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INTERCOMPARISON EXERCISE ON EXTERNAL GAMMA DOSE RATE UNDER FIELD CONDITIONS AT THE LABORATORY OF NATURAL RADIATION (SAELICES EL CHICO, SPAIN)

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The last nuclear accident in Fukushima nuclear power plant has increased the necessity for measuring radiation in the environment. Therefore, radiation monitors providing results traceable throughout the country become essential and it is very important to test them under the same environmental conditions. The first intercomparison of natural radioactivity under field conditions was held in Saelices el Chico (Salamanca, Spain) in May 2011, including an exercise on environmental dose rate. This article presents the results achieved by 19 instruments belonging to 12 institutions from 7 different countries. The tested detectors are proportional counters, ionisation chambers, Geiger–Müller and scintillators measuring dose rate in three stations with reference values from 110 to 1800 nGy h^{-1}. All the results were given in terms of air kerma (nGy h^{-1}) and the measurements show agreement within 25 % in all the sites. Evaluation criteria based on accuracy and statistical uncertainty were also carried out and 25 % of participants passed the test in all sites.

INTRODUCTION

The demand for measuring radiation in the environment has been increased since the last nuclear accident in Fukushima nuclear power plant[(1)]. The need for this type of measurements has been essential during the last decades especially after nuclear events such as Chernobyl accident or the Three Miles Island's event. These accidents had transboundary impacts[(2)], and hence, it is important to have radiation monitors available whose results can be traceable throughout the country. In addition to the radiation in the environment coming from artificial sources, the natural component of environmental radiation is also important. Environmental external radiation contains two main components, terrestrial radiation and secondary cosmic radiation[(3)]. Terrestrial radiation consists of photons coming from the radioactive isotopes present in the Earth's crust. These isotopes are mainly ^{40}K and those from the radioactive decay series of uranium and thorium, and the photon energy ranges from a few keV to 2.6 MeV, which corresponds to ^{208}Tl. As a consequence, the contribution of terrestrial component is very dependent on the local geology. Secondary cosmic radiation is originated by nuclear reactions, which involve different types of particles (neutrons, protons, pions and photons) and its contribution increases with the altitude.

Many different instruments are currently being used to measure the environmental dose rate exhibiting various designs, features and calibration techniques. Furthermore, there is no general agreement in the radiation quantity that should be addressed. As a result, the comparison of data from different instruments is far from simplicity and a consideration of all these aspects must be made to perform a valid comparison. To this aim, it becomes very important to test these devices under the same environmental conditions. This objective is usually achieved by means of intercomparison exercises either under laboratory or under field conditions. Such exercises provide unique opportunities for participants to check the adequacy of their home calibrations and field measurements[(4)]. The recommendations of the EURADOS working group 12[(5)] propose that the organisations responsible for environmental measurements should participate in national or international intercomparisons.

The Radon group from University of Cantabria in Spain has established a site where the values of natural radioactivity allow testing instruments and detectors under typical variations of temperature, humidity and atmospheric pressure, which can be found in occupancy places (dwellings and working places). Such a place is located in an old uranium mine site and the first intercomparison exercise was

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carried out at that site under field conditions in May 2011. The old uranium mine site was shut down in 2004. Since then, the restoration process has been taking place. During these activities, one of the buildings used for the treatment of uranium mineral was chosen to be a laboratory of natural radiation (LNR) in order to be used for the calibration and testing of instruments and detectors for the measurement of natural radiation. The Radon Group in collaboration with ENUSA was in charge of the activities of adaptation of this building to the new situation. Radon concentrations and external gamma radiation are subjected to daily variations due to changes in environmental conditions. Thus, the LNR is the perfect place for the performance of experiments devoted to the analysis of environmental radioactivity as well as a location for testing instruments specialised for the measurement of natural radiation.

The first intercomparison on natural radioactivity under field conditions was held in the old uranium mine of ENUSA in the municipality of Saelices el Chico (Salamanca, Spain) from 23 to 27 of May 2011. The meeting was organised by the Radon group of University of Cantabria (Spain) and the main objective of this event was to test different instruments and detectors for the measurement of radon gas and external gamma radiation (dose rate) in real conditions in a place where the levels of natural radiation are quite high. A total number of 45 participant institutions (~100 persons) from the following countries decided to take part in the exercise: Austria, Belarus, Belgium, Czech Republic, France, Germany, Hungary, Italy, Norway, Poland, Romania, Slovenia, Spain, Sweden and the UK. The institutions involved were universities, reference laboratories and commercial companies whose main activities are related to the measurement of natural radiation and radon gas and external gamma radiation in particular.

For the particular case of intercomparison of environmental dose rate instruments, the exercise was organised within the framework of the international intercomparison under field conditions. Twelve institutions followed the intercomparison on environmental dose rate from seven countries. In this work, the main results obtained in the activity as well as the characterisation of the working field are summarised. The aim of the activity was to test radiation monitors in three different areas where the air kerma rate has a wide range of values from 110 to 1800 nGy h^{-1}. Before the exercise, an introductory keynote lecture was given explaining the measurement of environmental dose rate as well as some examples of previous international intercomparisons on the same subject[6–8].

The quantity recommended by ICRP[9] for use in radiological protection, including the assessment of risks in general terms, is the effective dose. In the same way, the Council Directive 96/29/EURATOM[10] requires employing the same radiation risk related quantity, the effective dose. However, effective dose is not a measurable quantity and, therefore, the operational quantity ambient dose equivalent to a depth of 10 mm of soft tissue, $H^*(10)$, is recommended as operational radiation quantity for environmental monitoring of strongly penetrating radiation. For instance, the participants in the second EURADOS intercomparison of dose rate detectors employed in early-warning network systems were asked to provide their results in terms of $H^*(10)$[6]. It is also common to find environmental dose rate monitors that are still calibrated in terms of air kerma, absorbed dose in air and even in exposure or photon dose equivalent, which have the benefit that they are purely physical quantities whose calibration can be provided directly in metrology laboratories, without using any conversion factors or conventions in the definitions that could change in the future. In this exercise, participants were asked to provide the results in terms of air kerma rate in nGy h^{-1}, using if necessary the adequate conversion factors for 661 keV photons (^{137}Cs)[4, 11].

RADIOLOGICAL CHARACTERISATION OF TESTING POINTS

The organisers selected three areas as working field for the measurement of external ambient dose in the facilities of ENUSA located in Saelices el Chico (Salamanca, Spain). The radiological characterisation of the areas was done by the Spanish Metrologic world reference centre, CIEMAT (Centre for Energy-Related, Environmental and Technological Research), at the end of April 2011. The instruments used for establishing the reference levels were calibrated at the Dosimetry Standards Laboratory of CIEMAT. These instruments consist of two high-pressure ion chambers Reuter–Stokes models RSS-112 and RSS-131, and they are the worldwide reference devices employed in external ambient dose assessment. They were calibrated in terms of exposure by means of the method 'Shadow/Shield' using a point source of ^{60}Co. A conversion factor of 8.764 nGy μR^{-1} from exposure to air kerma was employed.Station 1 was located in a hillside composed of uranium tailings modified for the exercise. A surface of 25 m^2 (5 × 5 m^2) was excavated up to a depth of 1.5 m and refilled with soil containing a mixture of natural radionuclides whose average activity is of the same order as other Spanish soils and much lower than that of the surrounding areas in the same location. The measurement points were located at the vertices of this square and also at the geometrical centre (Figure 1). Station 2 was situated 500 m separated from Station 1 in a dry rock of uranium tailing materials pending of restoration, where a flat area of 25 m^2 was established for dose rate and gamma

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spectrometry measurements. The central point was marked as point A and the vertices as points B–E. The third area named point X was located 50 m to the north of the first station in an area composed of uranium tailings without modification and also characterised by CIEMAT.

The position of each measuring point was determined with a Javad global positioning system model Gismore with 1 m of precision both altitude and latitude above sea level. Geographical coordinates are expressed in terms of universal transverse mercator (UTM) zone (29). The radiation determinations were done with the equipment at 1.00 ± 0.03 m above ground level for 30 min. Each instrument acquired 300 readings of exposure rate. In the case of Station 2, the measurement time was 10 min due to the high radiation in this area. One hundred and fifty readings were acquired in this station.

Table 1 shows that the two Reuter–Stokes instruments have good agreement in the readings in both stations. The homogeneity is quite good in points A–E from Station 1 with an average value of 110 ± 5 nGy h^{-1}. The same homogeneity is observed in Station 2 with an average value of 1900 ± 50 nGy h^{-1}. Table 2 shows the reference values used for the intercomparison exercise, including position X.

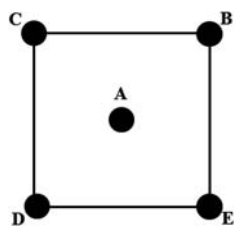

Figure 1. Distribution of measurement points located at Station 1.

In situ gamma spectrometry was also performed in the three locations with portable high-purity germanium detectors in order to estimate the activity concentration in soil. While the three sites present similar concentrations on ^{40}K (0.8–1.2 Bq g^{-1}) and natural thorium series (0.1–0.5 Bq g^{-1}), due to the differences in soil composition mentioned above the natural uranium series shows 0.05 Bq g^{-1} in Station 1, 0.3 Bq g^{-1} in point X and 5 Bq g^{-1} in Station 2. These measurements were performed at 1.00 ± 0.03 m above the ground and the efficiency calibration was calculated assuming a homogenous activity distribution in the soil and a standard soil composition named 'Dirt 1'[12].

These differences cause increases in the dose rate and changes in the photon spectra that could affect the monitor readings because of their photon energy response.

Prior to the exercise, a form was distributed where participants had to indicate the description of the instrument used (model, manufacturer and detector's type) and the calibration used (quantity, nuclide and photon energy). Each participant was identified with an alphanumeric code in order to keep anonymity (in those cases when the laboratory submitted two or more sets of detectors or instrument, the coding system is modified to allow distinguishing of different sets). Four different radiation monitors were used: scintillator, Geiger–Müller (GM), ionisation chamber and proportional counter. Figure 2 shows a pie chart where it can be observed that the majority of the detectors used correspond to scintillator and GM. Table 3 shows the description of the measurement equipment used by each participant, a total of

Table 1. Air kerma reference levels in Stations 1 and 2.

	Point					Average (A–E)
	A	B	C	D	E	
Station 1						
X-UTM, m	701704.7	701707.3	701708.5	701700.9	701700.8	
Y-UTM, m	4501191.2	4501196.1	4501187.0	4501186.5	4501195.6	
Z, m	719.3	719.1	719.1	720.0	719.1	
Reuter–Stokes 1, nGy h^{-1}	111 ± 4	112 ± 4	110 ± 5	110 ± 4	107 ± 4	110 ± 2
Reuter–Stokes 2, nGy h^{-1}	112 ± 4	113 ± 5	110 ± 4	111 ± 4	105 ± 4	110 ± 3
Station 2						
X-UTM, m	701541.8	701539.6	701537.7	701542.8	701545.6	
Y-UTM, m	4502440.1	4502436.4	4502440.6	4502442.9	4502439.0	
Z, m	719.2	719.5	720.1	720.0	719.3	
Reuter–Stokes 1, nGy h^{-1}	1862 ± 12	1831 ± 15	2008 ± 17	1901 ± 12	1815 ± 28	1883 ± 69
Reuter–Stokes 2, nGy h^{-1}	1803 ± 66	1785 ± 67	2005 ± 31	1908 ± 30	1798 ± 68	1860 ± 85

Uncertainty is expressed with k=2 (k means the coverage factor)

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Table 2. Air kerma reference values at Stations 1 and 2 and point X.

Point	Reference value (nGy h^{-1})	Uncertainty (nGy h^{-1})
Station 1	110	5
Station 2	1870	50
Point X	173	5

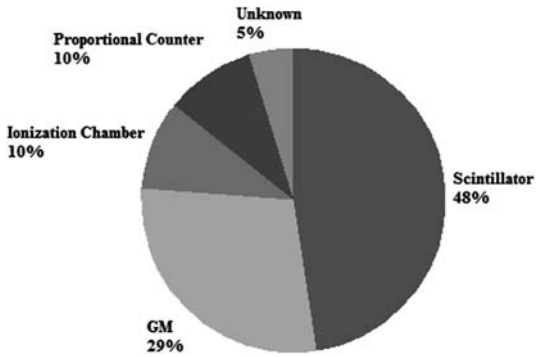

Figure 2. Pie chart showing the distribution of the different radiation monitors used in the intercomparison.

19 dose rate monitors (some institutions participated with more than one instrument).

RESULTS

The participants performed several dose rate measurements in different positions in Stations 1 and 2 and point X and the results can be seen in Table 4. Figure 3 shows the results obtained in Station 1 (mean value and standard deviation as bars). These error bars can indicate the monitor sensitivity, being smaller when the instrument is more sensitive. The solid grey line indicates the reference value provided by CIEMAT and the solid black line indicates the average of the mean values provided by all the participants. Dashed grey lines indicate one standard deviation up and down the average of the mean values. With some exceptions, the monitors present acceptable sensitivity as estimated from the error bars. It can also be seen that the average of the participant mean values is very close to the reference value and its relative standard deviation is ~25 %. While an ANOVA test shows that all the results are normally distributed and they are comparable, the data of seven monitors are further than 25 % from the average and the reference values. This could be related to the calibration factor but instrument Lab4 shows an important uncertainty, which could be related to poor sensitivity. One participant (noted as Lab13) repeated the measurement with four different NaI scintillator detectors, because they experienced several problems related to temperature influence (temperature range was 10–40°C) and other errors.

Figure 4 shows the results of the measurements for Station 2. Here most of the participants obtained

Table 3. Description of the measurement equipment used (GM), manufacturer and quantity used for the calibration of the instrument.

Code	Detector type	Quantity (calibration nuclide)
Lab1	GM	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab2	GM	Air kerma (^{137}Cs)
Lab3	GM	Not available
Lab4_a	Scintillator	Air kerma
Lab4_b	Scintillator	Air kerma
Lab4_c	Scintillator	Air kerma
Lab5	GM	Ambient dose equivalent $H^*(10)$, photon dose equivalent H_x
Lab7	Scintillation NaI (Tl)	Ambient dose equivalent (natural pechblend and thorium source)
Lab8	Energy-compensated GM detector and a beta shield	Exposure (X) (^{137}Cs)
Lab9	GM	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab10	Ion chamber/ proportional counter	Photon dose equivalent $H_x(10)$ (^{137}Cs)
Lab11_a	1 × 1 NaI scintillator	Exposure (X) (^{137}Cs)
Lab11_b	3 × 3 NaI scintillator	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab12	Proportional counter	Ambient dose equivalent $H^*(10)$ (^{137}Cs and ^{60}Co)
Lab13_a	Scintillator	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab13_b	Ion chamber/ proportional counter	Photon dose equivalent $H_x(10)$ (^{137}Cs)
Lab13_c	Scintillator	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab13_d	Scintillator	Ambient dose equivalent $H^*(10)$ (^{137}Cs)
Lab13_e	Scintillator	Ambient dose equivalent $H^*(10)$ (^{137}Cs)

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Table 4. Results of intercomparison external gamma dose rate obtained by the participants in the international intercomparison under field conditions (Saelices el Chico, Spain, 2011).

Code	Air kerma rate (nGy h^{-1}), Station 1	Air kerma rate (nGy h^{-1}), Station 2	Air kerma rate (nGy h^{-1}), point X
Lab1	112 ± 15	1932 ± 111	100 ± 8
Lab2	180 ± 10	1917 ± 194	265 ± 7
Lab3	175 ± 7	2133 ± 162	260 ± 50
Lab4_a	176 ± 45	2429 ± 177	209
Lab4_b	99 ± 6	2182 ± 259	174
Lab4_c	149 ± 10	2624 ± 198	181
Lab5	127 ± 8	2042 ± 123	193 ± 12
Lab7	132 ± 40	3237 ± 141	261 ± 14
Lab8	138 ± 8	2262 ± 103	204 ± 25
Lab9	160 ± 29	2197 ± 154	317 ± 63
Lab10	110 ± 5	1800 ± 50	173 ± 6
Lab11_a	65 ± 3	1643 ± 136	129 ± 1
Lab11_b	132 ± 7	2958 ± 477	129 ± 1
Lab12	140 ± 20	2500 ± 300	230 ± 30
Lab13_a	20 ± 6	1152 ± 113	35 ± 2
Lab13_b	100 ± 8	1995 ± 97	182 ± 9
Lab13_c	55 ± 5	1465 ± 136	117 ± 6
Lab13_d	38 ± 5	980 ± 53	117 ± 6
Lab13_e	53 ± 5	1248 ± 66	117 ± 6

results ~15 % higher than the reference value and the data show a normal distribution, concluding that they are comparable according to the ANOVA test. With the exception of Lab11, the instruments improved their uncertainty due to the higher dose rate in Station 2. Figure 5 shows the results obtained at point X, where the gamma spectrum is slightly different because of the presence of the uranium tailings that were not covered. Here there is no significant difference between the reference value and the average of the participant values. However, in this case all the instruments agree within 25 %, with the exception of instrument Lab9, which exhibits again poor sensitivity. As in Station 1, the values are normally distributed although in this case the ANOVA test shows that these values are not comparable.

It is interesting to perform an evaluation test of participants in order to quantify the competence of the different radiation monitors. To do this, three criteria have been used following the scheme used by IAEA in proficiency test[13]: accuracy, relative bias and variation coefficient. Each criterion has been analysed using the following equations:

$$A_{\text{lab}} = \frac{K_{\text{lab}} - K_{\text{ref}}}{2.58\sqrt{\sigma_{\text{lab}}^2 + \sigma_{\text{ref}}^2}}, \quad (1)$$

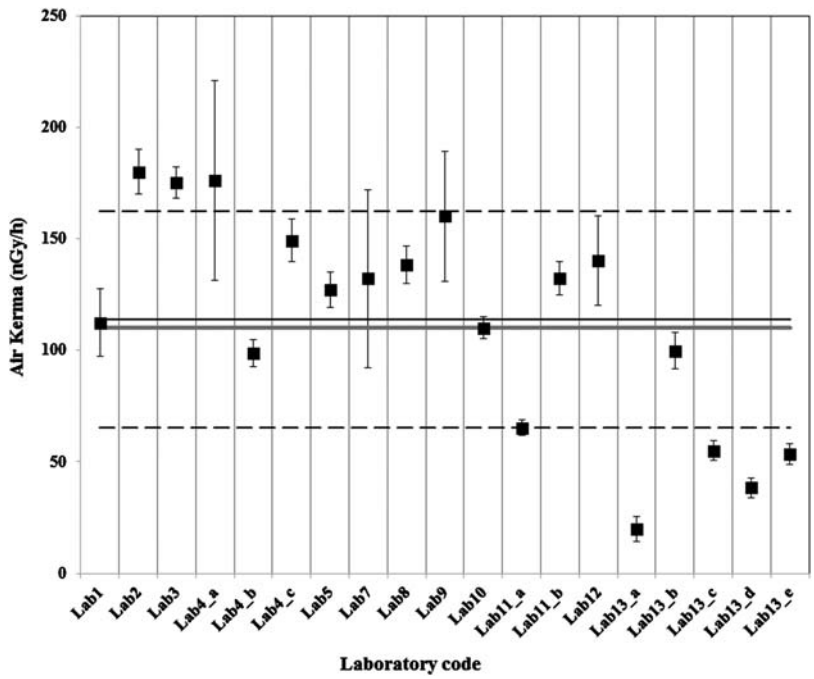

Figure 3. Results obtained at Station 1 (restored area). The grey line indicates the reference value by CIEMAT. The black line indicates the average of the participant mean values and the light grey dashed lines one standard deviation up and below the average value.

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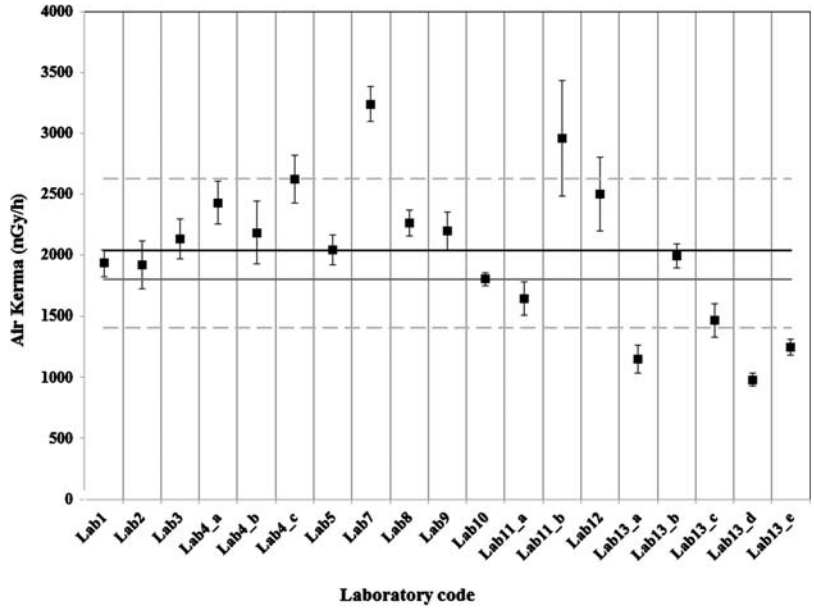

Figure 4. Results obtained at Station 2 (no restored uranium tailings area). The grey line indicates the reference value by CIEMAT. The black line indicates the average of the participant mean values and the light grey dashed lines one standard deviation up and below the average value.

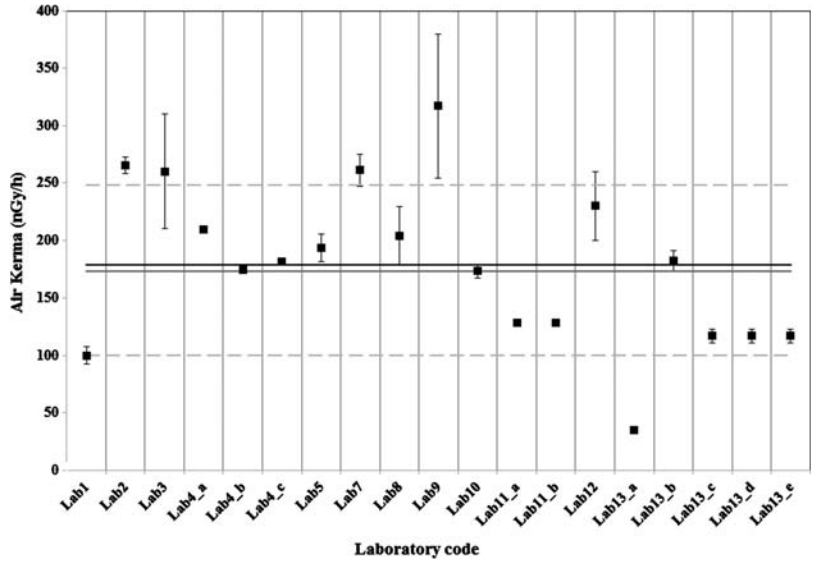

Figure 5. Results obtained at point X (uranium tailings). The grey line indicates the reference value by CIEMAT. The black line indicates the average of the participant mean values and the light grey dashed lines one standard deviation up and below the average value.

where K is the value of air kerma, σ is the uncertainty of air kerma and A is the accuracy value for the corresponding radiation. The radiation monitor was considered acceptable for performing environmental gamma dose rate determination if the result of the accuracy test is less than or equal to one. The

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Table 5. Evaluation test performed in the environmental gamma dose rate intercomparison exercise.

Detector type	Accuracy			Relative bias (%)			Variation coefficient (%)		
	Station 1	Station 2	Point X	Station 1	Station 2	Point X	Station 1	Station 2	Point X
GM	0.06	0.42	3.14	2	7	−42	14	6	8
GM	2.43	0.23	4.12	64	6	53	6	10	3
GM	2.91	0.76	0.67	59	18	50	4	8	19
Scintillator	0.57	1.32	NA	60	35	21	25	7	NA
Scintillator	0.56	0.56	NA	−10	21	1	6	12	NA
Scintillator	1.40	1.57	NA	36	46	5	6	8	NA
GM	0.70	0.71	0.60	15	13	12	6	6	6
Scintillation NaI (Tl)	0.21	3.72	2.29	20	80	51	30	4	5
Energy-compensated GM detector and a beta shield	1.12	1.57	0.47	26	26	18	6	5	12
GM	0.66	0.95	0.88	46	22	83	18	7	20
Ion chamber/proportional counter	0.00	0.00	0.00	0	0	0	5	3	3
1 × 1 NaI scintillator	2.88	0.42	3.37	−41	−9	−26	5	8	1
3 × 3 NaI scintillator	0.98	0.94	3.37	20	64	−26	6	16	1
Proportional counter	0.56	0.89	0.73	27	39	33	14	12	13
Ion chamber/proportional counter	0.42	0.69	0.34	−9	11	5	8	5	5
Scintillator	4.65	2.04	9.93	−82	−36	−80	28	10	6
Scintillator	3.15	0.90	2.78	−50	−19	−32	8	9	5
Scintillator	4.10	4.37	2.78	−65	−46	−32	12	5	5
Scintillator	3.24	2.59	2.78	−52	−31	−32	9	5	5

NA means that the participant did not provide enough data for performing the test.

second evaluation test applied was the relative bias from the reference value:

$$\text{Relative bias} = \frac{K_{\text{lab}} - K_{\text{ref}}}{K_{\text{ref}}} \times 100, \quad (2)$$

and finally another criterion such as variation coefficient was used:

$$V_{\text{lab}} = \frac{\sigma_{\text{lab}}}{K_{\text{lab}}} \times 100. \quad (3)$$

Table 5 shows the results obtained for the evaluation test in Stations 1 and 2 and point X.

In order to compare the results, the results of each detector were analysed taking into account these two criteria: accuracy test less than or equal to one and variation coefficient <10 %. According to these criteria, only two detectors of type GM and ion chamber/proportional counter pass the two tests in the three testing points.

CONCLUSIONS

The dose rate exercise within the international intercomparison carried out in Saelices el Chico proved the benefits and effectiveness for participants. Measurements of most of the dose rate instruments agree within 25 % in the three studied sites, despite the differences in dose rate and soil composition. However, this figure could be easily improved when considering corrections for photon energy response or harmonisation in the calibration technique. Limitations of some monitors when used in practice due to poor sensitivity or environmental influences such as temperature were also learnt.

It was also demonstrated that the measurement of dose rate is not an easy task. Although most of the participants obtained results that fall within 25 % in each testing site, when performing evaluation criteria only 25 % of participants passed the test in the three testing sites. This result shows that calibrating the radiation detectors in the laboratory could be a necessary condition but never enough to achieve good results. The evaluation criteria carried out using IAEA recommendations are quite strict for such kinds of measurements. Nevertheless, IAEA criteria should be applied in all intercomparison exercises as described in this paper. Therefore, it is possible to compare results from different intercomparisons. In conclusion, this paper reveals the significance of calibrating instruments under field conditions and such calibrations will continue at LNR in the future.

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