

EXPERIMENTAL CHARACTERIZATION OF AN *IN-SITU* SPECTRO-TRACER USED IN GEOPHYSICAL EXPLORATIONS

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Abstract. The selection of an appropriate detector for field measurements, in case of natural radioactivity, is an important task taking into account constraints given by data reliability, time and cost effectiveness. This paper provides the necessary data when choosing a portable spectrometer Gamma Surveyor based on BGO crystal (6.3 in³). The proven efficiency of the crystal, the weaker energy resolution but still good comparable results with HPGe results, obtained for ²³⁸U, ²³²Th, ⁴⁰K often makes it a good choice. Thus, in this paper some functional parameters, as energy calibration, energy resolution and background of the system are analyzed. In addition, field data, concentrations of radionuclides ²³⁸U, ²³²Th, ⁴⁰K (ppm) in surface soil on the Bucharest area are compared with laboratory measurements using HPGe detector. The obtained data, for all three radionuclides, are well correlated.

Key words: soil survey; geophysical techniques; gamma ray spectrometry; *in situ* measurements.

1. INTRODUCTION

Gamma spectroscopy is a very powerful technique, widespread used in applications others than nuclear physics, such environmental studies, geochronology, life sciences, industrial applications etc. For environmental studies this technique is based on two classes of the detector materials: scintillation detectors and semiconductors detectors. Different factors are important in the choice of a type of gamma detector: a) the resolving power of the peaks which determines the complexity of the spectrum; b) the detection efficiency which dictates the source strength; c) the simplicity of the arrangement and the ease of the data acquisition; d) the time and cost-effective techniques; and e) other factors such as the response linearity, the stability etc.

Taking into account the above factors, the choice of the spectrometer also depends on the applications and requirements. It is known that an HPGe spectrometer

has an excellent energy resolution, but often lack in sensitivity and always requires low operating temperatures. Instead, a scintillator detector excels in sensitivity, but lower in energy resolution. However, the latter is easy to handle, operate at room temperature and can be manufactured in a wide range of sizes and shapes that make it suitable for environmental studies [1, 2] especially on the field regarding radiological mapping campaigns and others [3].

The use of the scintillator detector for *in-situ* measurements has been well proven by [1] where large size BGO detector is used for detection of radioactive particles found at depths down to 10 cm in the ground or NaI [2] and CsI specially design for airborne gamma-ray survey [3]. As was mentioned the choice of a detector type and size depends of how much in depth this shall detect. If only the surface layer is under consideration *in-situ* surveys can also be conducted using hand-held portable spectrometers. These spectrometers have the advantages of being very flexible regarding the collection of the data, spatial resolution, sampling time and survey design. In addition, where anomalies or large variations of radiometric measurements occur, samples can also be collected during survey. Variations of natural emitted gamma radiation have been used in geological prospecting for over 20 years to detect variation of soil material [4]. Subsequently, *in-situ* gamma results can be compared with laboratory measurements. However, a comprehensive characterization of these portable spectrometers a must be well performed, so that the interpretation of the results will be correct.

In this paper an *in-situ* survey tool, used for determination of ^{40}K , ^{238}U , ^{232}Th concentrations and of natural gamma dose rate in the environmental monitoring, geological mapping and geophysical methods combined with seismological studies [5, 6], is characterized and its results are compared with laboratory measurements. The system must be well evaluated for the accuracy and precision of its data in order to have confidence in the results. The results showed good correlation for all three radionuclides.

The paper describes the calibration of a commercially available portable spectrometer and comparative measurements to laboratory based, HPGe gamma spectrometry results. The novelty consists in the validation of *in-situ* measurements for the portable spectrometer. This paper it is important, because there are not so many data in the literature about this viable alternative for *in situ* measurements and also highlights the cost-effective way to obtain concentrations of natural radionuclides in soil for geophysical studies.

2. MATERIALS AND METHODS

In this paper a survey tool used for geophysical exploration is characterized regarding its resolution, response to various radiation sources and assessment of the results. Gamma Surveyor II is a hand-held gamma portable spectrometer, commercially available from GF Instruments, Brno, Czech Republic. The detector consists of a

scintillator crystal of bismuth germanate ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) of 103 cm^3 volume. The entire associated electronic is embedded in a water proof assembly. Each measured data file has an identifying file number and is stored in the memory of Gamma Surveyor II [7]. The spectrometer allows connecting GPS receiver to register electronically the position of each measuring point in the field. It has an LCD display (128×64 pixels) which help the user to choose operation mode, measuring time and other settings, and internal memory – up to 100 files with 400,000 full spectra and assay results [7]. The spectral analyzer measures in 1024 channels that corresponds to 30 keV – 3 MeV energy range. The instrument determines ^{40}K [%], ^{238}U [ppm], ^{232}Th [ppm] concentrations and measures natural gamma dose rate [nSv/h]. The instrument is able to detect natural at the ground surface and boreholes covering wide range of applications like field, underwater, car-borne surveys.

The calibration of the spectrometer is performed by the manufacturer on high volume K, U, Th and background standards according to IAEA recommendation [7, 8]. The factory calibration is assuming a homogenous half space mode (2π) valid for usual field measurement. However, the user can perform and add its own calibration using a complete set of calibration pads (K, U, Th, background standards) with known K, U, Th concentrations. Nominal concentration values for individual standards must be multiplied by geometrical factor $G = 1 - h / r$, where h is the height of the middle of the detector above the pad and r is the diameter of the pad. The spectrometer undergoes fast auto-stabilization using natural radioactivity and stable calibration.

The accuracy of the measurements is influenced by measuring time. In this case, for field measurements, the relative error obtained must be judged individually for each measuring task and then the convenient measuring time must be chosen. For low natural gamma radiation, high measuring time is needed and *vice versa*. According to the manufacturer specification the relative error for low radioactivity measurements and a measuring time of only 3 min is 6% for ^{40}K , 30% for ^{238}U and 16% for ^{232}Th . These are also due to the fact that the region of interest (ROI) for ^{40}K is less influenced by the background and is well defined in the spectrum evaluation, since for the assessment of ^{238}U and ^{232}Th the ROIs are influenced by the background and peak interferences.

In the field, the usual mod of measurement is Spectrum & Assay that, a spectral measurement with determination of concentrations of K, U, Th and of natural gamma dose rate. The system allows spectrum view and has integrated a 17 isotopes library and selectable measuring time from 10 s to 30 min. An assessment of the optimum measuring time was performed for 5 min, 10 min and 30 min at the same point. After evaluating the results and their associated errors but also taking into account time effectiveness for a field day measurement it was concluded that the optimum measuring time was 10 min.

To confirm the reliability results of the field measurements with Gamma Surveyor II for the natural radioactivity ^{40}K , ^{238}U and ^{232}Th soil samples were collected

to be analyzed in the laboratory on a HPGe detector. The laboratory gamma spectrometry system consists of coaxial p-type HPGe detector with a 35% relative efficiency and 1.73 keV energy resolution at 1332.5 keV (^{60}Co) connected with a digiDART (ORTEC) and is described elsewhere [9]. The samples are prepared and placed in plastic cylinders and measured for 86400 sec acquisition time.

3. RESULTS AND DISCUSSION

3.1. GAMMA SURVEYOR II CHARACTERIZATION

To characterize the Gamma Surveyor II spectrometer a set of quasi-point sources, such as ^{60}Co , ^{133}Ba , ^{137}Cs , ^{152}Eu has been used to verify the energy calibration against the one given by the manufacturer. From the differential pulse height distributions representing the response of the spectrometer to the energy lines of the sources, the energy calibration was performed and the energy resolution of the detectors was determined, by calculating the full width at half maximum (FWHM), which indicates its capability to separate close energy lines. The energy calibration curve is described by the following linear function: energy [keV] = $a_0 + a_1 \times \text{channel}$, where $a = 6.40 \pm 16.12$ and $b = 2.95 \pm 0.05$ are the coefficients of the linear fit curve shown in Fig. 1. Table 1 presents the isotopes used with the prominent peaks in the spectrum and corresponding centroid channel. The response of spectrometer to the ^{133}Ba source is presented in Fig. 2, using the incorporate software. Measurements were performed for 10 min acquisition live time.

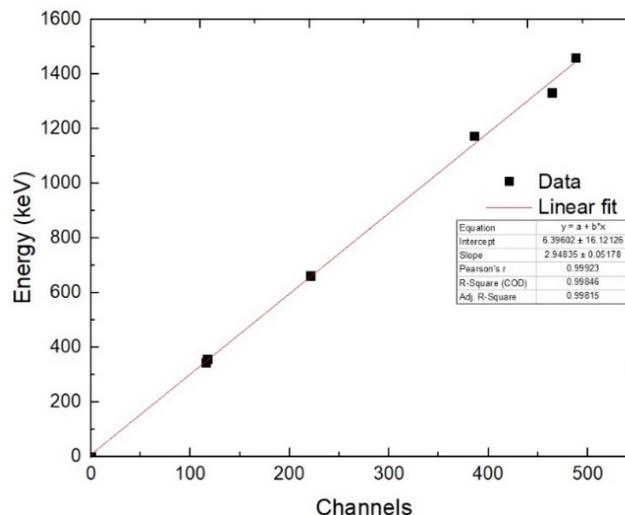


Fig. 1 – Energy calibration curve of the Gamma Surveyor II spectrometer.

Table 1

The centroid for each ROI for the isotopes used for energy calibration

Radionuclide	Energy (keV) / I_γ (%)	Channel
^{152}Eu	344.28 / 26.50	116
^{133}Ba	356.02 / 62.05	117
^{137}Cs	661.66 / 85.10	221
^{60}Co	1173.24 / 99.97	386
^{60}Co	1332.50 / 99.98	464
^{40}K	1460.83 / 11.00	488

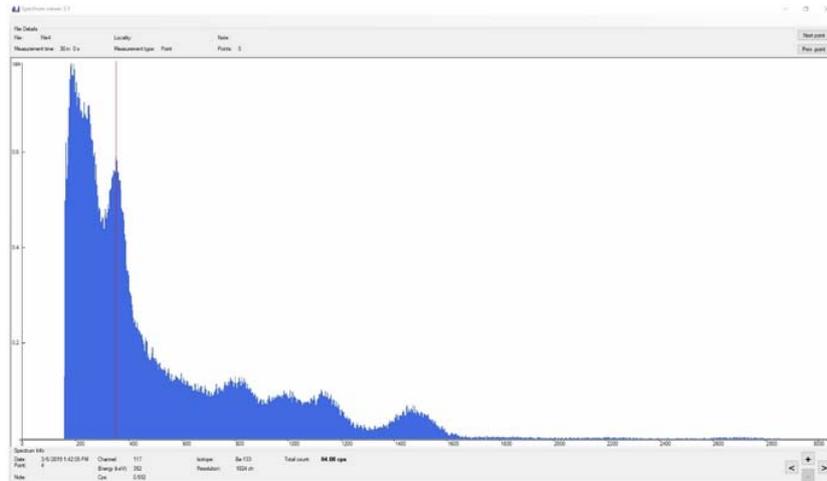


Fig. 2 – The response of the Gamma Surveyor II spectrometer to the ^{133}Ba source.

The energy resolution is defined as the full-width-half-maximum (FWHM) per centroid of the peak. However, the statistical broadening of the peaks predominates over other potential sources of resolution loss, then the variation of the resolution with energy is given by the FWHM of the peak that is proportional to the square root of energy (Eq. 1) [10, 11].

$$R = \frac{\text{FWHM}}{H_0} = K \frac{\sqrt{E}}{E}, \quad (1)$$

where K is a constant of proportionality. In experimentally measured resolution the slope is not so steep as the predicted one, then a more adequate representation of measured data is given by the following equation (Eq. 2):

$$R = \frac{(A + B \times E)^{1/2}}{E}, \quad (2)$$

where A and B are constants particular to a specific spectrometer.

The values of the resolution for the energy lines of the radionuclides presented in Table 1 are plotted in Fig. 3. Two functions have been used to obtain a proper fit of the data: the first is the one from Eq. 2 (defined as Fit Function 2 and the values of the parameters are presented in the table embedded in Fig. 3); the second is a power function, very close to the Eq. 2, but for which the slope is bit more steeper (its parameters are shown in Fig. 3). Nevertheless, the chi-squared and chi-squared reduced have very close values, resulting that both functions can be used to validate data.

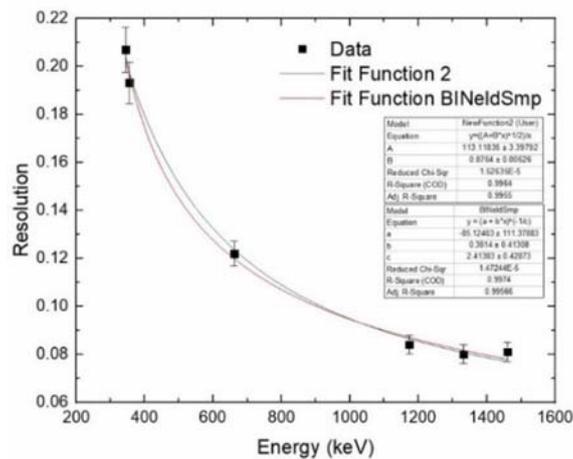


Fig. 3 – Energy resolution as a function of energy for the Gamma Surveyor II spectrometer.

Table 2

The FWHM and resolution for the measured isotopes

Radionuclide	Energy (keV)	FWHM (keV)	R (%)
^{152}Eu	344.28	11.8	3.4
^{133}Ba	356.02	11.9	3.3
^{137}Cs	661.66	12.8	1.9
^{60}Co	1173.24	14.2	1.2
^{60}Co	1332.50	14.7	1.1
^{40}K	1460.83	15.0	1.0

The background of the detector was measured placing the detector in a 10-cm thick lead castle to shield it from building materials as much as possible. In the background spectrum (the darker-lower spectrum in Fig. 4) can be observe the gamma rays from ^{207}Bi (570 keV and the sum peak 1633 keV, one wider peak is also visible at 1063 keV). These peaks stem from the crystal itself. Figure 4 shows also measured spectra using point sources of ^{133}Ba , ^{137}Cs and ^{60}Co . In the spectra, ^{40}K and the peak ^{208}Tl are observed, most likely from the environmental background [12].

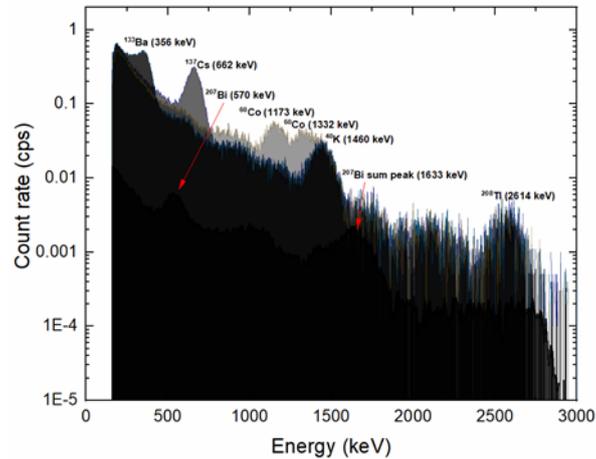


Fig. 4 – Measured spectra for ^{133}Ba , ^{137}Cs , ^{60}Co radionuclides and a background spectrum for the Gamma Surveyor II spectrometer.

According to its specifications the spectrometer calculates and gives the dose rate [nSv/h] which takes into account all contribution from ^{238}U (^{226}Ra), ^{232}Th and ^{40}K . This is highlighted by the data presented in Fig. 5 where the variation of dose rate increases with the increasing concentrations of the radionuclides taken into account. The Surveyor manual does not specify the calculation method for the dose or parameters considered, however from Fig. 5 is established a linear relation between the contribution of all three radionuclides and the dose. Obvious, the analytical equation should comprise the three components and associated coefficients as in [13, 14]. Though, at this point this analytical equation was avoided on the basis that the contribution for ^{40}K given by the Surveyor is expressed in percentage since for ^{238}U and ^{232}Th are given in ppm. The purpose of data presented in Fig. 5 is emphasizing the reliability of data given by the device.

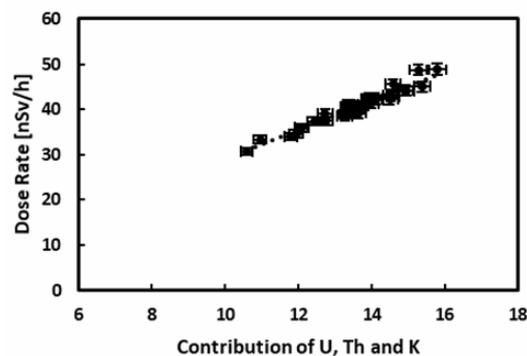


Fig. 5 – The variation of the dose rate with the contribution concentration of ^{238}U , ^{232}Th and ^{40}K as is calculated by the Gamma Surveyor II.

As mentioned before the Gamma Surveyor II is able to identify in the spectrum 17 radionuclides (Table 3), by their primary gamma energy line for all natural and artificial radionuclides. For those coming from natural decay serie of ^{232}Th two lines are considered 911.20 keV of ^{228}Ac and 2614.51 keV of ^{208}Tl . This means that the activity calculation takes into account that the latter represents only 36% of the first one. For the natural decay serie of ^{238}U the only line that is considered belongs to ^{214}Bi at 1764.49 keV which is assigned to ^{226}Ra because the energy line at 186.21 keV is entirely assigned to ^{235}U . This assumption is valid for the ^{238}U radionuclides at equilibrium. For a proper determination of nuclides from ^{238}U series the line of ^{214}Bi at 609.32 keV could be easily taken into account because as can be observed from Table 3 between ^{85}Sr (514 keV) and ^{137}Cs (662 keV) no other peak is considered in the spectrum.

Table 3

Radionuclides identified in the spectrum by the Spectrum Viewer and the ROI for each isotope

	Radionuclide	Energy* (keV)	ROI (keV)
1.	^{241}Am	59.54	52–70
2.	^{67}Ga	93.31	85–103
3.	^{57}Co	122.06	112–130
4.	$^{99\text{m}}\text{Tc}$	140.51	133–151
5.	^{235}U	185.72	178–196
6.	^{111}In	245.35	235–253
7.	^{51}Cr	320.08	310–328
8.	^{133}Ba	356.01	346–364
9.	^{85}Sr	514.00	505–523
10.	^{137}Cs	661.66	652–670
11.	^{228}Ac	911.20	901–919
12.	^{60}Co	1173.23	1165–1183
13.	^{22}Na	1274.54	1267–1285
14.	^{60}Co	1332.49	1324–1342
15.	^{40}K	1460.82	1453–1471
16.	^{226}Ra (^{214}Bi)	1764.49	1756–1774
17.	^{232}Th (^{208}Tl)	2614.51	2605–2623
18.	^{88}Y	1836.07	1828–1846

* Energy taken from [15]

3.2. *IN-SITU* MEASUREMENTS

For validation of the *in-situ* results measurements, soil samples were taken from three different locations from Bucharest (Fig. 6), but also to assure a high degree of variability of the results. Moreover, in the same point where the *in-situ* measurement was performed with the Gamma Surveyor II, a soil sample was collected. The results for ^{238}U , ^{232}Th and ^{40}K were plotted against HPGe results to show the consistency of the measurements, depicted in Figs. 7–9. There are linear correlations between the concentrations (ppm) of measured by Surveyor and activity

concentration [Bq/kg] measured by HPGe detector for the samples (Figs. 7–9). As it was expected when the concentration given by the HPGe increases also the concentration given by the Surveyor increases. However, the variations values for the concentrations are somewhat lower for the Surveyor being 35% lower for the ^{238}U , 47% lower for the ^{232}Th and 14% lower for the 40 K, compare with variations values given for HPGe. This small variation of values denotes a slightly lower sensitivity of the Surveyor (in comparison with HPGe) that could be due to statistical processes or evaluation method for the concentration of a radionuclide (window method) and, in addition, radionuclide interference can cause problems in their correct assessment.



Fig. 6 – Map with three different location to assure a high degree of variability of the results. More measurements were done from the place in the left-bottom of the corner (University Polytechnic yard), few were done in right-upper corner (Agronomic Faculty yard) and the last were in the Botanical Garden, center-bottom (also in Fig. 11).

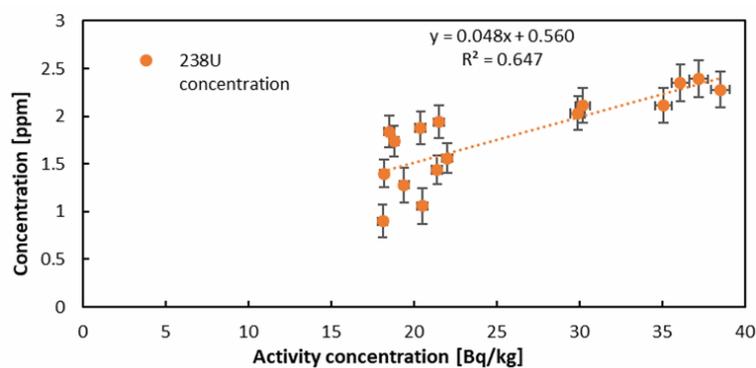


Fig. 7 – Linear correlation between concentration [ppm] of ^{238}U given by the Surveyor and activity concentration [Bq/kg] obtained from the HPGe detector.

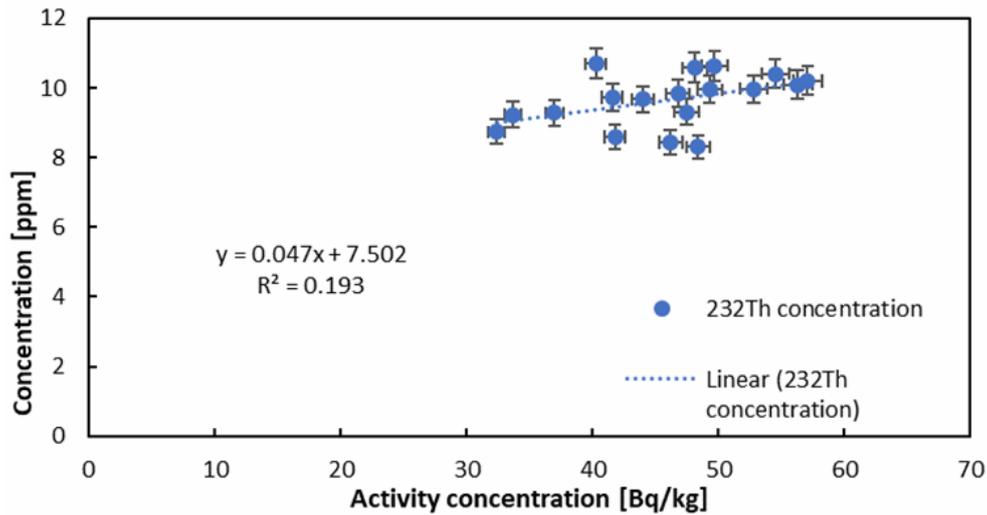


Fig. 8 – Linear correlation between concentration [ppm] of ^{232}Th given by the Surveyor and activity concentration [Bq/kg] obtained from the HPGe detector.

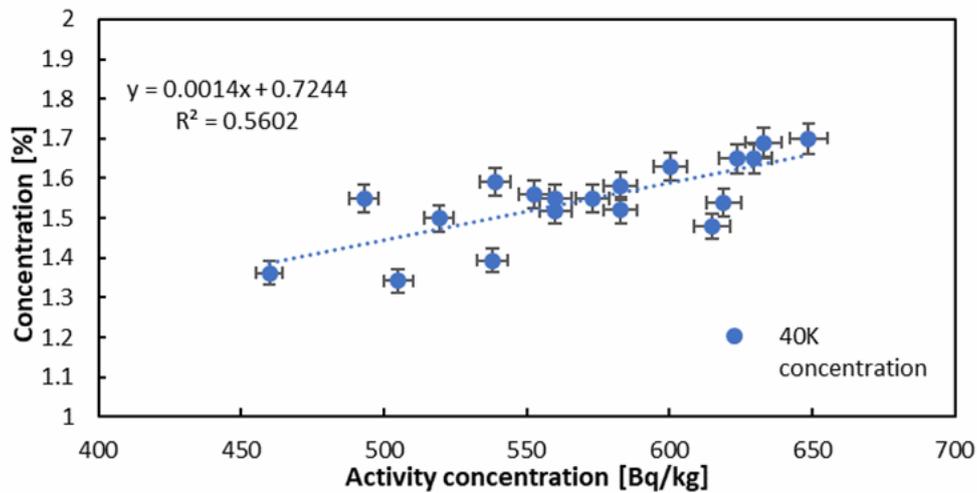


Fig. 9 – Linear correlation between concentration [ppm] of ^{40}K given by the Surveyor and activity concentration [Bq/kg] obtained from the HPGe detector.

Relatively good correlations can be observed between the Surveyor data and HPGe data as the values of R-squared are 0.65 and 0.56, for ^{238}U and ^{40}K , since for ^{232}Th this is weak and the value is about 0.20. The signification of the data correlation between both measurement systems is analyzed in Fig. 10, where it is shown the reliability and ability of Gamma Surveyor II to measure natural radiation *in-situ*, with acceptable measurements errors. Although, there are low

data variations (concentrations [ppm, %]) it follows the same trend as data calculated for the HPGe detector, which demonstrate the correspondence of the measurements.

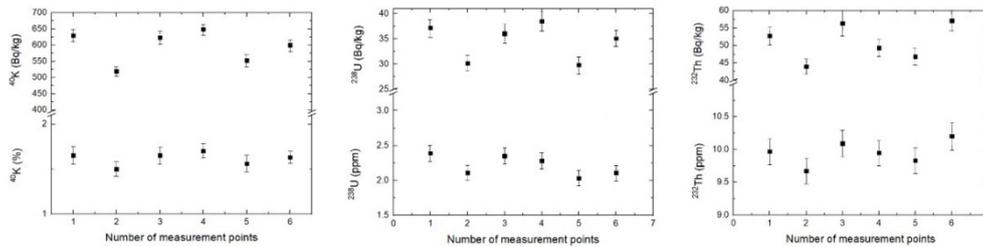


Fig. 10 – Variations of ^{40}K , ^{238}U and ^{232}Th concentrations point by point for both measurement systems.

The results for ^{238}U , ^{232}Th and ^{40}K plotted against HPGe showed the consistency of the measurements (Fig. 10). The results obtained for the Surveyor measurement system recommend using this type of system for *in situ* gamma spectrometry related to geophysical methods [16–20].

The advantages of using this system include:

- less time-consuming regarding acquisition and analysis time, the reporting of the data being instantaneous, at the elapsed time;
- does not require liquid nitrogen, as for HPGe detectors;
- cost-effective way to obtain concentrations of natural radionuclides in soil.

Field measurements and laboratory data using Gamma Surveyor II and HPGe spectrometers are plotted in Fig. 11 for the concentrations of ^{40}K , ^{238}U , ^{232}Th radionuclides. The contour maps were made using radial base functions for interpolation, extrapolation and approximation of scattered data. The spatial distribution resulted for both spectrometers shows good comparison. Although Gamma Surveyor II showed a lower sensibility compared to the HPGe detector (Figs. 7–9), but field data showed good consistency when they are either large or small values. For instance, point P3 showed the highest concentration for ^{40}K in both maps (Fig. 11, upper left and right). Also, the measurement point P8 (Fig. 11, bottom left and right) showed the highest concentration for ^{238}U , since P11 exhibit one of the lowest for both spectrometers measurements. Only for ^{232}Th , the concentrations seem to be rather weak correlated, as shown also in Fig. 8, where the correlation factor R -squared is the weakest (0.19).

However, in practical studies, for geophysical explorations and to measure the degree of regional variability or irregularities activities concentrations of various locations the Gamma Surveyor II spectrometer proves to be a reliable alternative. Even if in the above figures (Fig. 11), the variations of the concentrations do not match point by point, the general aspect of increase or decrease is emphasized for the area studied.

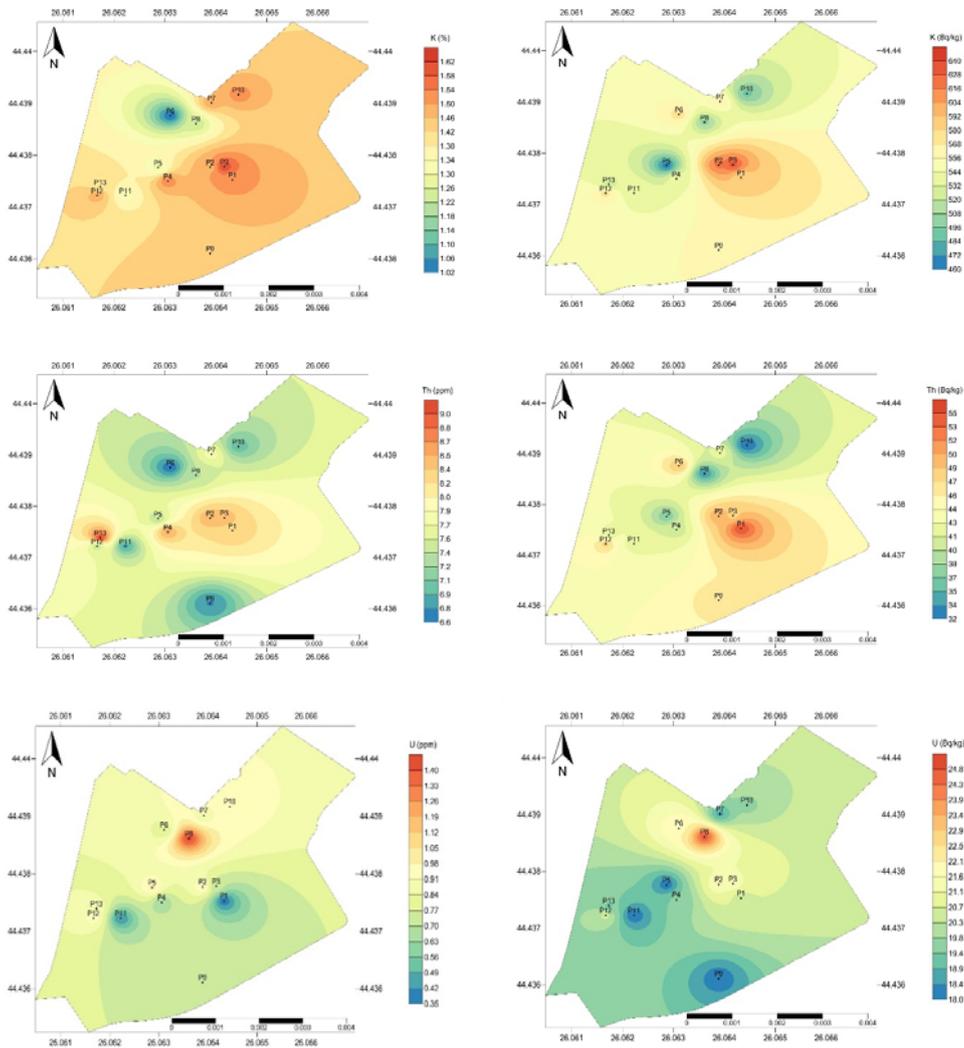


Fig. 11 – Spatial distributions of ^{40}K [%], ^{238}U [ppm], ^{232}Th [ppm] in soil measured with both systems (left side Gamma Surveyor II; right side HPGe detector).

4. CONCLUSIONS

The purpose of this paper was to verify the performances of the *in-situ* Spectro-tracer Gamma Surveyor II, the correctness of the data recorded for natural radionuclides ^{40}K , ^{232}Th and ^{238}U by comparison with data from a HPGe detector. Analyzing the presented results, it was shown the consistency and the reliability of the measured data using the Spectro-tracer. As part of multi-disciplinary studies as

survey of radioactive heat production from rocks or studies of tectonic structures using simple geophysical methods this type of equipment represent a viable alternative for *in situ* measurements.

There are many advantages of using this system, such as: fast reporting data, less time-consuming measurements, no liquid nitrogen requirement as for the *in-situ* HPGe detectors, easy to handle, BGO crystal contributes to a better response to the presence of radionuclides in the soil as its density is higher compare to the NaI crystal, cost-effective way to obtain concentrations of natural radionuclides in soil.

Among the weaknesses or ways to improve and to complete the system would be the fact that although the system has a built-in library of 19 radionuclides (natural and artificial) distinguishable in the spectrum, it does not calculate their concentrations. From this point of view this represents a loss, because in all measured samples on the HPGe detectors, ^{137}Cs has been detected and calculated with activities between few tens up to more than a hundred Bq/kg. Also, a better stabilization of the spectrum would give better linear correlation of the data, R^2 close to 1, because has been observed that measurements in various days could lead to different response of the system, as the scintillation yield of the BGO crystal is temperature depended, about 10% between 20°C and 30°C. This was observed from the relative scattering data.

Baseline terrestrial gamma radiation maps, using *in situ* measurements with modern ad cost effective equipments such as Spectro-tracer, are indispensable for providing basic reference information that may be used in assessing the impact of a radiation related incident, performing epidemiological studies, remediating land contaminated with radioactive materials, assessment of land use applications and resource exploration. However, these systems must be well evaluated for the accuracy and precision of their data.

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This study was out to provide an assessment of the instrument used in various projects for *in situ* measurements and the authors declare that do not promote commercial interests.

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