

Research Article

Advances in Materials Science and Engineering Assessment of Gamma Radiation Hazards Related to Geologic Materials: Comparison of Results by Field Gamma Spectrometry and Laboratory Methods

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Geologic materials are a potential source of external gamma radiation in buildings due to the presence of radioisotopes (⁴⁰K and isotopes from U and Th decay families). The contents of these radioisotopes can be evaluated by laboratory analyses that require the extraction of samples, with associated financial and time constraints, and it is also difficult to perform on materials already applied in existing structures. In this work, portable gamma spectrometry results are compared with laboratory analyses in terms of radioisotopes estimations, and the activity concentration index is calculated from them (which assess the conjoint contribution of the different radioisotopes to the external gamma radiation). The studied objects are rocks and their “in situ” weathering products. The results obtained indicate that despite several problems posed by field measurements with portable gamma spectrometry, namely, in terms of the geometric characteristics of the objects measured, this technique has the potential to identify materials with different hazard levels related to external gamma radiation.

1. Introduction

Natural ionizing radiation includes external and internal sources. While internal sources comprise naturally occurring radioisotopes that are taken into the human body, external ones are mainly cosmic rays and gamma radiation emissions from both terrains and building materials [1]. Isotopes present different activity values (radioactive decay rate, which is expressed in SI units as Becquerel, Bq, with $1 \text{ Bq} = 1 \text{ s}^{-1}$), and the ratio of the activity of the isotope to the total mass can be considered as an expression of its concentration, referred as activity concentration or specific activity (Bq/kg). The external gamma radiation related to terrestrial sources is mainly due to primordial radionuclides with radioactive decay half-lives that are approximately Earth’s age or older [2]. They are mainly ⁴⁰K and radioisotopes

of the radioactive decay series of ²³⁸U and ²³²Th (daughter isotopes of these radioisotopes) present in minerals and rocks used in building materials, being the main cause of indoor radiation exposure, which is about 40% greater than outdoor exposures in middle and high latitudes [3].

Different approaches have been considered to assess the dose that a person will receive from building materials in a given indoor space. The 2013/59/EURATOM directive of the Council of Europe (CEU [4]), which is directed to setting “basic safety standards for protection against the dangers arising from exposure to ionising radiation,” indicates the use of an activity concentration index (I) which is based on the sum of factors obtained by dividing the activity concentrations (or specific activities) of isotopes by constants that correspond to certain reference conditions in terms of room characteristics, materials application, and bulk density and exposition time.

$$I = \left(\frac{C_{\text{Ra-226}}}{300} \right) + \left(\frac{C_{\text{Th-232}}}{200} \right) + \left(\frac{C_{\text{K-40}}}{3000} \right), \quad (1)$$

where $C_{\text{Ra-226}}$ is the activity concentration of ^{226}Ra in Bq/kg, $C_{\text{Th-232}}$ is the activity concentration of ^{232}Th in Bq/kg, and $C_{\text{K-40}}$ is the activity concentration of ^{40}K in Bq/kg.

A value of one for this activity concentration index is indicated as a conservative screening tool with the main goal of distinguishing building materials with radioisotope levels that may have a contribution to external gamma radiation in excess of typical outdoor exposure above the reference level. This contribution of the materials will be different for different conditions of those considered to obtain the index, and the question of the typical outdoor exposure might also be a subject of discussion as it can show marked differences depending on the terrain characteristics [5].

There is hence an interest (that might be increased in the future as this directive is implemented) in assessing the concentrations of radioisotopes in building materials. However, direct measurements of isotopes activities are costly and time-consuming and can be considered destructive in the sense that they require sampling, which limits its application to building materials applied in existing structures. This limitation of the extraction of a sample also applies to other, eventually faster, laboratory techniques such as inductively coupled mass spectrometry (ICP-MS) and X-ray fluorescence (XRF) that allow the measuring of the elements amounts (from which specific activities of isotopes can be calculated, e.g., for U, Th, and K by the factors indicated in IAEA [6]).

Portable gamma-ray spectrometry (which is referred here as field gamma spectrometry—FGS) is a classical geophysical technique that has been used for geological exploration as well as for assessing the dose rate in terrains. It can be a fast procedure, and there are variants (such as the one that is considered here) that can be used by a one-person team. When FGS measurements are carried out outdoor on rock outcrops, a $2\pi r$ geometry (plain areas) is needed to avoid over or underestimation of the radioisotopes (K, U, and Th) and gamma radiation [6]. There is, consequently, interest in studying the usability of FGS at several perspectives related to building materials and their contribution to external gamma radiation:

- (i) To assess natural variability in quarries (which can be used to avoid portions of the rock mass with undesirable amounts of the radioisotopes)
- (ii) To control the products which are going out (again to avoid situations above certain values)
- (iii) To assess the potential hazards levels related to a given material applied in existing structures

There are several studies comparing field and laboratory analyses of radioisotopes, some of them regarding building materials, such as dose measurements in spaces and in relation to the applications of materials [7, 8]. In the studies of Mueller et al. [9] and Nuccetelli et al. [10], one can find detailed discussions regarding the issues concerning the comparison of laboratory and field measurements of radioisotopes. However, studies with the perspective that is presented here (assessment in relation to the activity concentration index as a screening

tool of building materials) are much scarcer. As far as we know, Stals et al. [11], concerning different types of building materials, and Döse [12], concerning concrete slabs with different types of rock aggregates, are the only works oriented to the assessment of activity concentration index of building materials with portable gamma spectrometry. These two publications report excellent positive linear correlations between portable and laboratory results. Stals et al. [11] found that ^{226}Ra and (especially) ^{40}K results by portable equipment were lower than results from laboratory analyses while ^{232}Th results were higher in the portable equipment results. The final result in terms of activity concentration index (I) was slightly lower for portable measurements (between <0.1 and 3.65) than laboratory analyses (<0.1 to 3.91), getting activity concentration index values between 0.15 and 2.04 for laboratory measurements and between 0.28 and 1.6 for the portable measurements. The results of laboratory analyses were lower for the lower I values and higher by 15% – 20% for the higher I range (above 1 in the laboratory results). These studies [11, 12] were both made with laboratory well-defined geometrical specimens, while our study considers outcrops with variable geometrical conditions (hence harder conditions for correlations), which are nearer the field conditions that one can find in quarry mapping. In a field study with a perspective near to this paper (but not in terms of the activity index as screening tool of building materials), Bochiolo et al. [13] found that field measurements gave higher measurements in all the considered radioisotopes (which means, consequently, higher activity concentration index values), in a study performed in an old mine tunnel (therefore in geometrical conditions that are more homogeneous than those considered here).

Our main goal in this paper was to assess the performance of field measurements by portable gamma spectrometry in rock outcrops (including their weathering products) by comparison with laboratory analyses of samples from the same places where the measurements were made.

2. Materials and Methods

2.1. Studied Geologic Objects. The present study considered samples from igneous and metamorphic rocks and their weathering products in outcrops near the towns of A Coruña (NW Spain) and Braga (NW Portugal), which were based on the available geological mapping (IGME [14, 15]; Ferreira et al., [16]), and our field studies can be described as gabbro (3 samples in A Coruña), leucocratic body in gabbro (1 sample in A Coruña), biotite-rich granites (4 samples in Braga), leucocratic granite (3 samples in A Coruña and 5 samples in Braga), and Silurian siliciclastic metamorphic rocks (7 samples in Braga).

2.2. Analytical Methods. The present work considers results from different methods to assess the concentration of radionuclides in such rocks and weathering products to assess their activity index and possible limitations in their use as building materials under the present European regulations established by the directive CEU [4]. The main aim of the work is to compare the assessments of radioisotopes concentrations in these geological materials, namely, in the perspective of the European directive through the results of



FIGURE 1: Picture of the portable gamma-ray spectrometer GF Instruments model GRS-2000 used for field measurements in this work. On the left side, the probe with the detector can be seen, and on the right side, the computer system with electric supply can be seen.

the concentration activity index (which can be considered a synthesis parameter of the contribution of the different isotopes in relation to a certain reference state).

2.3. Field Gamma Spectrometry (FGS). Gamma spectra were acquired in situ in the field with a portable gamma spectrometer GF Instruments GRS-2000, equipped with a probe BGO ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) having a $51\text{ mm} \times 51\text{ mm}$ detector and a shielded photomultiplier (Figure 1). The equipment allows measuring energies up to 3 MeV. Measurements were made by direct probe contact at outcrops with variable geometry. The recommendations given by the IAEA [6] for static measurements (recording a spectrum in a given point during a certain amount of time) were followed. Each acquired spectrum corresponds to a counting period of 3 minutes. This time period is recommended by the manufacturer to optimize time and precision. According to the manufacturer's specifications for low gamma-ray emissions, statistical uncertainties are estimated to be 6% for K, 30% for U, and 16% for Th by measuring for 180 s. This error is due to background radiation that is basically caused by three main sources: atmospheric radon and its daughters, cosmic rays, and instrument background. Moreover, the geometry of measured surface must be planar ($2\pi r$ geometry) being both possible surface roughness of the measured terrains and any nearby topographic feature possible causes of inaccuracies in the measurements.

From the obtained spectra (an example is presented in Figure 2), we have used some peaks to assess the contents of K (potassium, mass percent), eU (uranium equivalent, in mass parts per million or ppm), and eTh (thorium equivalent, also in ppm). While potassium contents are estimated from the peak of ^{40}K (energy range 1.366–1.564 keV), there are several radioactive isotopes in the uranium and thorium decay series, and hence, “equivalents” of uranium and thorium (eU and eTh, resp.) can be used, assuming secular equilibrium. Estimations of eU and eTh were made from peaks of the spectra corresponding to ^{214}Bi (energy range 1.57–1.959 keV) and ^{208}Tl (energy range 2.42–2.81), respectively, on the decay chain of uranium and thorium. The specific activities of ^{226}Ra (or ^{238}U), ^{232}Th , and ^{40}K were

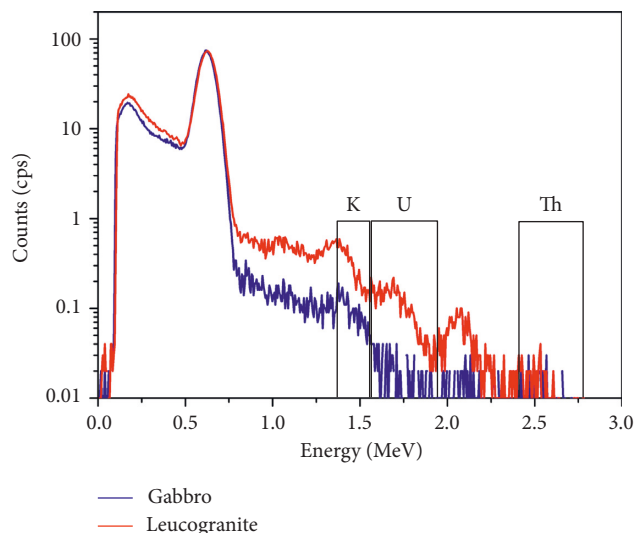


FIGURE 2: Example of two different field gamma spectrometry (FGS) spectra acquired on outcrops of gabbro (blue) and leucogranite (red) with indication of the energy windows for element estimations.

calculated from eU, eTh, and K with the conversion factors given in IAEA [6].

2.4. Sample Preparation for Laboratory Analyses. For laboratory analyses of the samples, the obtained samples were powdered at a particle size less than 0.5 mm and dried at 100°C to constant weight. Once dried, the powder was fired at 500°C for 20 hours to remove organic matter. A part of the powder was separated for elemental analyses and most of them for low background gamma spectrometry.

2.5. Laboratory Gamma Spectrometry (LGS). For laboratory gamma spectrometry (which is referred here as LGS), a mass of 250 g of each sample was placed in Marinelli polystyrene containers and sealed with a sellotape and left for more than 4 weeks in order to allow for Ra and its short-lived progeny to reach radioactive equilibrium. The radioactivity concentrations were measured in coaxial HPGe Canberra GR6022 of the “Servicio de Radioisotopos (CITIUS)” of the University of Seville (Spain). The detector was shielded with 10 cm thick lead on all sides including the top, to reduce the background contribution from the surroundings. The shielded volume is 180 cm^3 ; the relative efficiency of the detector is 60%, and it has a resolution of 1.4 keV at 2.3 keV for energies 122 keV and 1332 keV, respectively.

2.6. Element Analyses. The determination of both major and minor elements in the samples was obtained by X-ray fluorescence spectrometry (XRF). Powdered samples were measured in a Bruker-Nonius S4 Pioneer wavelength dispersive fluorescence spectrometer under helium purge at the University of A Coruña. To assess trace elements, inductively coupled plasma-mass spectrometry (ICP-MS) was used.

ICP-MS analyses were carried out in a Thermo Scientific™ ELEMENT XR™ ICP-MS which combines a dual mode secondary electron multiplier with a Faraday detector. Lithium metaborate fusion was used for sample preparation. The powdered samples were mixed with an equal amount of lithium tetraborate flux, placed in a carbon crucible, and fused at 1000°C in a furnace for 30 minutes. Once the melt was cooled, the resultant fusion bead was ground and dissolved in 100 mL of 4% HNO₃/2% HCl₃ solution which was then analysed by ICP-MS. The same factors (given in IAEA [6]) were used to convert U, Th, and K measurements to specific activities of ²²⁶Ra (or ²³⁸U), ²³²Th, and ⁴⁰K.

2.7. Data Treatment. Some notes on the statistical treatment are presented here (additional information can be found in statistical texts such as Dodge [17]). The statistical summary includes diverse measures of points of the value distribution: arithmetic mean (referred simply as mean from now on), harmonic mean, median, geometric mean, minimum, first (lower) quartile, third (upper) quartile, and maximum. While the minimum, mean, and maximum are certainly widely known, some notes are presented in relation to the other measures. The geometric mean is obtained by the n root of the product of the values (or by the arithmetic mean of their logarithms). The harmonic mean is obtained by the quotient between the number of elements of the data set and the sum of the inverse of each element of the data set. The quartiles are the points that divide a set of values into 4 equal parts. The middle quartile divides the data set into two equal parts and is equal to the median. The lower and upper quartiles correspond to the upper limit of the lower 25% and lower limit of the upper 25%. While it is not our goal to characterize the distribution of the sets of values from the studied rocks, dispersion statistics (standard deviation, standard error of the mean, and interquartile range) are presented for the reader to assess the dispersion of the data sets under study, which could be useful for comparison. While we will not discuss the question of the normal distribution of such a geologically heterogeneous set of measurements, one can highlight that, for a normal distribution, the 95% confidence limits of the mean are obtained by the mean more or less the product of the standard deviation by 1.96 (for a higher confidence limit, the error interval is, of course, higher).

As for there are groups of samples from some rock types and since our goal here is to compare techniques and not the geochemical characterization of the geological materials, data treatment is based on robust statistical techniques based on nonparametric approaches (that do not assume any premise concerning the shape of the data distribution or its parameters). The correlation between different measures is assessed by the rank-based Spearman correlation coefficient, which does not require each studied value set that presents a normal distribution, which is not affected by the specific values nor is it affected by arithmetic operations with constants (e.g., dividing by a constant) since it does not depend on the variance of the value sets. One will also present results of the coefficient of determination that assess

the fit of the data to a proposed regression equation (it is a measure of the contribution of a given factor to the variance of another under a given statistical model such as a regression straight line). The distribution of the values of the concentration activity index (I) obtained from results of each analytical technique is represented by a boxplot, representing the lower, middle, and upper quartile and raw data. Since the realization of several statistical tests with the same data set can cause an error I type inflation, we limit the use of other statistical tests to the comparison of the calculated values of the concentration activity index. Following the advice of the American Statistical Association (Wasserstein and Lazar [18]), the obtained p values are reported. Comparisons of I values from the different analytical techniques are based on two nonparametric statistical tests for paired statistical samples: the sign test and the Wilcoxon test. The sign test is based on the proportion of the signs of the differences between the paired results. The Wilcoxon test is based on the ranking of the differences between the compared statistical samples.

All statistic tests, the boxplot and the mean and standard error plots were prepared with *Statistica 11* (*Statsoft*).

3. Results and Discussion

Figure 3 presents a combined boxplot (quartiles) and raw data plot of the logarithms of ratios, in relation to estimations from field measurements, of laboratory estimations of ²²⁶Ra, ²³²Th, and ⁴⁰K specific activities by laboratory gamma spectrometry and those calculated from uranium, thorium, and potassium quantifications (the first two by MS-ICP and the later by XRF). It can be seen that, in almost all the cases, the ratios of laboratory estimations to field estimations are below one (negative logarithms), indicating that, for the different radioisotopes, estimations from field measurements are higher than laboratory ones.

Bivariate plots of laboratory measurements against the field ones as well as the values of the activity concentration index calculated from the different sets of isotopes estimations (laboratory and field) are plotted in Figure 4. It can be seen that there are clear trends toward correlations between the different sets of measurements, but there is some wider scatter in the case of ⁴⁰K estimations as shown by the coefficient of determination values, which are lower than those obtained by Stals et al. [11]. The plots also include the line of equality between plotted variables, showing that, in general (with few exceptions), values of radioisotopes estimations from laboratory measurements are less than those from field measurements and that this happens in the comparison of calculated values of the activity concentration index also (in this case, only one value obtained from field estimations is slightly lower).

The p values for the Spearman coefficients calculated for I value estimations from the three sets of analyses are presented in Table 1, showing that they markedly below 0.05 (the highest p value equals to 0.000001). It can also be seen from the Spearman coefficients in Table 1 that the correlation between the estimations from the laboratory analyses

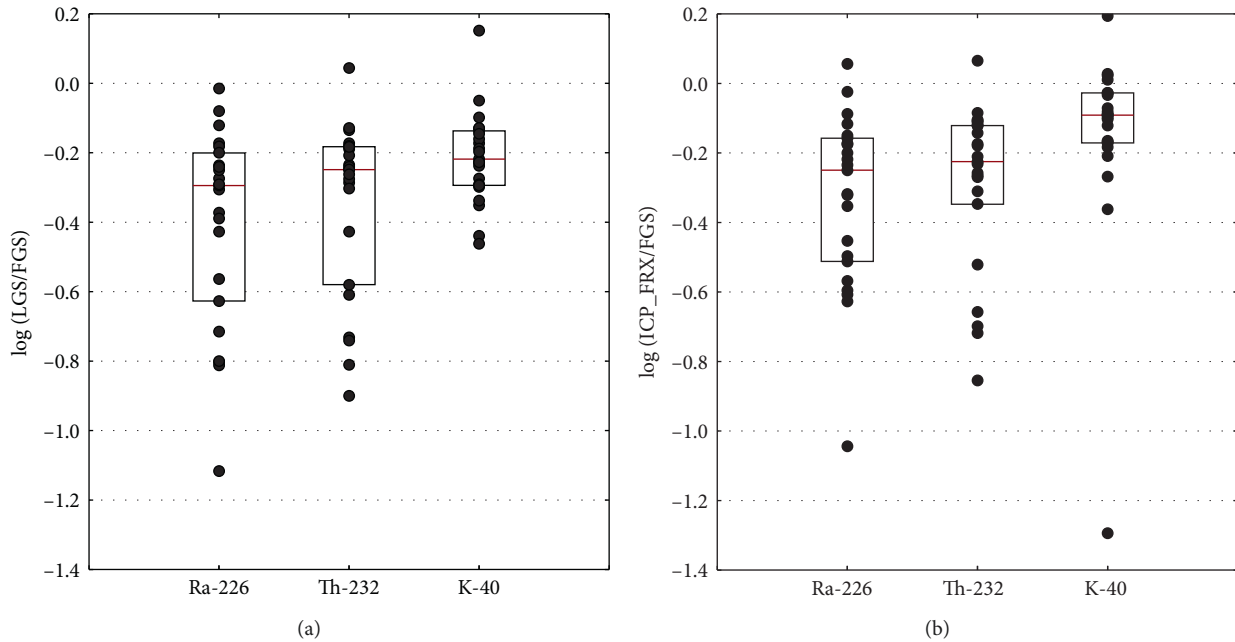


FIGURE 3: Combined boxplots (quartiles) and raw data plots of logarithms of radioisotopes estimations from laboratory analyses to those from field measurements: (a) comparison of laboratory gamma spectrometry (LGS) to field gamma spectrometry (FGS); (b) comparison of estimations from ICP and XRF to FGS.

is better than the correlations of the laboratory analyses with the field measurements.

Table 2 presents the univariate statistics of the activity concentration index (I) values calculated from the three procedures. It can be seen that all location measures are higher in the field set than in the ICP/XRF set that, in turn, are higher than those from the laboratory gamma spectrometry measurements. It can also be seen that I values based on laboratory analyses are always nearer each other than in relation to I values based on field measurements. One can also highlight the relation of the statistics presented in Table 2 in relation to the screening criteria indicated in the European directive ($I = 1$). Most of the average estimations (mean, geometric mean, and median) are above 1.0 for the set of I values from field measurements but does not happen in any of the averages for I values from laboratory measurements. The lower quartile of I values based on field measurements is very near this screening value. These relations are also illustrated in the plot of Figure 5(a) that combines a boxplot and the plotting of raw data. This plot also shows that the number of results that exceeds the $I = 1$ condition is clearly much higher for the field estimations.

The nonparametric statistical tests in the comparison of these paired values (Table 3) show very low p values (the higher one being 0.0005) and being noticeable that the comparison of field values with laboratory values produces lower p values. However, the statistical assessment of higher mean in the field set deserves more caution since for $p = 0.05$, the intervals of mean more or less 1.96 times the standard error of the mean of the different data sets overlap, as can be seen in the plot of Figure 5(b). Statistical non-parametric tests of paired samples also support these relations, with low p values, namely, in the comparison of I

values from field and laboratory measurements (with the highest being $p = 0.00003$).

To explain the scatter observed among ^{40}K estimates obtained by both XRF and LGS, two causes can be considered. On the one hand, the ^{40}K and ^{214}Bi peaks (i.e., used to assess the U concentration) are very close in the gamma-ray spectrum. The FGS equipment has low resolution when compared with HPGe used in LGS. Hence, when one of the ^{214}Bi or ^{40}K peaks is high, it can overlap to part of the nearby peak providing an overestimation of the other radioisotope [6]. To check if this effect is a cause of scatter in our results, we have compared eU and eTh estimates from FGS and the ratio of K assessed from FGS to K measured by XRF and LGS (Figure 5). The ratios K (FGS)/K (XRF) and K (FGS)/K (LGS) are near 1 when the estimates of K fit, being higher when the K (FGS) overestimates the K (XRF) or K (LGS) estimates. If the overlap of a high ^{214}Bi peak with the ^{40}K peak is the cause of K (FGS) overestimation, the ratios should be higher and above 1 when higher eU is measured. However, this is not observed when the K/K ratios are plotted versus eU (Figure 6). In fact, when the ratios are plotted versus the ^{208}Tl peak estimates (eTh), the results are similar to those in the former plot, being negligible any peak overlaps on the K and eU estimates by FGS. Anyway, it must not be forgotten that the large uncertainty of the FGS (6%, 30%, and 16% for K, U, and Th, resp.) hinders the assessment of this cause of scatter, and a possible larger data set could be required to get concluding results. Anyway, the disequilibrium in the ^{238}U decay chain should be expected in weathered rocks but not in nonweathered or slightly weathered rocks. In such rocks, secular equilibrium is observed [19]. Moreover, the weathering processes and derived disequilibrium are not necessarily similar in different rocks

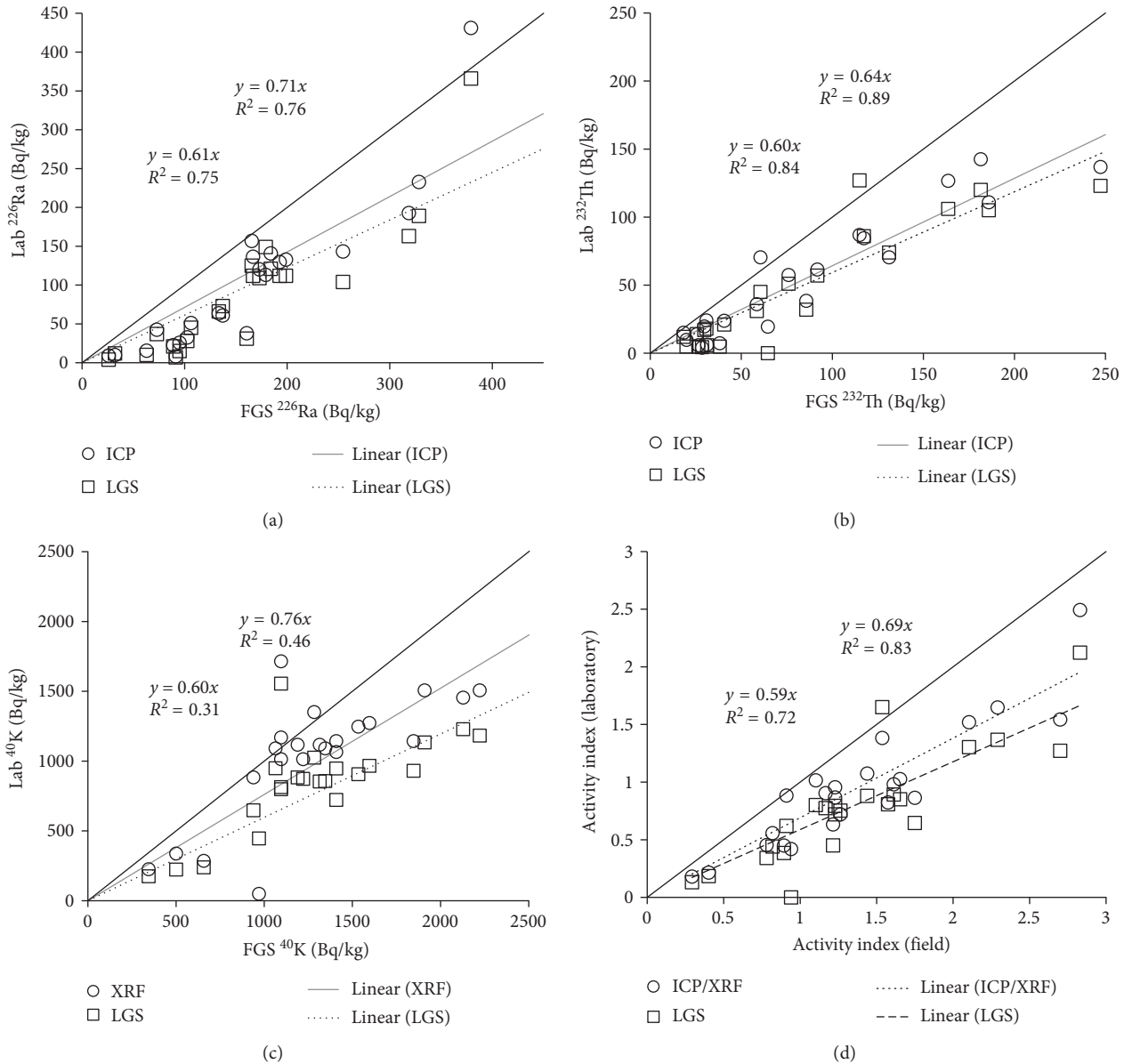


FIGURE 4: Plots of the results from laboratory analyses (ICP, XRF, and LGS: laboratory gamma spectrometry) against field estimations: (a) ^{226}Ra activity concentration; (b) ^{232}Th activity concentration; (c) ^{40}K activity concentration; (d) activity concentration index calculated from the isotopes estimations.

TABLE 1: Spearman correlation coefficients between the calculated values of the concentration activity index (I) from different procedures for estimation of isotopes specific activity and related p values.

	N	R	p value
I (field) and I (ICP/XRF)	23	0.83	1.08×10^{-6}
I (field) and I (LGS)	23	0.87	5.71×10^{-8}
I (ICP/XRF) and I (LGS)	23	0.95	3×10^{-12}

LGS: laboratory gamma spectrometry.

(e.g., gabbro and granite), being necessary to consider any rock type to assess this factor as a possible cause of the scatter results. Other sources of inaccuracy must be taken into account in the estimates, as it is not possible to assess its

effect on the results. A flat geometry is required for FGS measurements ($2\pi r$). Although planar surfaces have been chosen for the measurements, the work areas are not completely flat, being surface roughness and topographic features a possible cause of such inaccuracies and scattering.

4. Conclusions

Field portable gamma spectrometry measurements in igneous and metamorphic rocks and their weathering products showed good ordinal correlations with two laboratory techniques of assessment of radioisotopes specific activity, suggesting that it can be used to rank materials in terms of radiological hazard concerning the contribution of materials to the external gamma radiation dose. The results tend to be

TABLE 2: Statistic measures of the activity concentration index (I) sets from different procedures for estimation of isotopes specific activity and related p values.

	Minimum	Maximum	Mean	G.M.	H.M.	Med.	S.D.	S.E.	L.Q.	U.Q.	I.Q.R.
I (field)	0.29	2.83	1.38	1.22	1.04	1.23	0.65	0.13	0.91	1.65	0.74
I (ICP/XRF)	0.18	2.49	0.94	0.80	0.65	0.88	0.52	0.11	0.56	1.07	0.52
I (LGS)	0.13	2.12	0.80	0.69	0.52	0.77	0.48	0.10	0.44	0.89	0.45

G.M.: geometric mean; H.M.: harmonic mean; Med.: median; S.D.: standard deviation; S.E.: standard error of the mean; L.Q.: lower quartile; U.Q.: upper quartile; I.Q.R.: interquartile range.

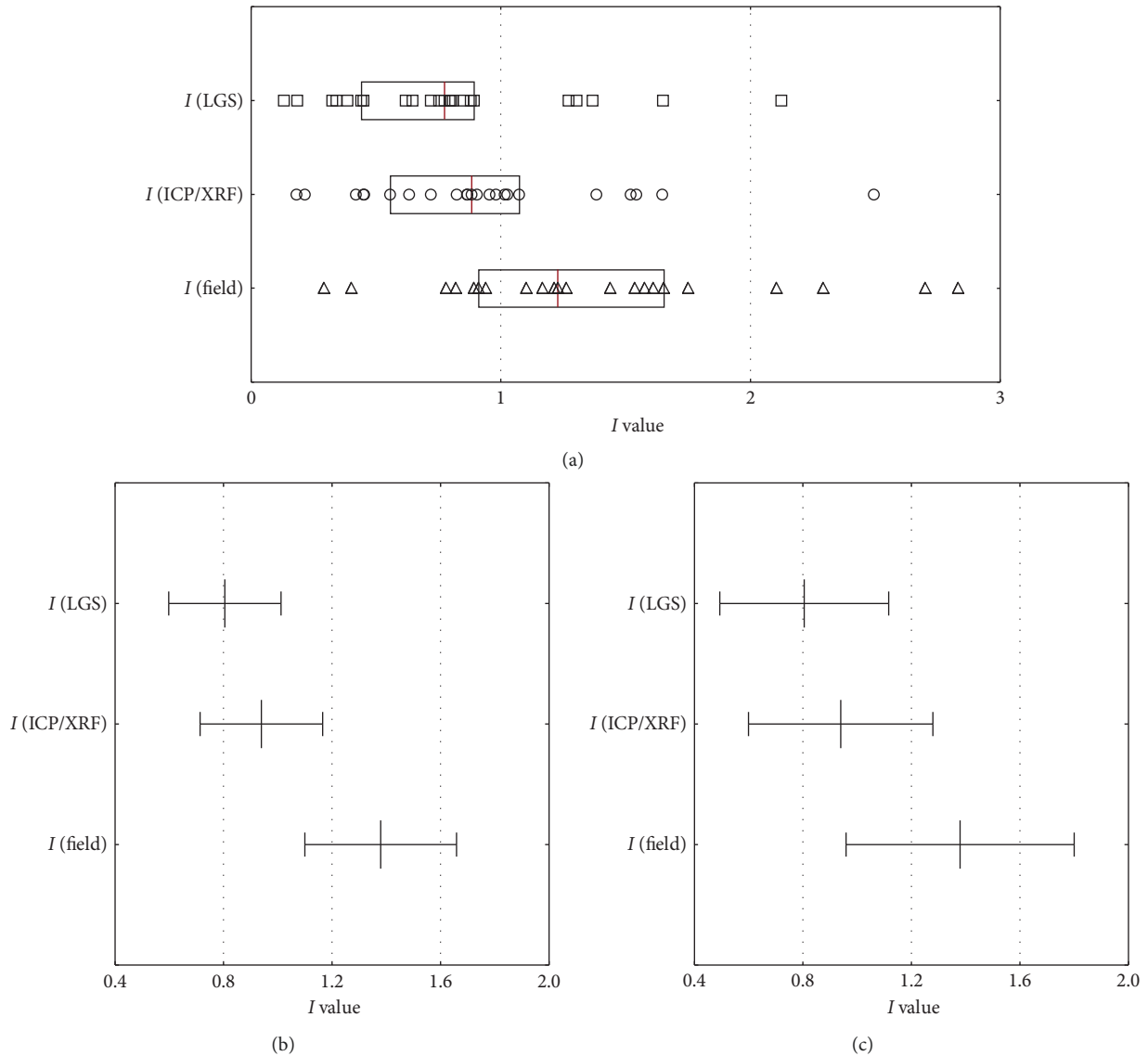


FIGURE 5: Plots for comparison of the activity concentration index values (I) calculated from the laboratory analyses (ICP, XRF, and LGS: laboratory gamma spectrometry) and field estimations: (a) boxplot (quartiles) and raw data plot; (b) mean and standard error plot for 95% confidence; (c) mean and standard error plot for 99.5% confidence.

TABLE 3: Results (p values) of statistical tests for paired comparisons of estimations of the activity concentration index (I) values.

	N	Sign test p value	Wilcoxon p value
I (field) and I (ICP)	23	0.000004	0.00003
I (field) and I (LGS)	23	0.000030	0.00003
I (ICP) and I (LGS)	23	0.000175	0.00047

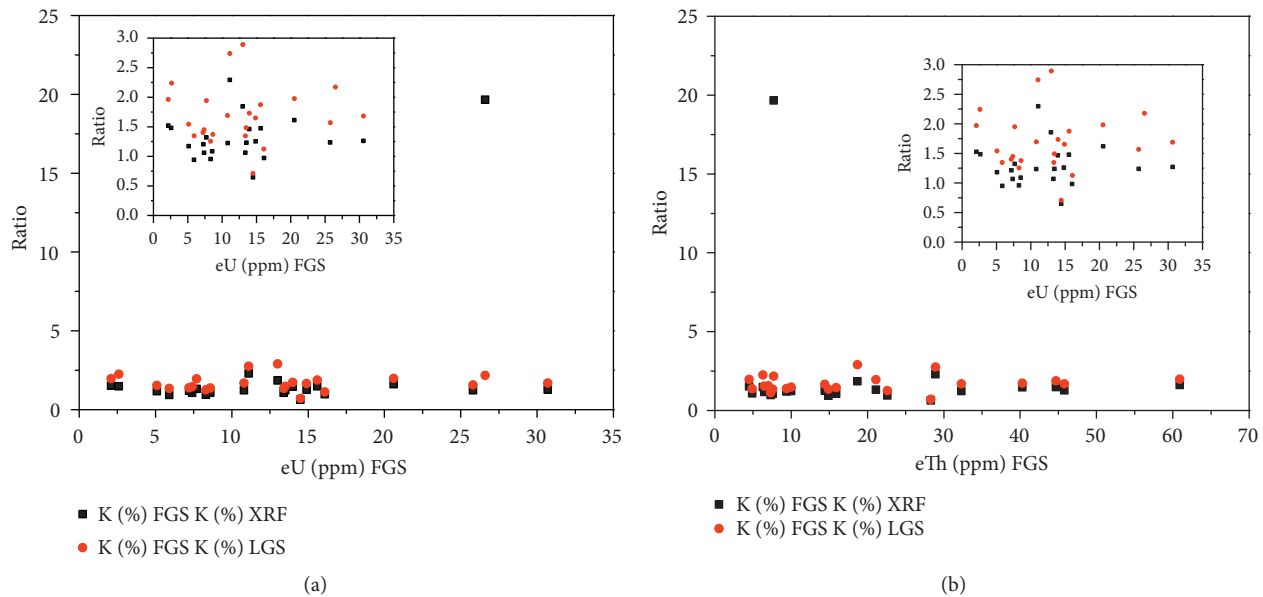


FIGURE 6: Plots for the comparison of estimates of eU (a) and eTh (b) versus K (%) ratios obtained by FGS, XRF, and LGS. The K ratios correspond to K (FGS)/K (XRF) and K (FGS)/K (LGS). The insets show a detail of the whole plots in the range 0–3 of the ratio and 0 to 35 ppm for eU and eTh. No correlation is observed between the data.

higher in the case of field measurements, which is reflected in a higher set of results above the guideline criteria $I = 1$. While one should read carefully in this matter, namely, given the limited sample set considered, these results strongly suggest that portable gamma spectrometry could be a conservative tool in assessing external gamma radiation posed by materials and, if this is confirmed, be used as a screening tool to separated samples to be sent for laboratory analyses.

It is possible that these outcrop measurements could work in a conservative way given the great amount of the geological substance that is present in the measuring point. But it is admitted that portable measurements might not be as efficient in the case of measuring specimens of materials as the effect of the surroundings could behave a higher impact (due to the smaller amount of the measured object).

Data Availability

Results of measurements and laboratory analyses are available from the authors upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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Supplementary Materials

Three tables with the results of the different samples and indication of the associated errors are presented as supplementary material. (*Supplementary Materials*)

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