



The Laboratory of Natural Radiation (LNR) – a place to test radon instruments under variable conditions of radon concentration and climatic variables

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Abstract. The publication of the new European Union Basic Safety Standards represents a remarkable milestone in the field of radiological protection in terms of adding radon exposure to this framework. Therefore, the coming years will bring the need to measure radon not only in the workplaces but also in the living spaces as a direct outcome of the application of the new EU Directive. So, the importance of having reliable instruments is evident and interlaboratory exercises are becoming more and more popular. However, most of them are carried out under constant conditions of meteorological variables. We present in this paper a facility to broaden the interlaboratory comparisons further by adding the study of radon exposures under real conditions of changes in climatic parameters. In addition, the facility has the possibility to verify the response of radon monitors when the radon concentration changes several orders of magnitude in a short period of time. Our work shows some results of one of the interlaboratory exercises carried out in the premises, where the radon levels were rather homogeneous in the testing room.

Key words: active monitors • interlaboratory comparison • *in-situ* • radon

Introduction

Natural radioactivity is the main component of the annual effective dose received by the general public. Among them, radon contributes around 50% to the total amount of radioactive dose for the general public [1]. The European Directive 96/29/EURATOM mandates the monitoring of occupational radiation exposures, which must be done by approved dosimetry services [2]. The new basic safety standards (BSS) [3] for protection against the dangers arising from exposure to ionizing radiation were issued in January 2014. The subject matter of the proposed directive is to establish a community framework for the BSS for protection of the health of the people. In particular, the Directive applies to the management of existing exposure situations, including the exposure of members of public to indoor radon, the external exposure from building materials and cases of lasting exposure resulting from the after-effects of an emergency or a past activity. The Annex XVIII of the document summarizes the list of items to be covered in the national action plan to manage long-term risks from radon exposures.

Radon is the biggest contributor to the total amount of radioactive dose for the general public. However, legislation in different countries differs from obligatory control of radon gas in some countries (Ireland, the Nordic countries and the Czech Republic), to recommended monitoring in other

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countries (Spain or Italy for instance). Moreover, there are two recommendations on radon in the European legislation suggesting maximum levels of this gas for new and existing houses [4] as well as the EU Directive that establishes a radon reference level for drinking water [5].

Application of the new European Directive will require competent measurement services in all member states. Thus, it is very important to assess that values provided by different laboratories are accurate and precise. One of the most common ways to assure the quality of the results of laboratories is by means of interlaboratory comparisons carried out by approved services of reference laboratories. Here, we can cite those radon intercomparison exercises done annually by Bundesamt für Strahlenschutz (BfS) in Germany and Public Health England (PHE) in the UK. There are international organization for standardization guidelines that provide information on how to perform such exercises. The guidelines include parameters that must be used to analyse the results [6]. The scenario for the typical intercomparison exercise is the exposure of the instrument to a reference atmosphere of the parameter to control (i.e. radon gas) under temperature, humidity and atmospheric pressure stable conditions. However, these are not the common situations that we can find in a normal dwelling when measuring this gas. Hence the existence of facilities to test instruments for the measurement of radon gas under changing conditions of meteorological parameters becomes necessary.

We present in this paper, the premises where we have carried out interlaboratory exercises aiming to test the proficiency of laboratories to measure radon gas under variable ambient conditions. Therefore, we include results of one of those intercomparison exercises corresponding to 19 participants and one week of exposure time.

Materials and methods

The Radon Group from University of Cantabria in Spain has established a site where the values of natural radioactivity are high enough to test instruments and detectors under typical variations of temperature, humidity and atmospheric pressure, which we can find in places of occupancy (dwellings and working places). Such a place is located in an old uranium mine site, where the first intercomparison exercise under field conditions were held in May 2011 [7]. The laboratory is located at a site where the values of natural radioactivity allow testing instruments and detectors under typical variations of temperature, pressure and atmospheric pressure.

The mine site was shut down in 2004 and since then the restoration process has been taking place. During these activities, one of the buildings used for monitoring activities in the mine was chosen to house the Laboratory of Natural Radiation (LNR) for calibration and testing of instruments and detectors for the measurement of natural radiation. The Radon Group in collaboration with the National

Uranium Company of Spain (ENUSA) and the Spanish Nuclear Safety Council (CSN) was in charge of modifying this building for 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 performing experiments devoted to the analysis of environmental radioactivity, as well as a location for testing specialized instruments for measuring natural radiation. Figure 1 shows a general view of the main building of LNR.

The building is a two-storey house with four rooms on the ground floor. There is one room on the ground floor that is used for radon in water calibration purposes. In the another room are 30 workstations for participants of the exercises and two more spaces designed as radon chambers. One of these chambers has an artificial ventilation system installed. Both radon chambers have the same volume and the source of radon is the underground soil. Therefore, this source is of natural origin and is affected by external meteorological parameters (temperature, humidity, pressure) as we observe in Fig. 2.

The upper floor of the building consists of a conference hall and a big room, which can also be used as radon chamber for studying very low exposures



Fig. 1. The Laboratory of Natural Radiation is located in the facilities of ENUSA uranium mine in Saelices el Chico (Salamanca, Spain).

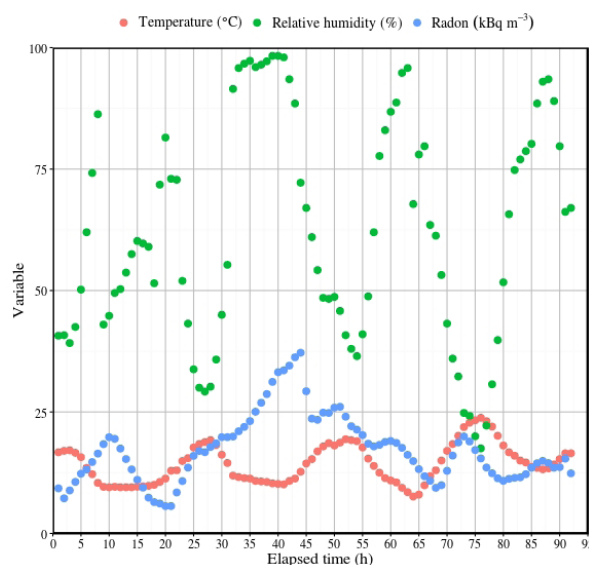


Fig. 2. Example of behaviour of external parameters (temperature, relative humidity and pressure) at the Laboratory of Natural Radiation premises.

(radon concentrations are usually within the range 100–200 Bq·m⁻³). This room has also a ventilation system to reduce radon levels if required.

The interlaboratory exercises normally consist of exposure of radon passive detectors to a radon atmosphere in one or two rooms (approx. 45 m³ volume each). These rooms are affected by daily variations of temperature, humidity and pressure. Therefore, we try to simulate the same conditions as one can expect to find in a real place of occupancy (either workplaces or dwellings). Due to the specific climatic conditions of the area, the temperature gradient can be high (up to 20°C in a 24-hour period), while the rest of parameters are quite stable. The laboratory allows carrying out the following tests: radon indoors and outdoors, radon in water, radon exhalation rate from building materials and external gamma dose rate.

The results presented in this paper correspond to the radon interlaboratory comparison carried out in 2013 by 19 laboratories. Each participant was requested to send a set of 30 radon passive detectors (CR39, Makrofol, activated charcoal) and five detectors to be used as transits. Each exposure consisted of 10 detectors. At the end of radon exposure, the detectors were moved to a room where radon concentrations were much lower comparing with the exposure values to perform degassing. Then the detectors were wrapped in aluminium bags and sent back to the participants for analysis.

To obtain a reference value to be used for analysing the results, we used Radon Scout monitors installed at different points inside the room. These are semiconductor detectors that can detect alpha particles emitted by radon decay daughters (²¹⁸Po and ²¹⁴Po) with a sensitivity of 1.8 cpm at 1000 Bq·m⁻³. Each Radon Scout monitor was calibrated with a reference instrument in a radon chamber under standard ambient conditions (humidity and temperature ranging from 30 to 70% and from 10 to 30°C, respectively). The reference instrument is annually recalibrated by the BfS (German Federal Office for Radiation Protection), which is an accredited institution for radon calibration purposes by PTB (Physikalisch-Technische Bundesanstalt). The measurement range varies from 0 to 10 MBq·m⁻³ and they also provide extra information on temperature and humidity [8]. We selected an integration time of 1 h. Figure 3 shows the position of the radon monitors used in the intercomparisons.

Results and discussion

We tested the homogeneity of radon gas concentration in the room by means of a very simple statistical analysis of the obtained data. Each monitor provided 91 values of radon concentration, and we compared the results of the six devices to find out whether we can assume that all monitors have the same response to this variable. In addition to that, we checked if radon concentration in the room was homogeneous. This is very important due to the large number of passive detectors exposed during the exercise. The



Fig. 3. Position of Radon Scout monitors used during the interlaboratory exercises to obtain reference levels of radon exposure.

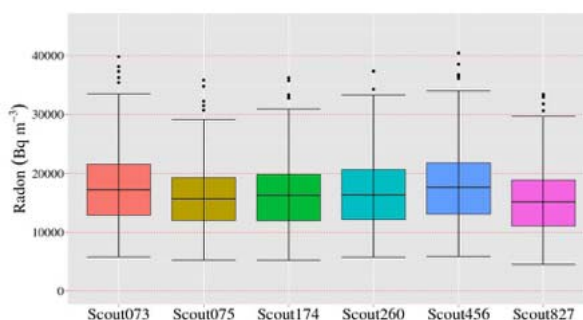


Fig. 4. Exploratory graph showing results of six radon monitors installed in the intercomparison room.

room was not large and we had to place the detectors at different points in the exposure area.

First, we will pay attention to the box plot of the results corresponding to six monitors shown in Fig. 4. The horizontal line through the box indicates the median or second quartile. By looking at the box size of the six devices, we observed that the interquartile range was quite similar, and thus the expected variability of the data was similar in all cases. Also, the boundaries of the boxes (1st and 3rd) quartiles correspond to comparable values, respectively. We can see that there are some outliers registered by the Radon Scouts, but only in the case of high radon concentrations. These can be related to peaks on the radon exposures in the room. However, we cannot identify outliers for low concentrations.

We can analyse if the time series data of the six Radon Scouts are comparable or not. From Fig. 4, it seems reasonable that this assumption is correct. As had expected, the radon distribution registered was log-normal for all equipments (*p*-values ranging from 0.288 to 0.5362). We performed a non-parametric statistical test (Kruskal-Wallis) using the R software [9], and concluded that there was no evidence to suggest differences exist among the radon distributions of the six Radon Scouts installed in the room during the intercomparison. Also, the Fligner-Killeen test of homogeneity of variances shows that variances were similar in all the six radon monitors used as references.

We calculated the mean value for the six monitors in one hour intervals, and the result was considered

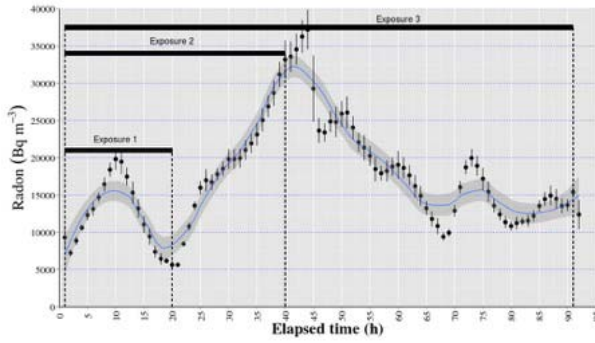


Fig. 5. Radon concentration in the room during the exercise. Grey colour represents polynomial smooth of the time series.

to be the reference exposure level for analysing data provided by the participants. Figure 5 represents the variation of radon concentration during the exercise. If we look at this figure, we note big changes on radon concentration in the room during the entire exercise. We also observe that the individual uncertainties of data are low (between 5% and 15%). The minimum value of radon concentration was $5626 \text{ Bq} \cdot \text{m}^{-3}$ and the maximum was $37\,204 \text{ Bq} \cdot \text{m}^{-3}$. Meanwhile, the three exposures were considered to represent non-constant radon concentration, which was one of the objectives of the interlaboratory comparison: to test the response of radon-passive detectors under real conditions of changes in radon activity concentrations one can find in a real occupancy building.

As we have noticed, the parameters of this type of intercomparison are very different from those normally used for testing radon detectors in reference laboratories, where detectors are exposed to constant radon exposures.

We have performed a two-fold analysis. First, the analysis consists of the evaluation of the results of different laboratories by means of Mandel's h statistic [10, 11]. Following an interlaboratories analysis, Youden graphs can compare the results of each participant. Figure 6 represents Mandel's h statistic calculated for all laboratories and exposures. If we have n participants in an interlaboratory test and each laboratory reports X_i as the mean value for a certain reference level, then $\bar{X}$ is the mean value of all results. Therefore, Mandel's h statistic is calculated for each laboratory as follows:

$$(1) \quad h_i = \frac{X_i - \bar{X}}{S}, \quad i = 1, \dots, n$$

We suppose that the random variables X_i are independent and are normally distributed. This is the case of an interlaboratory comparison. Also, we have shown that the results in our interlaboratory comparison are normally distributed for the three exposures. In Eq. (1) S is:

$$(2) \quad S^2 = \frac{Q}{n-1}$$

and

$$(3) \quad Q = \sum_{i=1}^n (X_i - \bar{X})^2$$

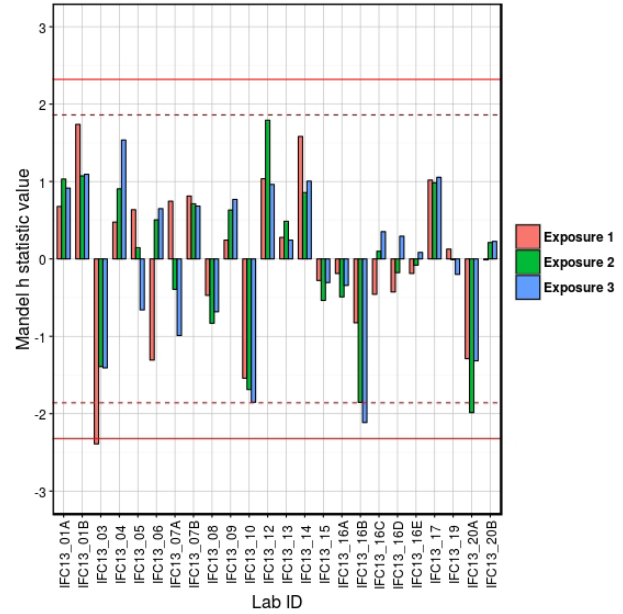


Fig. 6. Mandel's h statistic for the three radon exposures. Red line represents 5% confidence level and dashed line 1%.

Mandel's h statistic is an index that permits the evaluation of the interlaboratory's consistency. We can see in a graph the standardized bias obtained by one particular laboratory and the mean value of the rest of the participants in a particular reference level. We can also define critical confidence levels from this statistic. Figure 6 shows the interlaboratory data grouped by laboratory and gives a view of the laboratory bias and relative precision in the three radon exposures. Looking into this figure, we noticed that six laboratories showed a trend of giving lower values than the whole group, particularly three of them, which were identified as IFC13_03, IFC13_10 and IFC13_20A; these laboratories had a response that was significantly lower than the group in all exposures. The situation for the laboratories giving higher values is always within the interval corresponding to 1% confidence level.

We will look into how laboratories achieve results using a graphical tool called Youden plot [12]. In this graph, we represent pairs of values corresponding to the results of the same participant in two levels of the studied parameter. Each plot was divided into four quadrants, with the circle around the centre of the plot being a representation of the 95% confidence level. In case of only random errors, we would expect to find a cloud of points homogeneously distributed around the centre. Upper right and lower left quadrants represent laboratories that showed systematically higher or lower values than the rest. This finding can be interpreted as the source of systematic errors in the participants.

Figures 7–9 show the Youden graphs of participants compared with pairs of the level of radon exposure. The graphs are an adaptation of the original concept of Youden graph, as we have standardized the results by analysing the differences with the median values for each exposure. We can observe that when we compare exposures 2 and 3 (Fig. 9),

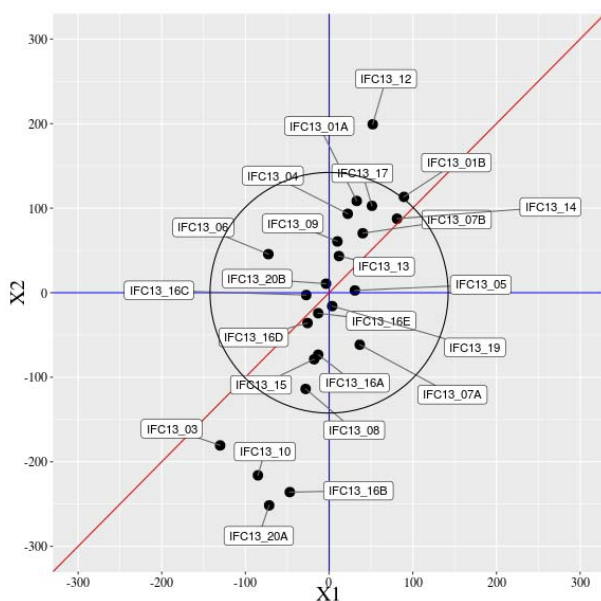


Fig. 7. Youden graph comparing results of exposures 1 and 2.

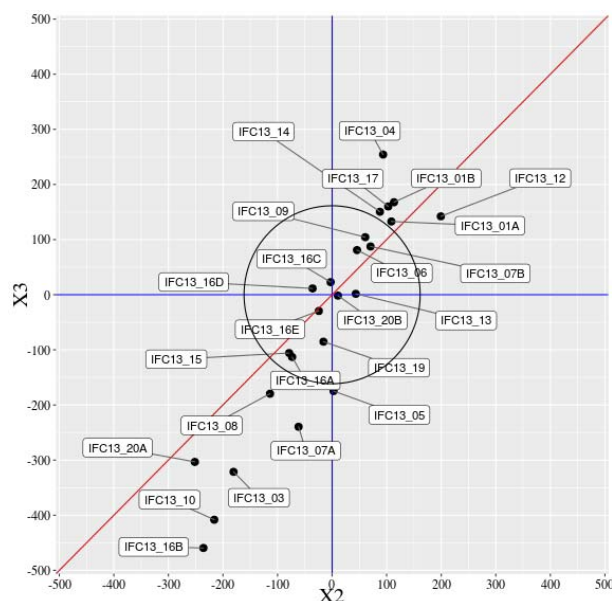


Fig. 9. Youden graph comparing results of exposures 2 and 3.

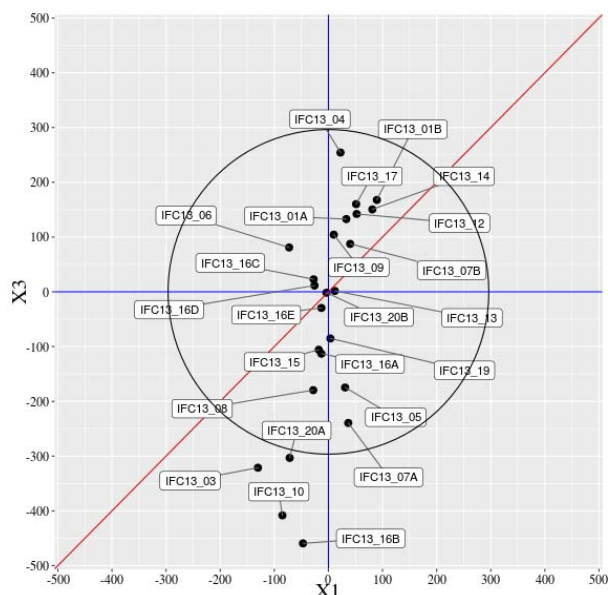


Fig. 8. Youden graph comparing results of exposures 1 and 3.

some laboratories systematically gave results higher or lower than the reference value. On the contrary, a comparison of exposures 1 and 2 and 1 and 3 shows the majority of the participants were within the 95% confidence level circle.

Conclusions

We have presented a summary of an interlaboratory exercise with the participation of 19 institutions from different EU countries. Participants submitted a total number of 24 detector series, which represents a good number of participants to carry out an acceptable interlaboratory exercise. After a detailed analysis of the data, we can sum up the next important outcomes from this experience:

- The results of participants are comparable in all exposures and there are no outliers, except for the case of the lowest exposure where one laboratory reported an anomalously lower value than others in the group.
- It is very important to carry out degassing of the detectors in a low radon atmosphere. This degassing process can be done by exposing radon monitors for several hours (2 h for the work shown on this paper) to outdoor air. Otherwise, it turns out that detectors give significantly higher values than the reference. This is important in the case of the exposure levels we have presented in this paper.
- We have shown in this exercise the importance of carrying out interlaboratory comparisons *in situ* where radon concentrations can change dramatically in a short period of time. Some of the discrepancies observed in the data could be due to problems with the reading systems of track-etched detectors. Therefore, both type of intercomparisons, constant values of radon exposures and changing values are needed to assess the performance of measurement laboratories.
- Measuring low exposures is complicated due to the large uncertainties observed. This is a problem when laboratories have to measure low radon concentrations.
- Some laboratories seem to have problems with systematic errors, which can be attributed to several reasons, and they will require further internal evaluation.

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