INTERNATIONAL INTERCOMPARISON EXERCISE ON NATURAL RADIATION MEASUREMENTS UNDER FIELD CONDITIONS (IFC11)*

BOTOND PAPP [1], ALEXANDRA CUCOŞ (DINU) [1*], MIRCEA MOLDOVAN [1], ROBERT BEGY[1], TIBERIU DICU [1], DAN C. NIŢĂ [1], CARLOS SAINZ [1,2], CONSTANTIN COSMA[1]

[1] „Babeş-Bolyai"University, Fântânele street, no.30, Cluj-Napoca, 400294, Romania
E-mail: dinualexandra2007@gmail.com
[2] Department of Medical Physics, Faculty of Medicine, University of Cantabria, Santander, 39011, Spain

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One of the aims of the First Intercomparison on Natural Radioactivity Under Field Conditions was to test different instruments and detectors for the measurement of radon gas and external gamma radiation (dose rate) in filed conditions, where the levels of natural radiation are quite high. The event was organized by the Radon group of University of Cantabria, (Spain) LaRUC, and was held in the end of May 2011, in the area of an old uranium mine of ENUSA, near Saelices el Chico (Salamanca, Spain). The following activities were performed: External gamma dose rate; Radon indoors with active and passive detectors; Radon outdoors; Radon in water; Radon exhalation rate from building materials; Radon exhalation rate from soil and Radon in soil gas. This work presents the main results of the intercomparison obtained by our group in all the exercises.

Key words: radon in water, gamma dose rate, radon in soil gas, indoors passive detectors, indoors active detectors, international intercomparison.

1. INTRODUCTION

Measuring and testing indoor radon levels and external radiation dose for the population due to natural sources are of high importance to the total effective dose and it is necessary to ensure that the values provided from measurements and tests are accurate. One of the most common ways to assure the quality of the tests is by the means of intercomparisons carried out by approved services or reference laboratories [1]. Intercomparisons are an important tool for the measurement

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services and laboratories in order to detect potential problems and perform rectifications as well as to provide calibrations for the instruments [2, 3].

The *First Intercomparison on Natural Radioactivity Under Field Conditions* was held in the area of an old uranium mine of ENUSA in the municipality of Saelices el Chico (Salamanca, Spain) between 23 and 27 of May, 2011, and was organized by the *Radon group of University of Cantabria* (Spain). 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.

The old uranium mine site was closed in 2004 and since then, the restoration process has been taking place. The *Radon group from University of Cantabria* in Spain has established a site with normal background and the second site where the exposure is more 20 time higher as normal background value in order to test and calibrates instruments and detectors for the measurement of natural radiation under typically variations of temperature, pressure and atmospheric pressure which we can find in occupancy places (dwellings and working places). The exercises were carried out in one of the buildings of the old uranium mine used in the past for uranium mineral treatment, where the Radon group of University of Cantabria arranged the house in order to convert it to a laboratory of natural radioactivity with natural levels of indoor radon gas. These levels are also affected by daily changes of weather conditions which make the place suitable for studying radon concentration variations indoors. The building is a two-storey house, where the ground level contains two radon chambers with higher radon levels. The possibility of using artificial ventilation systems allows controlling the natural radon concentrations. Another room was used for the radon in water exercise. In the surroundings of the building a 9×9 m^2 was used as field for external gamma dose rate measurements [4].

The main goal of the inter-comparison exercise was measurements of radon gas in water, soil gas, indoors and outdoors, radon exhalation rate and measurements of the external gamma dose rate (by the existence of radioactive tailings in the surroundings). Thus, the following activities was performed: *External gamma dose rate; Radon indoors with active and passive detectors; Radon outdoors; Radon in water; Radon exhalation rate from building materials; Radon exhalation rate from soil and Radon in soil gas.* A total number of 45 participant institution from different European countries took part in the activities, such as universities, reference laboratories and commercial companies which main activities are related to the measurement of natural radiation and radon gas and external gamma radiation in particular. This report presents the main results of the intercomparison, as well as discussions of the achieved results obtained by our group from *Babes-Bolyai University* (BBU) in all the exercises [4].

2. MATERIAL AND METHODS

2.1. RADON IN WATER

The aim of this exercise was to test different measuring systems of radon in water using a sample with a fix radon concentration. To do this, a barrel containing 100 L of water was connected to a small box containing a soil sample with high amount of uranium. Thus, radon gas generated from uranium radioactive decay is pumped into the barrel and was dissolved in the water. The barrel was closed in order to prevent radon gas leakages. The sampling was done using a tap installed in the wall of the barrel.

The method of measuring radon from water sample used by us based on the using of a LUK-VR system, which consists from a LUK-3A radon detector (based on scintillation technique with Lucas cells) and a VR scrubber for radon measurements in water samples [5, 6]. The VR-scrubber consists from a glass vessel of 500 cm^3 volume in which a known quantity of water sample (*i.e.* 300 cm^3) was introduced. The principle of operation is that the concentration of the radon dissolved in the water sample is mixed with the air that is on the top of the water level, within the scrubber volume. Following this, air is then transferred from the scrubber into the Lucas cell, to measure the radon activity concentration of the sample, by Lucas cell method. The extraction of radon gas from the water sample (*i.e.* the transfer process from the scrubber into the detection chamber of the detector), and the calibration of the method (*i.e.* calculations of radon activity concentration of a water sample and the efficiency detection of the Lucas cell) in [$Bq \cdot L^{-1}$] are described in details in [6, 7].

2.2. EXTERNAL GAMMA DOSE RATE

The aim of this exercise was to compare different instruments of dose rate meter at two selected sites from the area of the old uranium mine of ENUSA with highest natural gamma doses. One of these had the name of “Severiano green” consists from 17 points, and another site having high dose rates values consists from 4 points. A total number of 11 participants take part in this activity.

BBU group participated with an instrument in the name of “Gamma Scout” dose rate meter, equipped with a Geiger-Müller counter tube, which determines the dose rate in terms of the Equivalent Dose [$\mu Sv \cdot h^{-1}$]. This dose rate meter detect not only gamma radiation, but alpha and beta radiation as well. In this inter-comparison exercise it was used for the detection of the gamma radiation, only [8]. The calibration of the instrument (*i.e.* conversion from imp/sec. into the dose rate) was made using the Cs-137 isotope at the gamma energy of 661 keV, by which the conversion from the Equivalent Dose H*(10) [Sv] to Air Kerma (K_{air}) [Gy] was done with a factor of (Sv/Gy = 1.2) [4].

2.3. RADON IN SOIL GAS

Radon in soil gas comparisons tests the calibration of the instruments and the techniques of soil gas sampling, soil gas transfer into the detection chamber, radon-measuring procedures, stability of field measurements, and elimination of thoron as well as data processing [9].

The method of radon in soil measurements is composed from the sampling of soil gas and the detection of radon gas. In our case, for sampling the soil gas a "Neznal probe" was used (a steel pipe with 1 m length and 1 cm diameter), which was inserted into the soil to a given depth (80 cm regularly). To create an active volume at the end of the probe in the soil, it should remove a few cm. For soil gas sampling a Janet Syringe was used, with a volume of 145 mL (equal with the volume of the detection cell) [10, 11]. Radon activity concentration was measured using the LUK3C radon and thoron detector (Jiří Plch-SMM, Prague), which was developed for radon measurements in soil gas and is based on a scintillation technique with Lucas cell [5]. The characteristics of the detection system of the instrument are described in details in the papers under references [13, 14]. Table 1 shows the characteristics of the sampling system and of the instrument used by our group in this exercise.

Table 1

Characteristics of the sampling system and of the instrument used by BBU group gin the International exercise (Saelices del Chico) "Radon in soil gas"

SAMPLING SYSTEM DESCRIPTION	
Type (description) of the sampling probe	Neznal probe
Description of the sampling system	grab sampling - syringe (150 mL)
Typical volume of the soil-gas sample	150 mL
INSTRUMENT DESCRIPTION	
Instrument Model	LUK 3C
Manufacturer	Jiří Plch-SMM, Prague
Last calibration	from manufacturer
Principle of measurement	Lucas cell (scintillation), time of counting of 400 s; influence of thoron eliminated

The selected site for this exercise (previously analyzed by the Radon v.o.s. company, Czech Republic) was a site with high radon levels in soil gas. The measurements of radon in soil were performed at 8 field points.

2.4. RADON INDOORS PASSIVE DETECTORS

The Indoor Radon exercise (*i.e.* the exposures) with passive detectors were done in three separate rooms of the building, two rooms at the ground floor and one room in the first floor. In total, the 22 institutions were participated at this exercise, using five different types of detectors: CR39 (18 sets), LR-115 (3 sets), Makrofol (2 sets), activated charcoal (3 sets) and electrets (2 sets) [4]. In particular, BBU group used CR-39 solid state nuclear track detectors provided from Radosys Ltd, Hungary. The characteristics of the used CR-39 detectors are the following: thickness of 1 mm, total area of 100 mm^2, type and filter of air gap and the range of the exposure between 40-12000 $kBq\cdot h\cdot m^{-3}$ (http://www.radosys.com).

After the exposure to radon concentration, an etching process and an automatic reading of all the detectors had been made in the Environmental Radioactivity Laboratory of Babeş-Bolyai University, using RadoSys-2000 equipment (Elektronika, Budapest, Hungary) [13, 14, 15, 16].

For a good statistics of the result, 15 detectors were necessary for each exposure and 15 more to be used as transits. During the intercomparison exercise three different exposures were performed (exposure no.1, 2 and 3). Some detectors were used as transits, being considered as exposure no. 4. Results of the intercomparison were done in terms of exposure to radon [$kBq\cdot h\cdot m^{-3}$], but some of them (due to the characteristics of the used detectors) gave the result in terms of radon concentration [$kBq\cdot m^{-3}$] [4]. Table 2 shows the characteristics of the radon exposures and the number of participants in each. In addition, 9 laboratories used the transit exposures (*i.e.* exposure no.4) and they gave the results of the transits.

Table 2

Characteristics of the radon indoors exposures for CR-39 track detectors.

Exposure	**No. 1**	**No. 2**	**No. 3**
Start (dd.mm. hh:mm)	24.05. (12:30)	25.05. (17:30)	26.05. (11:30)
End (dd.mm. hh:mm)	08.06. (18:30)	16.06. (20:10)	13.06. (11:15)
Nr. of participants	24	22	17

2.5. RADON INDOORS ACTIVE DETECTORS

The exercise of the comparison of Radon indoors active detectors was performed in the two rooms located at the ground floor of the laboratory building, and the measurements were done exclusively during the meeting. The total number of the participant institutions was 17, and the following instruments were used: RADON SCOUT (31 pieces), Alpha Guard (12 pieces), RADIM (12 pieces), RAD7 (2 pieces) and ATMOS (2 pieces). BBU group was participated with RADON SCOUT (4 pieces), Alpha Guard (1 pieces) and RADIM (3 pieces) [4].

3. RESULTS AND DISCUSSIONS

3.1. RADON IN WATER

BBU group performed three measurements of radon in water, and gives the result of radon activity concentration in terms of $Bq\cdot L^{-1}$, with uncertainty. A preliminary analysis of the data of this exercise was made, based on the results from 13 participants, where no reference value was set (see Table 3) [4].

Table 3

Results of the exercise “Radon in water” (BBU results: CRn ± dCRn, and results of the statistical analysis: mean, standard deviation, etc).

Our results	
CRn ± dCRn [Bq L^{-1}]	358.3 ± 35.8
dCRn (%)	10
Statistical analysis of all the results	
Mean (SD) [Bq L^{-1}]	338 (37)
Min. [Bq L^{-1}]	252
Max. [Bq L^{-1}]	412
SD (%)	11

Statistical analysis show that our result using Lucas cell technique is quite close to the border of ±1 standard deviation. A possible explanation for the difference in our results and of other participants could be attributed to the sampling technique, where all the samples were taken in a short interval (2-3 hours). Therefore the participants were opening and closing the tap for the acquisition of the water sample. This situation possibly created disequilibrium in the radon concentration inside the barrel so the radon concentration in water samples was not exactly the same.

3.2. EXTERNAL GAMMA DOSE RATE

Each participant was evaluated according to the mean value of the Air Kerma Rate (K_{air}) [$nGy \cdot h^{-1}$] with standard deviation which was determined from the Equivalent Dose Rate H*(10) [$\mu Sv \cdot h^{-1}$], provided from the individual dose rate measurement. Finally, for the two sites, the mean value of a single participant was compared with the mean value of all the participants, with an uncertainty of 1 SD. In addition, was done reference values provided by CIEMAT, in terms of Air Kerma Rate (K_{air}). Results of the measurements (*i.e.* reference values for both sites, means and standard deviations of BBU results, as well as results of all the participants) were represented in Table 4.

Comparing the mean values of these results with the reference values of the sites, in case of the Severiano green site, BBU mean value is in agreement with the reference value in the range of the standard deviation. In case of the site with high dose rate, the mean value with the standard deviation is not in the range of the reference value but in both case the mean values is in agreement with the mean values from all the results, taking account the standard deviations.

Table 4

Results of the exercise “external gamma dose rate” (mean of our results *versus* mean of all the results with standard deviations, in units of Air Kerma Rate)

Site	**Reference value [nGy·h^{-1}]**	**Our results Mean ± S.D. [nGy·h^{-1}]**	**All results Mean (S.D.) [nGy·h^{-1}]**
Severiano green	110	144 ± 29	130 (35)
High gamma dose rate	1800	2613 ± 352	2257 (436)

We can conclude that it has been observed the need to take into account the energy response of the detector in the case of ambient dose rate determinations. In these types of measurements, the energy spectrum is quite different from that use in the calibration of the device (normally a ^{137}Cs source). Most of the errors are due to this phenomenon [4]. In these cases the devices are not designed for outdoors determinations in the presence of the natural radionuclides.

3.3. RADON IN SOIL GAS

Results of radon in soil measurements, *i.e.* radon activity concentrations were done in terms of [kBq·m^{-3}]. Table 5 shows results of our measurements (radon concentrations and uncertainties) for the 8 points, also mean values and standard deviations of radon concentrations provided from the results of 18 participants. It was different number of results provided from the participants in each points

Table 5

BBU results of the exercise “Radon in soil gas” and results of the statistical analysis (mean values and standard deviation) of all the participants

Point	**Our results**		**Means of all results**	
	Depth [m]	CRn ± dCRn [kBq·m^{-3}]	Nr. of results	CRn (SD) [kBq·m^{-3}]
A	0.6	8.10 ±1.00	12	13 (16)
B	0.5	29.16 ± 2.16	15	40 (19)
C	0.65	33.60 ± 2.09	17	48 (27)
D	0.65	17.23 ± 1.17	13	29 (16)
E	0.5	45.43 ± 3.79	18	72 (36)
F	0.5	35.03 ± 2.45	15	37 (39)
G	0.6	120.87 ± 8.14	16	126 (94)
H	0.5	13.17 ± 0.97	18	22 (8)

The distribution of the results in each point was checked and two different data distribution was found. In points A, B, D, F and H the values give a log-normal distribution, by which the results were done in terms of Geometric Mean and Geometric Standard Deviation. In the rest of the points C, E and G the distribution is normal and the results are characterized by Mean and Standard Deviation [4].

The comparison of these radon results with the means of all the results conducts to a good agreement of BBU concentration values with the mean concentrations determined from all the results, with a correlation coefficient of 0.98.

3.4. RADON INDOOR PASSIVE DETECTORS

From radon indoor by passive detectors measurements it can be seen that all the results provided from the participants are inside the limits of 30 % of the standard deviation from the mean value. The statistical analysis of all the data using CR-39 track detectors shows that the distributions of the results for the three radon exposures (no.1, no.2 and no.3) follow a normal distribution which is characterized by the mean value and standard deviation from all results. Table 6 shows a summary of the results from the measurements with our CR-39 track detectors participating in the three exposures (in addition for the exposure with transits), in comparison with the mean values and standard deviations from all the detectors provided by the participants.

Table 6

Results obtained by our CR-39 detectors in the exercise radon indoor, compared by the means and standard deviations from all CR-39 detectors results.

Exposure	**no. 1**	**no. 2**	**no. 3**	**no. 4**
Our results Mean (SD) [kBq·h·m^{-3}])	2978 (85)	670 (28)	98 (16)	24 (12)
SD (%)	3 %	4 %	17 %	50 %
All results Mean (SD) [kBq·h·m^{-3}]	3521 (478)	693 (67)	104 (16)	63
SD (%)	**14 %**	**10 %**	**15 %**	**23 %**
Category	**B**	**A**	**A**	**-**

The results corresponding to the transit detectors (exposure no. 4) had a mean value of 63 kBq·m^{-3} with a standard deviation of 23 %. Our result in this case is smaller because we also extract the background of detectors. For the two exposures (no.2 and no.3) the standard deviations from the mean value are less to 15 % for all participants and BBU result is much closed to mean value (3.3%, respectively 5.7%). In the case of exposure no.1 the result is with 15.4% less as the mean of all participants and we checked and reinstalled the software for our Radosys.

There was no reference value and we compared BBU results with the means of all the results. To do this, absolute difference (in %) between BBU mean and the mean value of all the results ≤ 10 % were ranked as category A; ≤ 15% as category B; ≤ 20 % as category C. These categories were done in the Table 6 (last line).

3.5. RADON INDOORS ACTIVE DETECTORS

As results of the radon indoors measurements using active detectors was give a graph of the variation of the radon concentrations (during the period of the exercise) for all the devices (from all the participants) (see Fig.1), where BBU devices were assigned by a code **IFC11_21** [4]. It can be observed and can be conclude that the agreement between all devices and our devices is good.

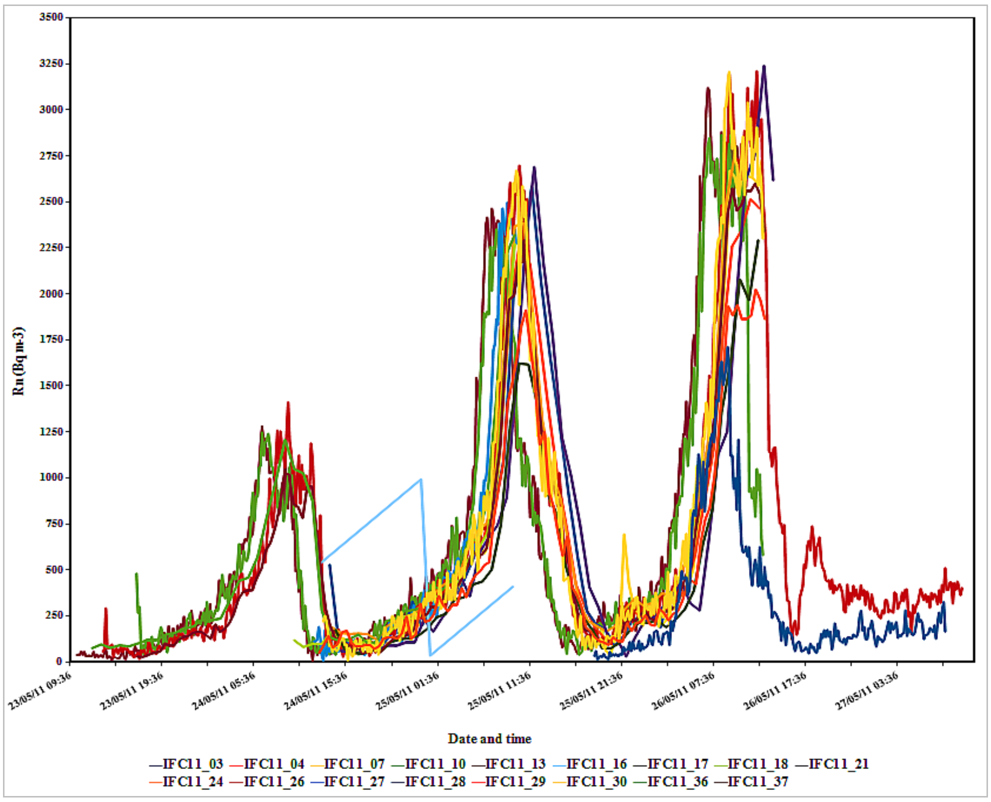

Fig. 1 – Results of radon indoors using active detectors for all participants (23-27 of May) [4].

4. CONCLUSIONS

Considering the frame of the *International Intercomparison on Natural Radioactivity Under Field Conditions* it has been tested Radon in water, External gamma dose rate, Radon in soil, Radon indoors with passive and active detectors measurement methods, with a number of 45 participant institution from European countries.

The results obtained from *Radon in water comparison measurements* are in good agreement with the limit of the standard deviation of the mean value provided from the results of all the participants. The uncertainty of the determination of the activity concentration of our result is in the order of 10 %.

The results obtained from *External gamma dose rate measurements* comparing with reference values of the two sites show that at low dose rate (*i.e.* in case of the Severiano green site) our mean value is in agreement with reference value in the range of the standard deviation, and in the case of high dose rate our mean value is higher (45%) than reference value but comparing with average value of all participants is higher only with 15%. The uncertainty (*i.e.* the standard deviation) of our determinations is lower than 20 %.

The *Radon in soil exercise* shows that BBU results of radon concentrations are in good agreement with the means of all the results radon concentrations provided from the participants, with a correlation coefficient of 0.98.

The *Radon indoors passive detectors measurements* based on three different exposures by CR 39 track detectors, show an absolute difference ≤ 15.4% in case of the high exposure (no.1), ≤ 6% in case of the medium and low exposure (no.2 as well as no.3, respectively).

The *Radon indoors active detectors measurement results are in good agreement* between all devices and BBU devices are applicable for the period of measurements.

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