

Application of a New In Situ Calibration Technique for Gamma Ray Spectrometry and Comparison of In Situ and Laboratory Measurements Taking into Account Realistic Croatian Conditions

Davor Rašeta, Branko Petrinc, Dinko Babić, Marko Šoštaric

Institute for Medical Research and Occupational Health

Ksaverska cesta 2, Zagreb, Croatia

draseta@imi.hr, petrinc@imi.hr, dbabic@imi.hr, marko.sostaric1@gmail.com

ABSTRACT

After a nuclear accident, it is important to quickly measure possible affected area to determine where emergency and remediation measures are needed. In situ gamma ray spectrometry was developed to quickly measure large areas of land following nuclear weapon tests and possible nuclear accidents. However, a proper calibration of detectors for in situ measurements is a long and complicated process. One tool designed to make this calibration quick is the InSiCal software.

InSiCal software can shorten the in-situ calibration procedure to a single measurement (combined with calculations). We decided to investigate if the method can be implemented in Croatia, within expected constraints of emergency situations (especially time constraints).

We compared in situ measurements made with two different HPGe detectors calibrated using the InSiCal software and laboratory measurements of samples collected at the same locations. Detector calibration and in situ measurements were optimized for quickness, simulating time pressure present in case of a nuclear accident.

Measurements of both in-situ detectors were reasonably close – in most cases the confidence intervals overlapped. In-situ measurements generally undershot laboratory measurements. Large uncertainty intervals at energies below 100 keV make short in-situ measurements unsuitable in that energy range. If the range below 100 keV is important, the duration of the measurements must be increased.

Our findings suggest that in situ gamma spectrometry using InSiCal software can provide reasonably accurate data, but some improvements may be needed.

Keywords: *emergency, HPGe spectrometers, InSiCal software, radiation, radionuclide measurements*

1 INTRODUCTION

A nuclear accident can leave vast areas of land potentially contaminated. Radiological contamination in areas where people live should be quickly determined to ensure that they are safe for living or to apply the required emergency measures. It is equally important to determine the contamination on agricultural land, both to restrict the use of contaminated fields and to ensure the public that the food produced on the fields cleared for use is safe. Uninterrupted food supply is, of course, one of the imperatives of emergency management.

In-situ gamma-ray spectrometry was first used to measure the fallout from nuclear testing, in addition to the naturally occurring isotopes. It is widely used since the 1960s [1], [2]. In the beginning, scintillators only were used. Since 1980s, HPGe (cooled with liquid nitrogen) often replace the scintillators in the field, because of their superior resolution. Since the turn of a century,

mechanically cooled HPGe detectors are replacing more cumbersome liquid nitrogen-cooled HPGe detectors.

The first elaborate description of the in-situ gamma-ray spectrometry method was published in 1972 [3]. In late 1990s the IAEA accepted the technique for characterization of contaminated sites and for monitoring following a nuclear or radiological emergency [4], [5]. An ISO guide on the in-situ gamma-ray spectrometry in soil was published in 2013 [6].

In-situ gamma-ray spectrometry is faster compared to the traditional sampling and laboratory measurements and less sensitive to localized variations. It is especially useful in searching for hot spots in large areas. However, calibration of the detector for work in the field is complicated and time-consuming, and lots of time and effort are needed to develop the methodology to the level where confidence in the results could be comparable to the laboratory results.

In 2018, a tool developed by the Norwegian Radiation Protection Authority was presented [7]. This tool, InSICal, enables much quicker and easier calibration of HPGe detectors for in-situ measurements.

Using InSICal, we calibrated two detectors with different efficiencies, different manufactures, one cooled with liquid nitrogen, the other electrically. We performed in-situ measurements at 7 locations in Croatia, chosen as likely locations of interest in a case of nuclear accident. The results were compared against the laboratory gamma-ray spectrometry measurements performed on the soil samples taken from the same locations.

The following radionuclides were analysed: ^{40}K (at 1460.8 keV), ^{238}U (through ^{234}Th at 63.3 keV and the double peak at 92.4 and 92.8 keV), ^{232}Th (through ^{228}Ac at 333.3 keV, 911.2 keV, and 969.0 keV), ^{214}Bi (at 609.3 keV and 1120.3 keV), ^{214}Pb (at 295.2 keV and 351.9 keV), and ^{137}Cs (at 661.7 keV).

The duration of calibration measurements and of in-situ measurements was determined taking into account speed over accuracy, as is expected in an emergency.

2 MATERIALS AND METHODS

2.1 Detector Calibration

Detectors used for in-situ measurements are Ortec Detective-Ex, 17 % relative efficiency at 1,332 keV, and Canberra GC, 54 % relative efficiency at 1,332 keV. For laboratory measurements, Canberra GC, 54 % relative efficiency at 1,332 keV detector was used.

Laboratory measurements were performed using standard laboratory geometry (200 ml cylinder) on a detector calibrated with a calibration source in the same geometry. All laboratory measurements lasted 80,000 seconds. For in-situ measurements, both detectors were calibrated using ^{152}Eu point source positioned 1m directly below the detector in the direction of its symmetry axis. Calibration measurements lasted 17,000 seconds, and in-situ measurements lasted 5,000 seconds.

2.2 Calibration Model

The efficiency of the detector at specified energy (ϵ_E) can be calculated from the measurement of the net detected photons from a known source at specific energy (R_{CE}), activity of the source (A_C) and emissivity of the source at that specific energy (p_E):

$$\epsilon_E = \frac{R_{CE}}{A_C p_E} \quad (1)$$

This efficiency can be expressed as a product of three separate factors, the intrinsic efficiency of the detector (η_0), the correction factor for different responses depending on the angle of the gamma ray (W) and correction factor for the geometry of the sample (G) [7]:

$$\epsilon_E = \eta_0 W G \quad (2)$$

In laboratory, there is no need to calculate separate factors, since using a calibration source in the same geometry as a sample removes the need for separate calculation of G and W . However, in

the field calibration source cannot be in the same geometry as a sample. The discussion of the factors for in-situ measurements is, for example, in [7]. A discussion about uncertainty calculation for in-situ measurements is in [8].

2.3 InSiCal

Using intrinsic efficiency calculations, a known geometry of the detector, an assumed distribution of the radionuclides, and Monte Carlo methods, InSiCal calculates W and G . For this work we used the InSiCal version 0.91 obtained through the IAEA. InSiCal requires input for each selected gamma ray energy separately. It has four possible radionuclide distributions – surface, uniform, step, and exponential [7]. Surface distribution assumes that the entire activity of the analysed radionuclide is distributed on the surface, and is an appropriate selection after a release. Uniform distribution assumes that the analysed radionuclide is uniformly distributed throughout the soil and is an appropriate selection for naturally occurring radionuclides. Step (or “slab”) and exponential distributions assume uneven distribution of radionuclides, leeching from the top layers toward bottom layers. Exponential distribution is the correct one to describe the physical process, but requires some knowledge of the actual distribution in soil. Step distribution is an approximation of the exponential distribution, where it is assumed that the analysed radionuclide is present uniformly in soil up to a certain depth. It may be appropriate for ^{137}Cs , especially in situations where relaxation length is not known.

2.4 Site selection

While Croatia does not have nuclear power plants, NPP Krško in Slovenia is located mere 10 km from the Croatian border, and NPP Paks in Hungary is but 65 km from the border. We therefore chose measurement sites in and around seven Croatian settlements closest to either of the NPPs. In case of a nuclear accident in one of those two NPPs, it is reasonable to expect that those are the locations where in-situ measurements will be needed. Coordinates of the selected sites are presented in Table 1.

Table 1: Site locations

Site	Altitude	Longitude (°N)	Latitude (°E)
Bregana (BR)	187 m	45.825	15.635
Klanjec (KL)	169 m	46.046	15.731
Plavić (PL)	202 m	46.116	15.613
Batina (BA)	80 m	45.840	18.848
Beli Manastir (BM)	102 m	45.753	18.637
Baranjsko Petrovo Selo (BP)	89 m	45.763	18.463
Zlatna Greda (ZG)	79 m	45.721	18.867

2.5 Measurements

We decided to measure each site for 5,000 s as an expected compromise between the confidence in the results and the lack of time in an emergency. The Measurements were performed an area with uncultivated land and low vegetation (forest clearing or a meadow). At each site soil sample, down to the depth of 10 cm, was taken. Soil was cleared of foliage and gravel, fully dried, mixed, sieved, and packed into 200 ml cylindrical plastic containers, where they were sealed with a duct tape. The whole process took almost three weeks, demonstrating why in-situ measurements are preferable in an emergency situation. The samples were measured immediately after they were packed, and again more than a month later, to ensure that secular equilibrium between ^{226}Ra and ^{222}Rn is reached again.

3 RESULTS

For laboratory measurements, 2σ confidence intervals overlapped for all the radionuclides in all the samples except for ^{226}Ra (measured through the daughter radionuclides ^{214}Bi and ^{214}Pb). The results of ^{226}Ra laboratory measurements are presented in Table 2.

Table 2: Activity concentration of ^{226}Ra in Bq/kg ($A \pm 2\sigma$)

Site	First measurement	Second measurement
Bregana	30±1	33±1
Klanjec	33±1	38±1
Plavić	42±1	55±1
Batina	38±1	44±1
Beli Manastir	44±1	57±1
Baranjsko Petrovo Selo	41±1	54±1
Zlatna Greda	54±1	60±2

For each sample measured activity concentration after the reestablishment of secular equilibrium was higher than the measurement immediately after sampling, as expected. The difference was between 10% and 32% of the initial value.

When measuring deposition after an emergency, measuring daughter radionuclides to assess the activity of the radionuclides of interest should be done very carefully. In the presented measurements we can see that only part of ^{222}Rn emanates from the soil during the sampling and sample manipulation. However, during deposition no connection between the deposition of parent and daughter radionuclides should be assumed, so until secular equilibrium has been re-established, measurement of daughter radionuclides does not provide information on parent radionuclide. This means that, for example, the quantity of ^{238}U cannot be accurately measured using only gamma ray spectrometry for a few months after deposition, but ^{235}U and ^{226}Rn can be accurately detected and measured (if their quantities are above the detection level).

The following figures present the comparison of laboratory measurements and in-situ measurements of ^{238}U (Figure 1), ^{214}Pb and ^{214}Bi (Figure 2), ^{232}Th (Figure 3), ^{137}Cs (Figure 4), and ^{40}K (Figure 5). All the figures show the mean value and 2σ confidence interval. Laboratory measurements shown are the results of the second set of laboratory measurements. Detector A is Canberra GC with 54% relative efficiency, and Detector B is Ortec Detective-Ex, with 17% relative efficiency. Location abbreviations are presented in Table 1.

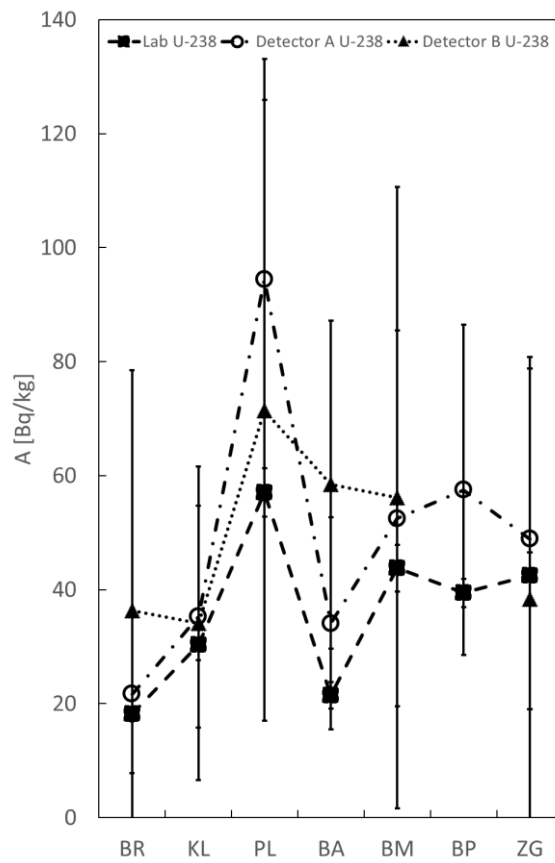


Figure 1: Results of ^{238}U measurements

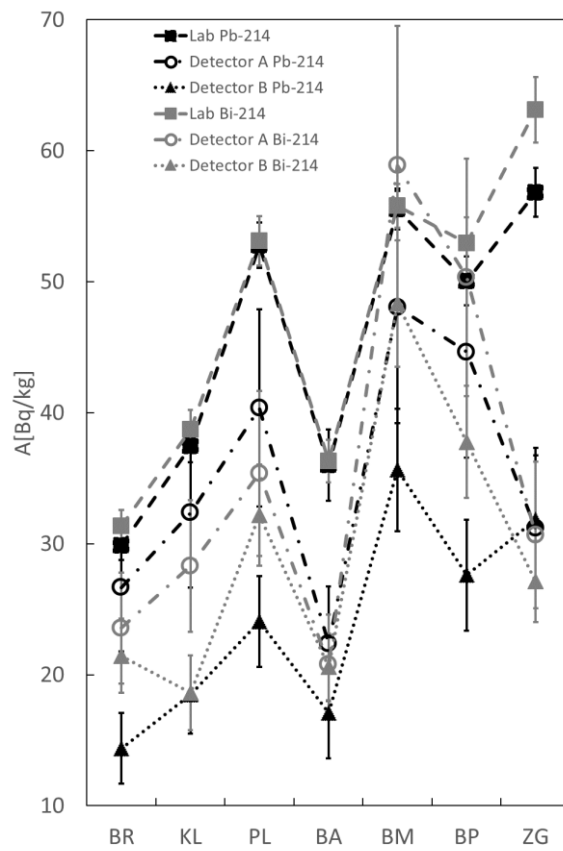


Figure 2: Results of ^{214}Pb and ^{214}Bi measurements

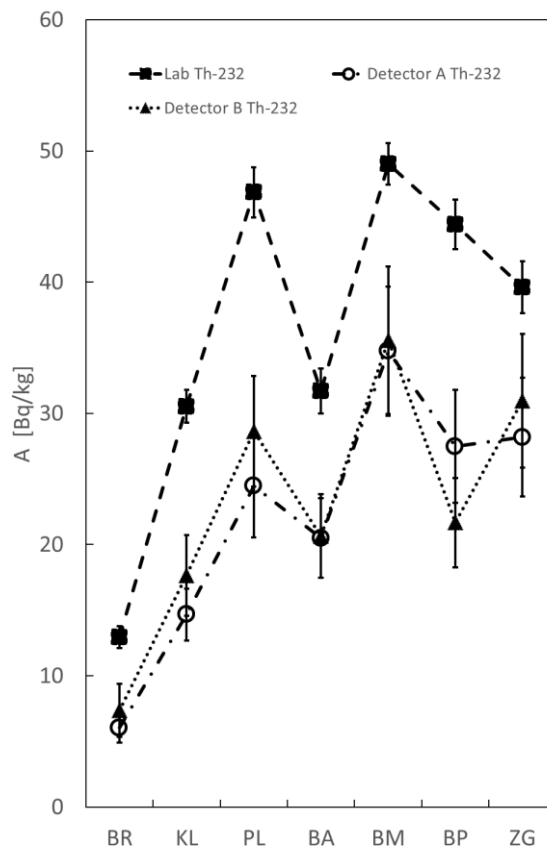


Figure 3: Results of ^{232}Th measurements

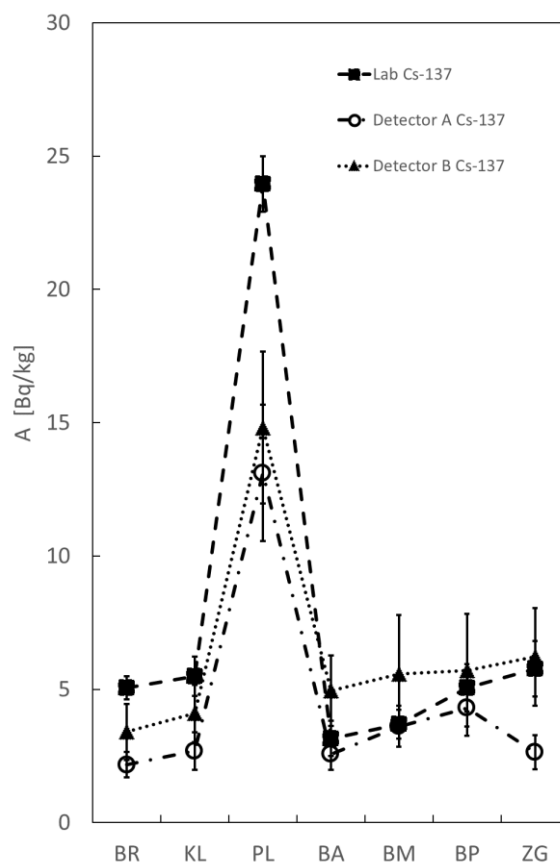


Figure 4: Results of ^{137}Cs measurements

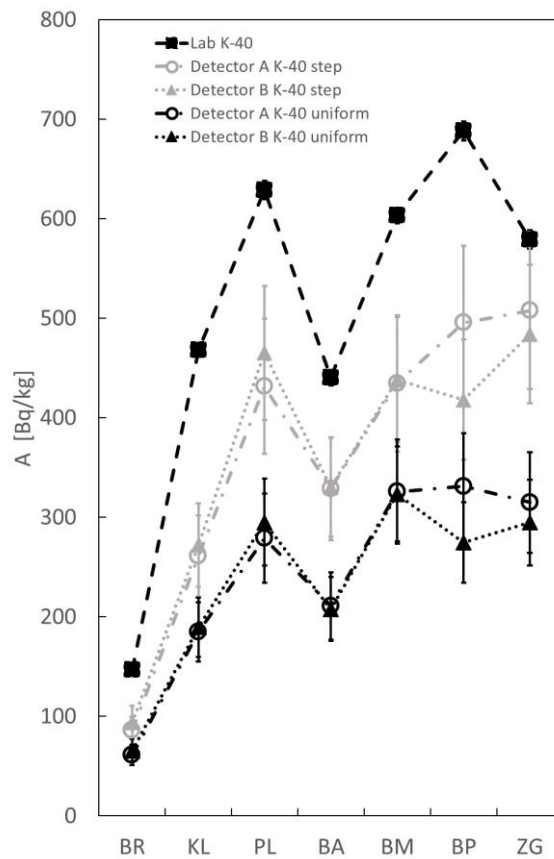


Figure 5: Results of ⁴⁰K measurements

The results presented in Figure 1 to Figure 5 show that 2σ intervals overlap for almost all in-situ measurements. For all radionuclides except ¹³⁷Cs, in-situ measurements made with Detector A gave higher values than in-situ measurements made with Detector B. In-situ measurements produced lower values than laboratory measurements. One reason may be that in-situ and laboratory measurements did not measure the same sample. Laboratory samples were cleared of foliage and gravel, which might have biased the measurements. Particularly big difference can be seen in ⁴⁰K measurements. Using uniform distribution, in-situ measurements produced only 47% of value of laboratory measurements, on average. Even if surface distribution is assumed (approximating the situation where foliage that was removed contains significantly more K than the soil below it), in-situ results only reach 70% of the laboratory results. This may indicate a calibration problem.

In-situ measurements have larger confidence intervals than laboratory measurements. For most of the energy spectrum (of gamma rays of interest) this presents little concern, especially in emergency situation. However, at energies around or below 100 keV, the effects of the Compton background are such that 5,000 s measurements cannot be relied on (as seen in Figure 1, where 2σ intervals were, on average, 56% for Detector A and 88% for Detector B).

4 CONCLUSION

Our findings suggest that in-situ gamma spectrometry using InSiCal software can replace laboratory measurements and should be a preferred method when time is a critical factor. When accuracy is more important than quickness, laboratory measurements provide a better option. InSiCal software successfully solves the problem of calibration of the detectors for in-situ measurements. The calibration procedure using InSiCal software can be applied on any HPGc detector irrespective of the manufacturer, efficiency or cooling method.

While in-situ measurements were reasonably accurate compared to the laboratory ones, some improvements are possible. Detecting low-energy gamma rays (below 100 keV) may be problematic because of high Compton background. It may be worth to do additional optimization of calibration times. Measurement times of 5000 s are generally sufficient, but may lead to higher detection levels for certain radionuclides. 5000 s measurements allow for a single team to measure two sites during the working hours, if travel to the location and back to the base is included, or up to four sites, if the detector stays in the field, and experts change every 8 hours. Longer measurements would, of course, increase the accuracy of the results and decrease the detection levels, but the trade-off is less total area covered.

REFERENCES

- [1] H.L. Beck, W.J. Condo, W.M. Lowder, Spectrometric techniques for measuring environmental gamma radiation, HASL-150, Health and Safety Laboratory, New York Operations Office, AEC New York, New York, 1964.
- [2] H.L. Beck, Environmental gamma radiation from deposited fission products, 1960-1964, *Health Physics* No. 3, pp. 313–322, 1966.
- [3] H.L. Beck, J. DeCampo, C. Gogolak, In Situ GE(Li) and Nai(TL) gamma-ray spectrometry, HASL-258, Health and Safety Laboratory (AEC) New York, New York, 1972.
- [4] International Atomic Energy Agency (IAEA), Characterization of radioactively contaminated sites for remediation purposes, IAEA-TECDOC-1017, 1998.
- [5] International Atomic Energy Agency (IAEA), Generic procedures for monitoring in a nuclear or radiological emergency, IAEA-TECDOC-1092, 1999
- [6] International Organization for Standardization, Measurement of radioactivity in the environment - Soil - Part 7: In situ measurement of gamma-emitting radionuclides, ISO 18589-7:2013.
- [7] A. Mairing, T. Vidmar, T. Gäfvert, J. Drefvelin, A. Fazio, InSiCal - A tool for calculating calibration factors and activity concentrations in in situ gamma spectrometry, *Journal of Environmental Radioactivity*, No. 188, pp. 58–66, 2018.
- [8] Y. Nir-El, G. Haquin, Minimum detectable activity in in situ γ -ray spectrometry, *Applied Radiation and Isotopes*, Vol. 55, pp. 197–203, 2001.