# Performances of a lightweight collimated γ-ray spectrometer for in-situ surveys

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# Abstract

In this study we present the design and the performances of Cava-rad system, a portable collimated  $\gamma$ -ray spectrometer based on lead-plate method. The system is calibrated using the full-spectrum analysis method with non-negative least square constraint in order to obtain the fundamental standard spectra of background, potassium, uranium, thorium and cesium. The results are validated by comparing the activity concentration of natural radionuclides  $^{40}$ K,  $^{238}$ U and  $^{232}$ Th measured in 35 test sites with those measured using HPGe detectors. The mean discrepancies between in situ survey and laboratory measurements are approximately of 20% for  $^{40}$ K and for  $^{232}$ Th and 30% for  $^{238}$ U.

# Keywords

Collimated  $\gamma$ -ray spectrometry; *In situ*  $\gamma$ -ray spectroscopy; Environmental radioactivity; Natural radioactivity; Full spectrum analysis

# **1. Introduction**

In situ  $\gamma$ -ray spectrometry with NaI(Tl) detectors is a well consolidated as a technique for local scale radioactive surveys. It is employed in a wide field of applications, from mineral exploration to environmental radiation monitoring, providing quantitative information on the radioactivity content of the investigated sites, especially for the most abundant natural radioisotopes, <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th (Nuccetelli 2008, Chiozzi et al. 2000). Factors influencing the reliability and the quality of *in situ* measurements are related to site location features (e.g. morphology and geoengineering characteristics), environmental conditions (e.g. changes in water content) and to the acquisition and analysis (e.g. detector type and spectral analysis). Sometime the accuracy of *in situ*  $\gamma$ -ray survey is affected by the vertical migration of radionuclides in the soil, in particular in case of presence of <sup>137</sup>Cs or of natural radionuclides enriched in industrial residues. The determination of <sup>137</sup>Cs content using scintillation detectors stimulated over years two main scientific challenges: the first is the overcoming of the physical

limitations of *in situ* measurements, such as the intrinsic energy resolution of NaI(Tl) detectors (Caciolli et al. 2012, Kovler et al. 2013), and the second concerns the study of radionuclide vertical migration. In this framework, Korun et al. (1994) developed the so-called lead-plate method in order to observe the changes in the vertical distribution of radioactive contaminants (e.g.  $^{137}$ Cs): this method uses a circular *lead-plate* positioned between the detector and the ground to shield  $\gamma$ -rays from small angles. The contribution to the flux from large angles varies with the depth parameter and thus the ratio of the peak counts obtained with the *lead-plate* in place to that obtained without the lead plate will give an indication of the activity-depth distribution.

Efforts for improving the accuracy of *in situ* measurements are highly boosted in order to make such technique an effective screening method to fulfill the requirements of the new Euratom Basic Safety Standards (**BSS 2014**) regarding building materials and naturally occurring radioactive materials (NORM residues) (**Stals et al. 2014**, **Kovler et al. 2013**). In situations such as site remediation and monitoring, building materials quarrying, mineral exploration, both the contamination history and geochemical processes can lead to a spatial variability of the radionuclide concentrations (**Zhao et al. 2012**; **Haddad et al. 2014**). Some attempts to limit the detection of unattenuated  $\gamma$ -rays from a selected range of polar angles are proposed using lead as collimator (**Benke and Kearfott 2002**; **Lovborg et al. 1971**). The major drawbacks of reducing the detectors' field of view by folding them with lead are the increase of instrument's weight and of the time response.

Taking advantage of the existing state-of-art, this study focuses on the feasibility of attenuating the environmental noise without compromises the detectors' portability and extends the counting time. We propose to obtain the collimation effect subtracting the spectra acquired with the *lead plate* to the spectra acquired without the *lead plate* in place. The difference spectrum corresponds to the signal originating from a selected range of polar angles. A similar approach was formerly introduced in order to improve the signal to background ratio for location of point sources in mobile  $\gamma$ -ray spectrometry measurements (Cresswell and Sanderson 2009) applying the filtering "rolling" average background spectra. The optimization of the acquisition time and the reduction of the weight are obtained also adopting the Full Spectrum Analysis method with Non-Negative Least Squares constraints (Caciolli et al 2012) to perform the analysis on the difference spectra. The calibration process produces *standard spectra* of background, potassium, uranium, thorium and caesium, which are used to analyze *in situ* measurements. The quality of the results is controlled by 35 *in situ* tests, with various morphologies, physical and environmental configurations. A comparison with the data obtained with high-resolution  $\gamma$ -ray spectrometers (HPGe) is also discussed.

## 2. Material and methods

#### 2.1 Cava-rad system set-up design and operation

The portable collimated  $\gamma$ -ray spectrometer called Cava-rad system is essentially made up of a *lead plate* of 9.0 cm × 9.0 cm × 3.0 cm dimensions, which is fully automated to shield on/off a 3 in. cubic shaped NaI(Tl) crystal having an energy resolution of 7.3 % at 662 keV (<sup>137</sup>Cs), 5.2 % at 1172 and 1332 keV (<sup>60</sup>Co). The *lead plate* is used to partially shield a target area of approximately 100 cm<sup>2</sup> that we define as Field Of View (FOV) of the detector. Subtracting from the spectrum acquired with shielding off (PbOut of the FOV), the spectrum obtained with the shielding on (PbIn the FOV) the Difference Spectrum (DS) is obtained (**Figure 1**). We estimate that DS is contains about 20% of the total counts collected with PbOut. The dimension of the *lead plate* and in particular the shielding thickness (in our case 34 g/cm<sup>2</sup> was chosen as appropriate) is crucial since it determines the quality of the DS and therefore the accuracy of

the measurement. The obtained DS reproduces the collimation effect by filtering the surrounding background, including the escaped  $\gamma$ -rays from the target area.

The system is mounted on rugged case in PVC where the NaI(Tl) detector is rigidly arranged in order to prevent possible damages on the field (**Figure 1**). The NaI(Tl) detector is coupled with a photomultiplier (PMT) base with integrated bias supply, preamplifier and digital multichannel analyzer (MCA). It is powered through a universal serial bus (USB) communication connector by a notebook which is also used for data storage. The movement of the lead shield upon a conveyor belt is driven by an electric motor powered by a 9 V battery. The lead plate movement is automatically governed by an electronic set-up together with JAVA program. The user can set also some data taking parameters (e.g. live time).

Possible influences on the spectral shape due to the presence of the *lead plate* can be neglected by selecting an appropriate energy range. Indeed, the lead used as shielding material can add some extra background due to the presence of <sup>210</sup>Pb: this radionuclide has a half-life of 22.3 years and emits characteristic  $\gamma$  energy at 46.5 keV and a bremsstrahlung continuum from beta decay of its daughter <sup>210</sup>Bi, extending from low energy up to 1162 keV. Furthermore, when hitting the lead surface, characteristic lead X-rays may escape and reach the detector. The *lead plate* gives rise also to backscattered events within a broad range of energy, between 200 keV and 300 keV.



**Figure 1.** Cava-rad (panel D) and a scheme of data taking: the configurations PbOut and PbIn are reported in panels A and B respectively. The 10 cm x 10 cm target area is in the panel C.

The advantage using the *lead plate* method consists on improving the *in situ* measurements increasing the capability to filter the background especially due to atmospheric radon or the presence of spatial variability. The indirect collimation allows also to reduce the weight of the

instruments and to optimize the counting time, even with a relatively small volume of NaI(Tl).

#### 2.2 Calibration of the Cava-rad system

The Cava-rad system is designed for *in situ* gamma spectroscopy for quantifying the abundances of radionuclides (i.e. <sup>40</sup>K, <sup>238</sup>U, <sup>232</sup>Th and <sup>137</sup>Cs) in a restricted area underneath the scintillation detector (NaI(Tl)). Its calibration is based on the selection of calibration sites characterized by a prevalent concentration of a single radionuclide with the purpose to adopt the Full Spectrum Analysis (FSA) with the Non-Negative Least Square (NNLS) method (Caciolli et al. 2012). We identify ten sites, which are characterized for the abundances of <sup>40</sup>K, <sup>238</sup>U, <sup>232</sup>Th and <sup>137</sup>Cs by collecting a sample underneath the target area of Cava-rad. A bulk sample is also collected around the target position within a radius of 1 meter, with the aim to check the homogeneity with respect to the target area. Attention must be done in the case of NORM residues, where due to technological processes; <sup>238</sup>U and <sup>232</sup>Th are typically found in secular disequilibrium with long lived daughters, e.g. <sup>226</sup>Ra and <sup>228</sup>Ra/<sup>228</sup>Th. In these cases, additional surveys have to be done to control the disequilibrium and then the results must be expressed in terms of the radionuclides present in NORM samples. In this study were considered natural sites, where <sup>238</sup>U and <sup>232</sup>Th decay chains are reasonably assumed in secular equilibrium, therefore now on the results are expressed as U and Th.

Samples are homogenized up to 1 cm grain size, dried at 105°C temperature, placed in measurement containers and left undisturbed for at least four weeks in order to establish radioactive equilibrium in the <sup>238</sup>U decay chain segment after <sup>226</sup>Ra. The activity concentrations are measured on 180 cm<sup>3</sup> samples using the MCA\_Rad system (**Xhixha et al. 2013**). This fully automatic spectrometer consists of two 60% relative efficiency coaxial p-type HPGe  $\gamma$ -ray detectors, with an energy resolution of ~1.9 keV at 1332.5 keV (<sup>60</sup>Co). The absolute full energy peak efficiency of the MCA\_Rad is calibrated using certified standard point sources (<sup>152</sup>Eu and <sup>56</sup>Co). The overall uncertainty in the efficiency calibration is estimated to be < 5% (**Xhixha et al. 2013**).

In **Table 1** we report the abundances with their experimental uncertainties of radionuclides in the target area measured with MCA\_Rad system. These results will then be used as input concentrations in the calibration process (see flowchart in **Figure 2**). From measurements of the bulk samples surrounding the target area we infer that the variability in the calibration sites is generally < 30% for <sup>40</sup>K and <sup>238</sup>U and < 20% for <sup>232</sup>Th. The calibration spectra of <sup>137</sup>Cs allows us to better reconstruct the measured spectra in the case of the presence of this radionuclide: this one of the main advantage of using FSA with NNLS analysis. The quality of each calibration site can be assessed also according to the prevalent presence of a specific radionuclide compared to the others. In general we observe typical Th/U ratios ranging from 4.6 to 5.7. A markable anomaly of thorium enrichment was found in the placer deposit (Soleminis, Sardinia), corresponding to a Th/U ratio of 14.1. The K/U ratio shows typical values ranging from  $0.3 \times 10^3$  to  $1.2 \times 10^3$ . The uranium anomalies are visible in the Piovene Rocchette and Arsiero sites, together with placer deposit (Soleminis, Sardinia) site. On the other hand, not satisfactory anomalies are found for potassium, considering the K/Th ratios are in the range between  $0.1 \times 10^3$  and  $0.5 \times 10^3$ .

Table 1. The activity concentrations ( $\pm 1 \sigma$  statistical uncertainty) of radionuclides measured in 10 calibration sites. The ratios emphasize the anomaly presence of one radioisotope. The n/a annotation stay for non-available data, while data indicated as less than are the minimum detectable activities.

ID	Place	${}^{40}K\pm\sigma$ (%)	$\frac{^{238}U\pm\sigma}{(\mu g/g)}$	$^{232}Th \pm \sigma \\ (\mu g/g)$	$^{137}Cs \pm \sigma$ (Bq/kg)	Th/U	K/U (× 10 <sup>3</sup> )	K/Th (× 10 <sup>3</sup> )
K2	Galzignano terme	$6.0\pm0.1$	$4.9\pm0.2$	27.9 ± 1.1	$10.2 \pm 1.1$	5.5	1.2	0.2
K4	Recoaro	$2.8\pm0.1$	$2.5\pm0.3$	$13.4\pm0.8$	$6.5\pm0.8$	5.4	1.1	0.2
U1	Piovene Rocchette	< 0.04	$7.4\pm0.2$	<1.0	$2.0\pm0.6$	-	-	-
U3	Arsiero	$0.05\pm0.01$	$7.5\pm0.2$	<1.0	< 1.4	-	0.006	-
Th1	Castelvecchio (Altissimo)	$2.1\pm0.1$	$0.9\pm0.1$	$4.2\pm0.6$	< 1.6	4.7	2.3	0.5
Th	Placer deposit (Soleminis, Sardinia)	$2.3\pm0.1$	$25.6\pm0.5$	$360.7\pm3.3$	< 3.2	14.1	0.08	0.006
Cs1	Monte Novegno (Schio)	$0.5\pm0.1$	$1.2\pm0.2$	< 3.9	1496 ± 13	-	0.4	-
H1	Galzignano terme	$4.2\pm0.1$	$12.3\pm0.3$	$55.9 \pm 1.5$	< 2.8	4.6	0.3	0.08
H2	Galzignano terme	$4.1\pm0.1$	$8.0\pm0.2$	$36.8 \pm 1.3$	< 2.6	4.6	0.5	0.1
Bckg1	Lido di Spina	n/a	n/a	n/a	n/a	-	-	-

In each calibration site two measurements of 20 minutes each are performed placing the Cava-rad system on the target location. The first acquisition is encoded as PbOut (indicating that the *lead plate* is out of the FOV of the detector) and consequently the second one is the PbIn (the *lead plate* is in the FOV of the detector). A blank site, i.e. the site assumed to have negligible level of terrestrial radioactivity, is chosen 0.5 km offshore in the Adriatic Sea (Lido di Spina). The Cava-rad system is mounted on an inflatable boat over approximately 3 m of water, which is enough to shield more than 99% of the seabed radioactivity. The main sources of background events are cosmic radiation, atmospheric radon (<sup>222</sup>Rn) and the lead shield activity. The contribution from inflatable boat is considered negligible. Taking into account the low counting rate the acquisition time of this acquisition is extended to 1 hour.

Each measured spectra is energetically calibrated with a linear relationship on the base of the linearity of the NaI(Tl) in the energy range used. The rebinned spectrum is obtained using a pseudo-random number generator. The final spectrum is characterized by a zero offset and a fixed kev/channel parameter i.e. 12 keV/channel, corresponding to 256 channels. Since we are interested only in the energy range [300 - 2900 keV] the contamination by the *lead plate* on the spectrum quality is negligible. Following the FSA-NNLS method described in (Caciolli et al. 2012), we obtain the calibration standard spectra, as described in the calibration process flowchart (Figure 2). In particular for each acquisition the DS is calculated, and then the FSA-NNLS algorithm is applied including the concentration of the target area measured with MCA\_Rad. The standard spectra obtained are shown in (Figure 3). The DS are affected by a relatively low statistic and it produces some fluctuations in the standard spectra.



**Figure 2.** The flowchart represents the calibration process of Cava-rad system. For each acquisition, after the energy calibration, the DS is calculated. The FSA NNLS algorithm is applied including the concentration of the target area characterized by measurement with MCA\_Rad system.

# 3. Experimental cross-validation

The performances of Cav-rad system and the robustness of the calibration method are crossvalidated comparing the results obtained *in situ* with those from MCA\_Rad for 35 test sites. These locations are chosen with different geometrical configurations, variable abundances, densities and water content in order to have different source of environmental noise. In particular, we investigate up to 20 sites in Mt. Vulsini-Latera complex, a volcanic area of the Tuscan Magmatic Province (South Tuscany region, Italy), which is characterized by a thick quaternary coverage of pyroclastic rocks (ignimbrites) having high variability of radioactivity content. The outcrop shows a flat and smooth layering, whose stratigraphy is related with the chemical composition, according with the past activity of the volcano (Conticelli et al. 1986). In Euganean hills (Veneto region, Italy) we investigate 8 outcrops of acid effusive rocks (like trachytes and rhyolites) connected to a Paleogene volcanic activity (Milani et al. 1999) In Sardinia region the Cava-rad system is tested in 4 outcrops of upper Ordovician terrigenous succession (Barca, 1982), characterized by Quartz-sandstone affected by the layered deposition of heavy sand containing rare earth elements. The rest of the sites are chosen in random deposits of soil and sand. In each site a representative sample is collected from the target location investigated by the Cava-rad system and transported in laboratory for a measurement with MCA\_Rad system.



Figure 3. Fundamental standard spectra of background, potassium, uranium, thorium and cesium obtained with the FSA-NNLS method applied to Cava-rad system. In the last panel we report a typical measured/reconstructed spectrum having a reduced  $\chi^2$  of 1.2.

On the base of measurements in laboratory the test sites are characterized by ranges of abundances 0.2  $\div$  7.7 %, 1.3  $\div$  34.3  $\mu g/g$  and 1.0  $\div$  101.7  $\mu g/g$  for  $^{40}K,~^{238}U$  and  $^{232}Th$ respectively. The activity concentration of <sup>137</sup>Cs is found to be below the minimum detectable activity and therefore we neglect it for this comparison. The durations of the in situ acquisitions are of 5 minutes for both PbOut and PbIn configurations. The average relative uncertainties affecting the *in situ* measurement are approximately 8% for <sup>232</sup>Th, 10% for <sup>40</sup>K and 20% for <sup>238</sup>U. In situ measurements suffer higher statistical uncertainties respect to laboratory measurements: such result has been expectable since only approximately one fifth of the events of the measured spectra are considered in the analysis process. Globally we observe a good agreement between the results obtained in situ and those measured in laboratory: the coefficients ( $\Omega_{\rm K} = 0.93 \pm 0.21$ ,  $\Omega_{\rm U} = 0.93 \pm 0.29$  and  $\Omega_{\rm Th} = 0.82 \pm 0.18$ ) of linear correlation obtained weighting the experimental uncertainties (Figure 4) are compatible to unity at 1 sigma level. We note that Cava-rad system seems to underestimate weakly the abundances measured with MCA\_Rad. This result could be a consequence of the transparency of lead for photons with highest energy (i.e. 2614 keV of <sup>208</sup>Tl in the <sup>232</sup>Th decay chain). The values  $\Omega_{K}$ ,  $\Omega_{U}$  and  $\Omega_{Th}$  can be used as empirical corrections for dividing the measurements performed by Cavarad system. The relative percentage uncertainties  $\delta\Omega_{\rm K} = 22\% \ \delta\Omega_{\rm U} = 31\%$  and  $\delta\Omega_{\rm Th} = 22\%$  are used as systematic uncertainties on the concentration measurements obtained with Cava Rad, since they include the systematics due to method of analysis.



Figure 4. Correlations between measurements performed by Cav-rad and MCA\_Rad in 35 test sites. The coefficients of linear correlation obtained weighting the experimental uncertainties are  $\Omega_{\rm K} = 0.93 \pm 0.21$  (panel A),  $\Omega_{\rm U} = 0.93 \pm 0.29$  (panel B) and  $\Omega_{\rm Th} = 0.82 \pm 0.18$  (panel C). Error bars show statistical uncertainty (1  $\sigma$ ). In panel D the typical in-situ configuration with Cava-rad system is reported.

## 4. Conclusions and perspectives

The Cava-rad system is a portable (8 kg) collimated  $\gamma$ -ray detector designed for fast and in situ spectroscopy. A lead-plate method is improved in order to "filter" the background noise, reducing the instrument weights and the counting response time. Subtracting two spectra obtained with shielding on/off, one can calculate the DS which contain approximately 20% of the unattenuated  $\gamma$ -rays from the target area. The FSA method with NNLS constrains is successfully applied for the spectra analysis. On the base of data collected from ten natural calibration sites five fundamental standard spectra (K, U, Th, Cs and background,) are obtained for Cava-rad system. Finally, the calibrated Cava-rad system is tested for extracting in-situ the abundances of radionuclides in 35 different sites characterized with HPGe independent measurements on samples. The coefficients of linear correlation ( $\Omega_{\rm K} = 0.93 \pm 0.21$ ,  $\Omega_{\rm U} = 0.93$  $\pm$  0.29 and  $\Omega_{Th}$  = 0.82  $\pm$  0.18) between Cava-rad and laboratory results are comparable to the unity at 1 sigma level. We observe that Cava-rad system slightly underestimates the radionuclide concentrations: empirical coefficients  $\Omega_{K} = 0.93$ ,  $\Omega_{U} = 0.93$  and  $\Omega_{Th} = 0.82$  can be used for correcting the measurements. The relative percentage uncertainties  $\delta\Omega_{\rm K} = 22\% \ \delta\Omega_{\rm U}$ = 31% and  $\delta\Omega_{\rm Th}$  = 22% presently used as systematic uncertainties on the concentration measurements can be improved increasing the counting time during the calibration and reconstructing the expected signals with Monte Carlos simulations.

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