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Results of the 2015 national indoor radon intercomparison measurements in Serbia

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Abstract. Results and conclusions of interlaboratory comparison of indoor radon in 2015 in Serbia are presented. The participants were three accredited laboratories from Serbia: Serbian Institute of Occupational Health "Dr Dragomir Karajović", Laboratory for Radioactivity and Dose Measurements at the Faculty of Sciences, University of Novi Sad and Radiation and Environmental Protection Department, Vinča Institute of Nuclear Science. The laboratories make use of the same method for radon measurement, using charcoal canisters according to US Environmental Protection Agency (EPA) protocol 520/5-87-005. Calibration of detection efficiency was performed using EPA radium standard. Radon activity concentrations were determined on the basis of the intensity of short-living radon daughters, ^{214}Bi and ^{214}Pb, gamma lines. The results of intercomparison were evaluated by using the *u*-test, which was calculated according to the International Atomic Energy Agency criteria. In this paper, not only limitations but also the advantages and possibilities of application of this method for measuring levels of human exposure to radon are discussed.

Key words: charcoal canisters • indoor radon • interlaboratory comparison

Introduction

It is well known that radon and its short-lived progenies have the most impact to the population effective dose from radioactive sources [1]. Precise and accurate radon monitoring results should be the basis for the establishment of the national reference level and implementation of radon action plans. During the period February and March 2015, Laboratory for Radioactivity and Dose Measurements, University of Novi Sad, organized the first national interlaboratory comparison (ILC) of measurements of indoor radon activity concentrations according to international standard [2]. All accredited and authorized laboratories in Serbia for indoor radon measurements (there are three in total) that make use of the same method, using activated charcoal for radon adsorption according to US Environmental Protection Agency (EPA) protocol 520/5-87-005 [3], participated in the measurements. Passive charcoal canisters are dominantly used in Serbia for surveying radon levels in homes. This indoor radon ILC has been designed to identify analytical problems, if present, in transferring experience and knowledge among accredited laboratories and discussing the possibilities of radon adsorption on activated charcoal for radon dose assessment.

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Indoor radon measurements were carried out in real conditions on 14 different places (both rooms and classrooms) on the ground floor in Belgrade and Novi Sad. The charcoal canisters from each laboratory were exposed at the same time in the same place close to each other, thus providing the same conditions of exposure. The activities of radon concentrations were determined by gamma spectrometry in all three participating laboratories on high-purity germanium (HPGe) and sodium iodide (NaI) detectors independently. Each laboratory corrected its results with calibration and adjustment factors according to standard measurement procedure.

Materials and methods

Activated charcoal has a porous structure and an extremely large surface, making it suitable for adsorption of radon from the air [4]. Metal canisters with dimensions of the order of 10 cm filled with 70 g of activated charcoal were used to collect ^{222}Rn over a period of 2–3 days (Fig. 1). The typical time between the sealing of canisters and the beginning of the measurement was about 2 h, which is needed to establish the equilibrium between radon and its progeny. The activity concentrations of radon in charcoal canisters were measured from relative intensity of gamma rays emitted from short-living radon daughters ^{214}Pb and ^{214}Bi (^{214}Pb lines – 295 keV and 352 keV; ^{214}Bi lines – 609 keV and 1740 keV). Gamma spectrometry measurements could be carried out using both the high-resolution HPGe detector and NaI(Tl) scintillation spectrometer with a low level shield and known efficiency of gamma detection. In order to achieve 5% statistical accuracy at 100 Bq/m^3, the time of measurement was usually 1 h. Efficiency calibration of spectrometry systems was performed using certified EPA standards for radium activity and background canisters. By dividing the net count rate [counts/ks] by the known activity [Bq] of the standard, the efficiency [counts/(ks·Bq)] of the detector for that day was determined.

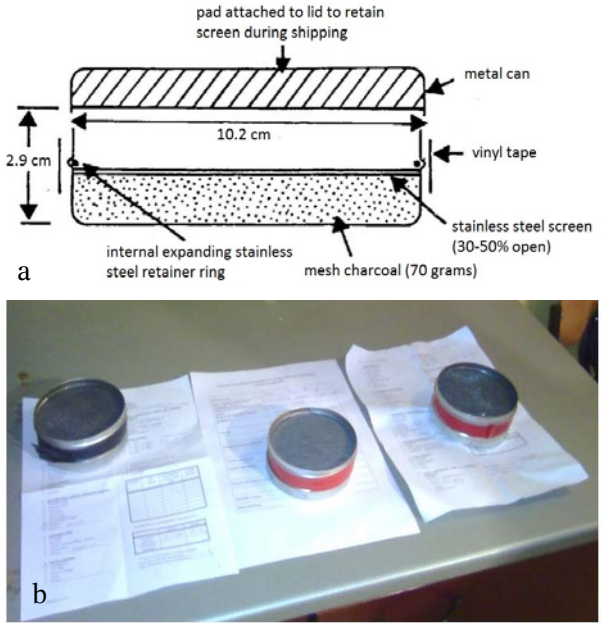

Fig. 1. (a) Charcoal canister assembly [3] and (b) photo of canisters' exposure during interlaboratory comparison in 2015.

In real conditions, charcoal's power of adsorption depends on the time of exposure and must be corrected for water content (i.e. difference between canister mass after sampling and mass before sampling). Based on data obtained from manufacturer for the used series of canisters, calibration curves of humidity for different times of exposures and different water gain were plotted in the software packages: Table Curve and Mathematica were used to determine the best mathematical fit of these functions (Fig. 2).

If the exposure period was longer than 48 h, the initial calibration factor (CF) had to be adjusted by using the adjustment factor (AF) curves representing the dependence of the adsorption rate from time of exposure. The AF curves were generated for low (20%), medium (50%) and high (80%) humidity according to criteria given in Table 1.

The activity concentrations of radon were calculated using the formula:

Table 1. Criteria for the selection of the appropriate adjustment factor curve

Canister water content [g]	Humidity [%]
<1.0	20
1.0–4.0	50
>4.0	80

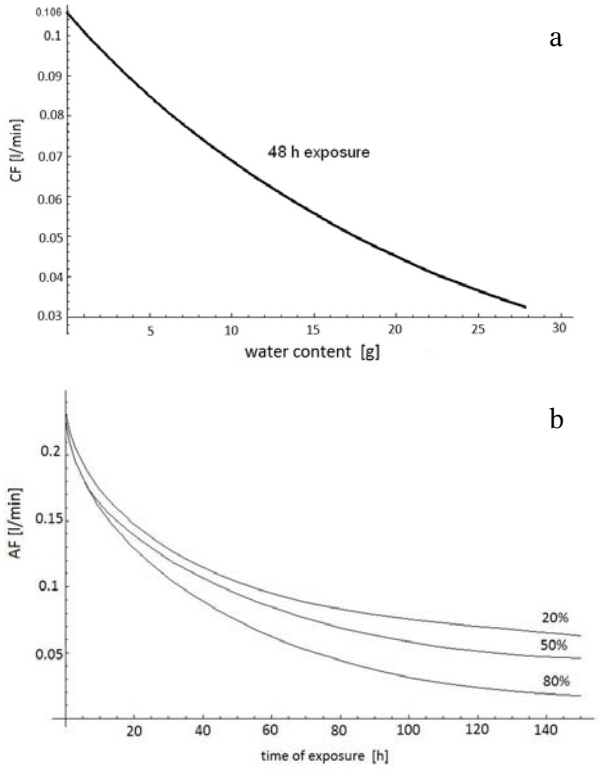

Fig. 2. The curves of humidity for different times of exposures and different water gains: (a) calibration factor curves for a 2-day exposure; (b) adjustment factor curves for low (20%), medium (50%) and high (80%) humidity of charcoal.

$$RN = \frac{I - I_F}{T_S \cdot E \cdot \mathrm{CF} \cdot \mathrm{DF}} \quad (1)$$

where: RN is radon concentration [Bq/m^3], I is total intensity of 295, 352, and 609 keV gamma ray energies [counts/ks], I_F is background spectral intensity [counts/ks], T_S is time of exposure [ks], E is detection efficiency [counts/(ks·Bq)], CF is calibration factor in [l/min] or [m^3/ks] and DF is decay correction, which could be estimated according to the equation:

$$\mathrm{DF} = e^{-\frac{0.693t}{T_{1/2(\mathrm{Rn})}}} \quad (2)$$

where t is time in days from the midpoint of exposure to the time of measurement and $T_{1/2(\mathrm{Rn})}$ is radon half-life (3.824 days).

All three laboratories made use of the same commercial canisters and EPA addendum calibration curves received from manufacturer. Detection efficiencies E for HPGe and NaI(Tl) instruments of different laboratories were different. The efficiencies of each gamma spectrometry system were determined on the date of measurement using the EPA radium standard.

Results of radon measurements

All 14 canisters from each laboratory were exposed under the same conditions, placed closely side-by-side in 14 different indoor places (living rooms and classrooms) on the ground floor in Belgrade and Novi Sad city. The activity concentrations of radon were measured in three laboratories separately and each laboratory was requested to report the results together with the corresponding combined standard uncertainties expressed as the square root of the sum of variances of all known sources of measurement uncertainty (Table 2). As the reference values for indoor radon concentrations in monitored places were not known, the average values RN_{AV} were calculated with standard deviations (σ [Bq/m^3]).

In the case of monitored places in Novi Sad numbered as 10, 11, 12, 13 and 14, the Laboratory for Radioactivity and Dose Measurements from Novi Sad had a problem with transport of exposed canisters to laboratories in Belgrade and too much time passed in order to precise determine indoor radon concentrations. This is one more problem with the charcoal canister method – safe and on-time transport to laboratory for gamma spectrometry measurements. Before the comparison of results, the laboratories harmonized the methods of uncertainty evaluation and also recalibrated the activity of standard canisters. Figure 3 shows the comparison of reported results for all 14 monitored places.

Statistical tools for data evaluation and results of ILC

The first step towards the evaluation of data is detecting and removing outliers – the values that significantly differ from the reference average. Maybe it was the wrong solution, because the true values of indoor radon concentrations were not known, and on the other side, outliers may be due to random

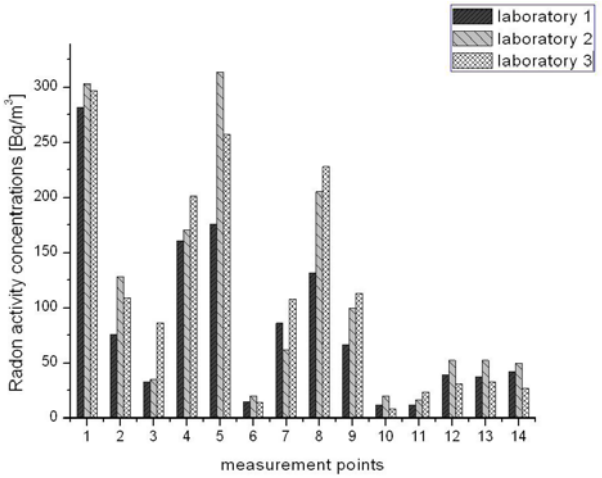

Fig. 3. Comparison of the results of all 14 monitored places.

Table 2. Reported results of indoor radon measurements with calculated average values as reference values with standard deviations (σ [Bq/m^3])

No.	Canister codes in the order of laboratories	RN_1 [Bq/m^3]	RN_2 [Bq/m^3]	RN_3 [Bq/m^3]	RN_{AV} [Bq/m^3]	σ [Bq/m^3]
1	175, 65, B16	282 ± 51	303 ± 37	297 ± 32	294	11
2	176, 66, B17	76 ± 14	128 ± 16	109 ± 12	104	26
3	178, 67, B18	33 ± 6	35 ± 11	86 ± 4	51	30
4	188, 68, B19	161 ± 29	170 ± 22	201 ± 6	177	21
5	194, 69, B20	176 ± 32	314 ± 36	257 ± 28	249	70
6	197, 70, B21	<15	<20	14.3 ± 1.1	–	–
7	163, 88, B10	86 ± 15	62 ± 10	108 ± 4	85	23
8	145, 63, B09	132 ± 24	205 ± 24	228 ± 7	188	50
9	165, 89, B08	67 ± 12	100 ± 13	113 ± 4	93	24
10	173, 63, B07	<12	<20	<8	–	–
11	162, 72, B06	<12	<17	23.5 ± 1.5	–	–
12	137, 45, B11	<39	<52	31 ± 4	–	–
13	105, 74, B12	<37	<52	33 ± 4	–	–
14	128, 79, B13	42 ± 8	<50	27 ± 3	–	–

Table 3. Grubb critical values for $n = 3$

n	Grubb critical values			
	One largest/smallest		Two largest/smallest	
	Upper 1%	Upper 5%	Lower 1%	Lower 5%
3	1.155	1.155	–	–

variation or an indication something scientifically interesting. However, we used the Grubbs' test based on the assumption of normal distribution of data to remove significant outliers.

The values of G parameter that determines the deviation of the maximum and minimum results $RN_{min/max}$ from the averages RN_{AV} for each monitored place were calculated using the formula:

$$G = \frac{\left| RN_{\frac{\min}{\max}} - RN_{AV} \right|}{\sigma} \tag{3}$$

The critical limit for Grubbs' test (Table 3) for one-sided test [5] is defined as:

$$G_{crit} = \frac{(n-1)t_{crit}}{\sqrt{n\left(n-2+t_{crit}^2\right)}} \tag{4}$$

where t_{crit} is the critical value of the t distribution with $(n - 2)$ degrees of freedom and significance level of α/n (α = 5% or 1%). Thus, the null hypothesis is rejected if $G > G_{crit}$ (5%).

Three statistical parameters: relative bias, z-score value and u-test value were used to compare the reported results of the participating laboratories to the reference values. The relative bias between the reported values and reference values expressed as a percentage were calculated using the formula:

$$\text{Relative bias} = \frac{\text{Value}_{lab} - \text{Value}_{ref}}{\text{Value}_{ref}} \times 100\% \tag{5}$$

The z-score values were calculated from the laboratory results Value_{lab}, the reference averages Value_{ref} and standard deviations of reference values σ:

$$z_{score} = \frac{\text{Value}_{lab} - \text{Value}_{ref}}{\sigma} \tag{6}$$

The target values for the standard deviation (σ) have been assigned on the basis of the reproducibility standard deviation (the standard deviation of the consensus made after outlier rejection). The laboratory performance was evaluated as satisfactory if $|z_{score}| \leq 2$; questionable for $2 < |z_{score}| < 3$, and unsatisfactory for $|z_{score}| \geq 3$.

The value of the u-test score was calculated according to the following equation:

$$u_{test} = \frac{\left|\text{Value}_{ref} - \text{Value}_{lab}\right|}{\sqrt{\sigma_{ref}^2 + \sigma_{lab}^2}} \tag{7}$$

The calculated u-test values were compared with the critical values listed in the t-statistical tables to determine if the reported result differed significantly from the expected value at a given level of probability [5].

For this ILC, we set the limiting value for the u-test parameter to 2.58 to determine the acceptance criteria for accuracy ($u_{test} \leq 2.58$) according to the International Atomic Energy Agency recommendations [6]. In that case, the reported result probably does not differ significantly from the reference value. Table 4 shows the comparison of laboratory results against reference averages.

Results discussion and conclusions

The overall evaluation of national indoor radon ILC results of 2015 showed that only one evaluated result did not fulfill the acceptance criteria for accuracy. This value was rejected as 'an outlier', although there was no real explanation of this deviation. Laboratory 3 had successfully participated in several international ILCs and achieved acceptable results [7–9]. Problem with the reference value could be avoided by exposition in radon chamber or using the active device in simultaneous regime with charcoal passive devices to estimate or obtain the reference value and that are the recommendation for the next intercomparisons. We also concluded that a larger number of charcoal canisters exposed to the same conditions from each laboratory would improve statistical evaluation. The distribution of u-test score results are shown in Fig. 4. Good agreement of results proves conformity assessment with standards and also stability of the performance of analytical systems in these laboratories. Applied statistical technique seems to be appropriate for data evaluation. Adsorption by charcoal is fast, inexpensive, but also enough precise and repeatable for fast

Table 4. Evaluated results of three different laboratories

Monitored places	Laboratory 1			Laboratory 2			Laboratory 3		
	Relative bias [%]	z-score	u-test score	Relative bias [%]	z-score	u-test score	Relative bias [%]	z-score	u-test score
1	4.1	1.09	0.23	–3.1	–0.82	0.23	–1.0	–0.27	0.09
2	26.9	1.08	1.32	–23	–0.92	0.79	–4.8	–0.19	0.17
3	2.9	0.71	0.16	–2.9	–0.71	0.09	–153	–37.14	12.27
4	9.0	0.76	0.45	4.0	0.33	0.23	–13.6	–1.14	1.10
5	29.3	1.04	0.95	–26.1	–0.93	0.83	–3.2	–0.11	0.11
6	–1.2	–0.04	0.04	27.1	–1.00	0.92	–27.1	–1.0	0.99
7	29.8	1.12	1.01	–9.0	–0.34	0.31	–53.2	–0.8	0.79
8	30.0	1.08	0.97	–7.5	–0.29	0.26	–21.5	–0.83	0.82

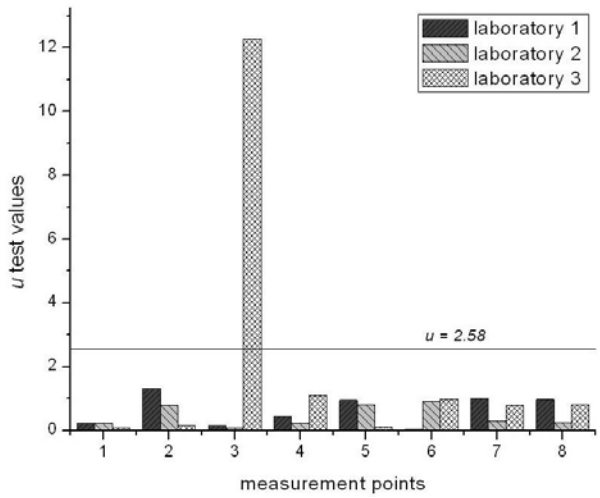

Fig. 4. Distribution of *u*-test values for eight monitored places.

judgment of indoor radon concentration, of course if the laboratory is equipped with a gamma spectrometry system. The possible application of this method could be in the selection of measuring sites for radon surveys, and mapping and exploring the prone areas to estimate human exposure to radon.

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