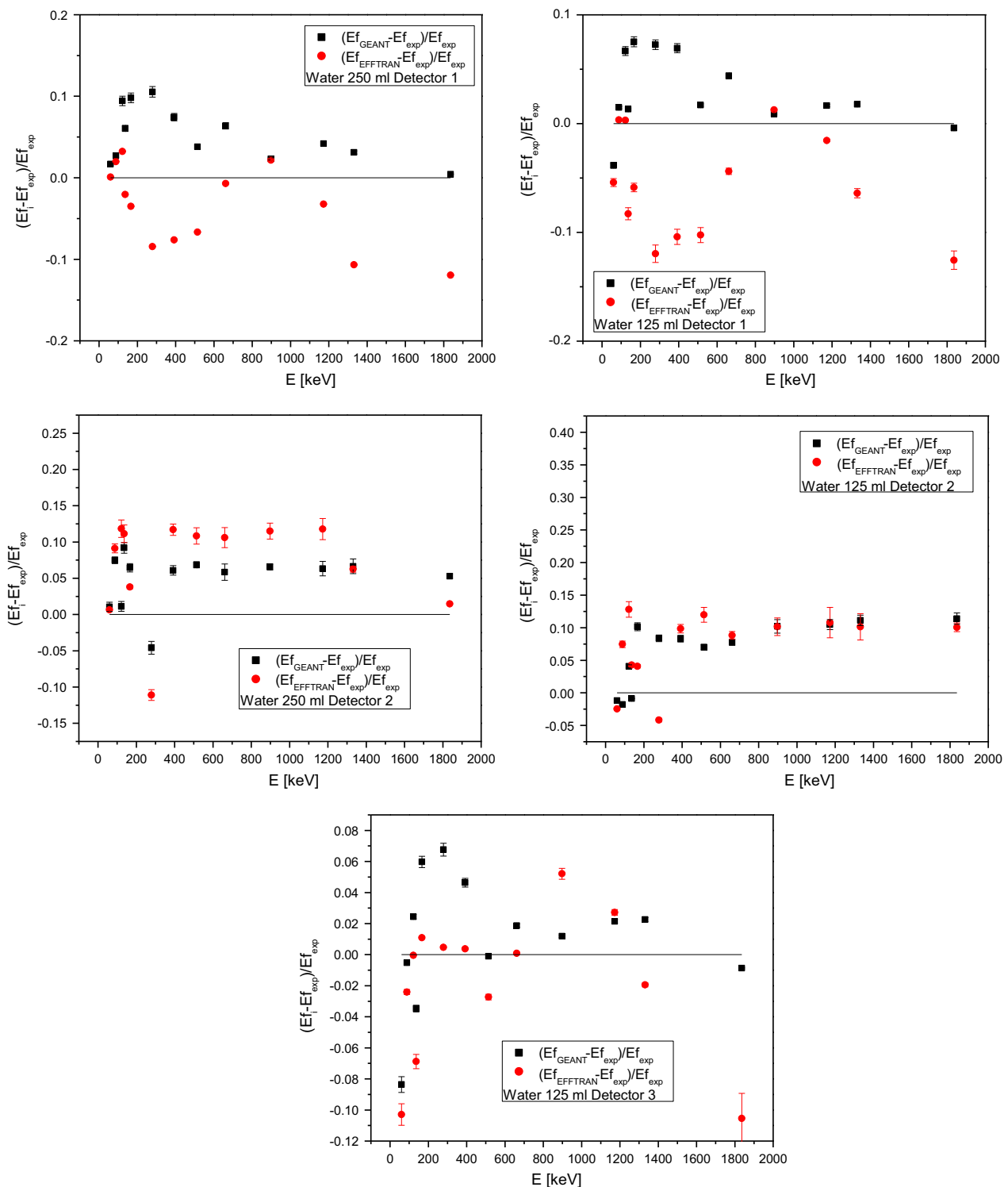


Fig. 1. Relative differences of efficiency obtained using GEANT4 and EFFTRAN with respect to experimental values for water matrix in 125 and 250 ml bottles. The squares represent points obtained using GEANT4 simulation and circles are points obtained using EFFTRAN.

$\pm 10\%$ of experimental results except for the low energy part. For Detector 1 and Detector 3, contrary to the soil matrix, simulated efficiencies are slightly overestimated. In case of Detector 2 the largest discrepancy is for the low part of the spectrum, where GEANT predicts efficiencies that are unacceptably lower than the experimental ones. It was noticed that the shape of the curve produced by GEANT is not in accordance with the experiment, since the slope is less steep than it should be. Since front dead layer thickness greatly influences the results in the part of

spectrum below 700 keV [11], this parameter was varied in order to achieve the best accordance with the experimental results. However, the discrepancy is still unacceptable, pointing that some other parameter (such as effective cross-section for the interaction) have to be revised. In case of EFFTRAN calculations, the discrepancies are similar in general and are mainly between 5 and 15%. In case of Detector 2, which has high efficiency in lower part of the spectrum, calculated efficiency curve follow the shape of the experimental curve. Contrary to GEANT, efficiency transfer

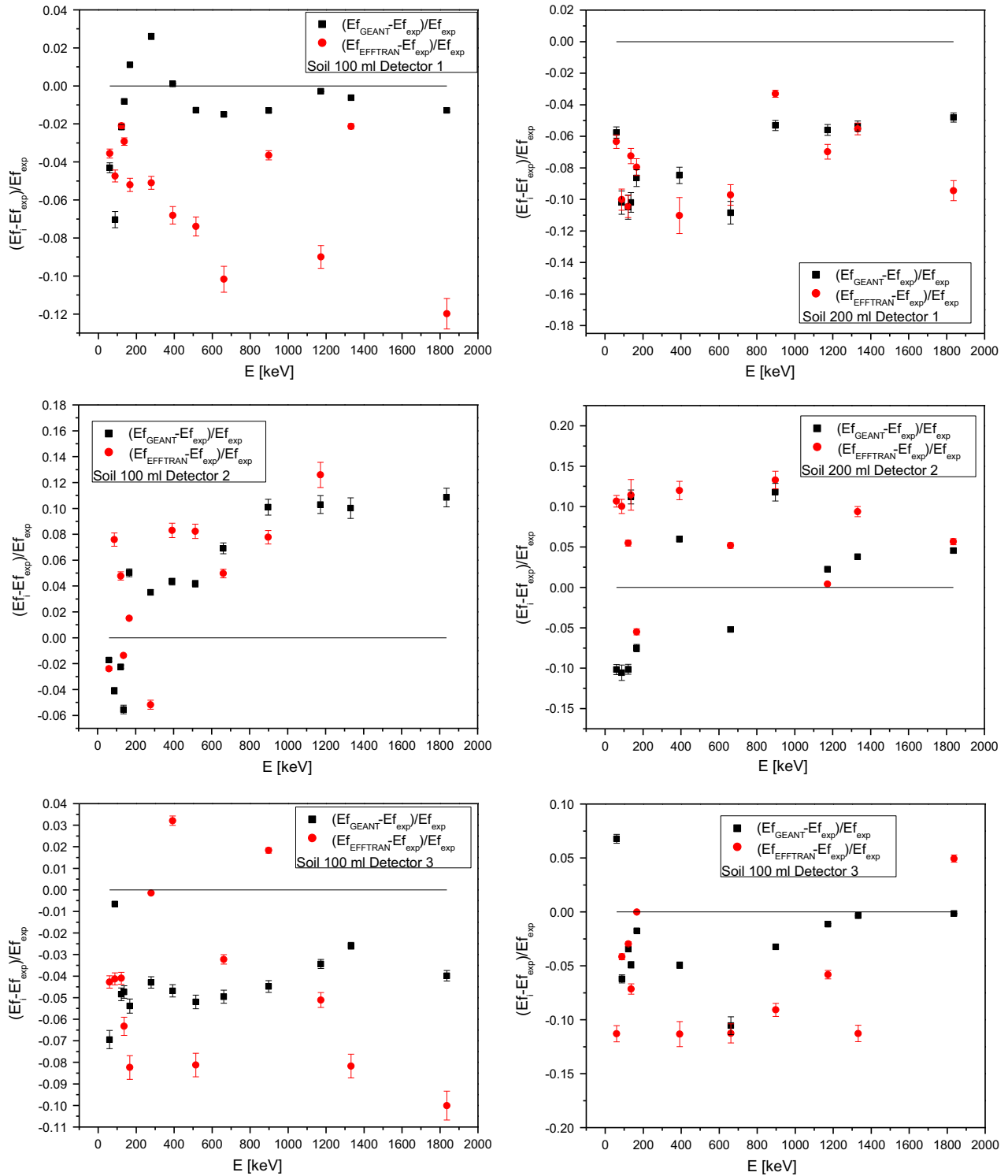


Fig. 2. Relative differences of efficiency obtained using GEANT4 and EFFTRAN with respect to experimental values for soil matrix in 100 and 200 ml cylindrical containers for all three detectors. The squares represent points obtained using GEANT4 simulation and circles are points obtained using EFFTRAN.

produced overestimated values for p-type and underestimated values for n-type detector.

Baring in mind that the calibration is performed with the aim to be used for environmental samples, discrepancy of 10% is acceptable, but it has to be included in the uncertainty budget for each routine measurement. This can be used as an indication of limitations for applicability of calculations or simulations. Namely,

both the GEANT4 simulation and the transfer of efficiency from point source to bulk source with water and aerosol matrix is acceptable in case of our measuring system. On the other hand, for n-type detector, GEANT simulation parameters need to be revised since some of the results fall out of the acceptable limits. In case of efficiency transfer for point source to bulk sample with large effective atomic number can produce large

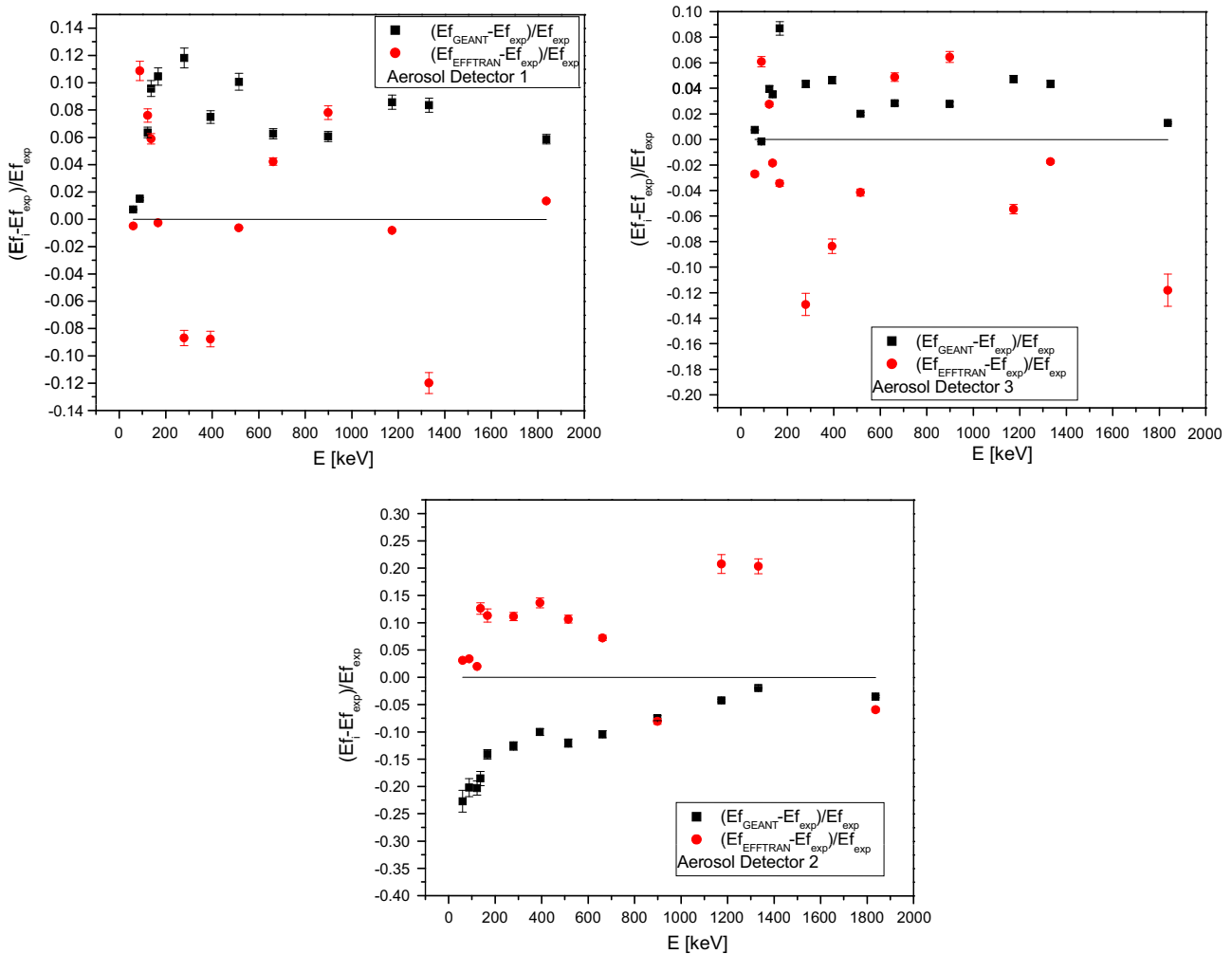


Fig. 3. Relative differences of efficiency obtained using GEANT4 and EFFTRAN with respect to experimental values for aerosol matrix for all three detectors. The squares represent points obtained using GEANT4 simulation and circles are points obtained using EFFTRAN.

discrepancies that span from -20 to $+20\%$. In such situation, efficiency transfer from similar geometry should produce better results. According to our investigation, efficiency transfer from 100 ml soil matrix to 200 ml soil matrix produces results that differ less than 10%. Also, by varying the geometrical characterization of the detectors, we can obtain better results using both GEANT4 and EFFTRAN [11–13].

Since the purpose of this calibration is to calibrate the measuring system for environmental samples, some comments can be made regarding the practical use of these methods, when applied to a realistic measurement. The GEANT4 simulation is capable of producing results with great accuracy, provided that all parameters of the measuring system and sample are available. This, in general, can be achieved in most of the cases. Furthermore, in all three types of samples investigated in this paper, GEANT4 produced satisfactory results in the mid range of the spectrum, which is of the interest when measuring samples that contain ^{214}Pb , ^{214}Bi , ^{232}Th (e.g. soil) or ^7Be (e.g. aerosols). However, when a lower part of spectrum is of interest (^{210}Pb , ^{238}U at 63 keV), and especially if n-type of detector is used, EFFTRAN produces results with greater accuracy. Also, it is recommendable that calibration sources with similar matrix as the real samples should be used for a starting point in efficiency transfer calculations. The other point is the processing time needed for each calculation. GEANT4 requires both time and proficiency in computer programming, which makes it

unsuitable for daily application, while EFFTRAN is easy to use and does not require large processor time, but lacks in precision.

4. Conclusion

This paper presents the results of calibration of three HPGe detectors using GEANT4 simulation and EFFTRAN software for efficiency transfer. The results were then compared with the experimental ones. For the purpose of experimental efficiency calibration, a set of 5 secondary reference materials was produced, two with water matrix, two with soil and two with aerosol matrix. For the EFFTRAN calculation, a set of point sources was measured in order to obtain a calibration curve needed for efficiency transfer. Both the GEANT4 simulation and the transfer of efficiency from point source to bulk source with water and aerosol matrix is acceptable in case of our measuring system. The main cause of the discrepancies can be attributed to the definition of detector geometry. GEANT4 produced most of the results with discrepancies smaller than 10%, while EFFTRAN, although the discrepancies were larger in respect to the experimental results, reproduced the shape of the curve better, especially for the n-type detector. However, since the starting point for EFFTRAN calculation was a set of point sources that did not cover the whole region of interest and taking into account that the geometry and composition of the measured samples were significantly different from point source, this method proved that it can be used with some

improvements regarding the setting of detector geometry and sample composition within the software.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.nima.2014.06.044>.

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