SIMULTANEOUS MEASUREMENTS OF RADON, THORON, AND EQUILIBRIUM EQUIVALENT CONCENTRATIONS IN FAMILY HOUSE – SINGLE CASE STUDY

by

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Scientific paper
https://doi.org/10.2298/NTRP190128017L

Simultaneous indoor measurements of radon, thoron and equilibrium equivalent concentrations, by three different continuous radon monitors in real ambient conditions, were performed and compared during two weeks period. Radon concentrations varied from 153 Bqm^{-3} to 870 Bqm^{-3} with variations of thoron concentrations in the range (2.17-219) Bqm^{-3}. Obtained arithmetic means of equilibrium equivalent radon and thoron concentrations were (130 50) Bqm^{-3} and (23 18) Bqm^{-3}, respectively. The inter-comparison of active radon devices was focused on consistency of results between themselves and the influence of environmental conditions on the behavior of each detector system. The obtained statistically significant correlation between the results of used monitors, validates them for application in radon diagnostic measurements as the first step in radon remediation and dose assessment.

Key words: radon, thoron, equilibrium equivalent radon concentration, equilibrium equivalent thoron concentration

INTRODUCTION

Exposure to radon, thoron and their progenies gives important amount of dose from ionizing radiation in indoor environment [1]. More detailed exploration is especially important in dwellings, where previously increased radon concentration has been detected. It is well known that higher concentrations of radon or thoron lead to the increase of their progenies in air. Due to different half-lives of radon and thoron, 3.82 days and 55.6 seconds, respectively, their distributions in indoor air are different. It is considered that the distribution of indoor radon is homogeneous, but thoron concentration exponentially decreases from its sources in the room (walls, floor *etc.*) [2, 3], even for the high rate of air-flow inside the room [4]. However, radon and thoron progenies have similar behavior in indoor air due to the relatively long half-lives.

According to EU directive, which implements basic safety standards for protection against the dangers arising from exposure to ionizing radiation [5], all member states (including candidate countries) shall establish the reference level for radon in dwellings and workplaces not to exceed 300 Bqm^{-3} and the national action plan for controlling public exposure due to radon indoors. Therefore, short-term indoor radon screening measurement by continuous monitors, followed by long-term measurement, is needed in dwellings where radon gas concentrations are near or above reference level [6]. For remediation of such residential areas and workplaces, it is necessary to use some active technique for finding radon and thoron paths of entry. Moreover, the use of passive detectors is associated with a large uncertainty due to inadequate handling and placement of detectors.

This work is a result of comprehensive study in one family house, where concentration of radon exceeded the level of 400 Bqm^{-3}, which represents the action level for average annual radon concentration in indoor air of existing dwellings, according to currently valid National Rulebook (Official Gazette RS 86/11 and Official Gazette RS 50/18). Previous measurements were performed several times by passive detectors, such as CR-39 and charcoal canisters [7, 8]. Pres-

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ent survey measurements were taken during two weeks in November 2018. Such a detailed analysis of the individual case emerged as a need for a better understanding of the behavior of radon, thoron and their progenies, over a longer period of time, under controlled conditions in one room.

Direct measurement and continuous monitoring of indoor radon and thoron progeny concentrations are very important in order to estimate their contribution to dose and to the precise determination of the equilibrium factors. Radon equilibrium factor F_{Rn} is defined as the ratio of the equilibrium equivalent radon concentration *EERC* and actual radon concentration C_{Rn} [9]

$$F_{\mathrm{Rn}} = \frac{EERC}{C_{\mathrm{Rn}}} \tag{1}$$

The same relation stands for thoron, Tn, equilibrium factor F_{Tn}, given by the ratio of the equilibrium equivalent thoron concentration *EETC* and actual thoron concentration C_{Tn}

$$F_{\mathrm{Tn}} = \frac{EETC}{C_{\mathrm{Tn}}} \tag{2}$$

MATERIALS AND METHODS

Simultaneous measurements of radon, thoron, *EERC* and *EETC*, in real indoor environment, were conducted continuously during November 2018, in a closed room with poor ventilation (<0.5 h^{-1}). The room was selected based on previous radon measurements and the detected, elevated radon level. The room, of approximately 75 m^3 volume, is a part of an old house built in the 1930s, constructed by adobe and bricks, with wooden floor and windows and without any concrete slab below the wooden floor. It was interesting to investigate *EERC* and the presence of thoron and *EETC* in indoor air. The measurement of thoron concentration is not as unambiguous as the measurement of the radon concentration and is highly dependent on the position of the instrument on which it is located in the room during the measurement [10]. Also, indoor thoron concentration is highly influenced by building material, used for the construction and covering, on the contrary to radon, where building material is a less dominant factor [11]. Therefore, simultaneous measurements of thoron gas, by RAD7 and RTM1688-2 active monitors, have been carried out at several positions in the room. For this study, three different radon/thoron monitors were used: AlphaGuard PQ2000 PRO, RPM2200 with RTM1688-2 (Sarad GmbH) and RAD7 (DURRIDGE Company). All active detectors have integrated sensors dedicated for temperature, relative humidity and atmospheric pressure measurements. AlphaGuard principle of detection is based on a pulse ionization chamber with an active volume of 0.56 dm^3. The unit AlphaPM was connected as front-end to AlphaGuard for simultaneous monitoring of radon and its decay products. RTM1688-2 monitor is an alpha spectrometer with semiconductor detector insensitive to ambient humidity. EERC and EETC were determined with RPM2200 by detection of the alpha activity collected on a membrane filter (1 m pore size), with constant air-flow. Differentiate mode is used for hourly determination of EETC, because of the long half-lives of thoron progenies until the ^{212}Po, the point in the decay chain at which such alpha-spectrometry measurements are possible. RAD7 is a commonly used, semi-conductor alpha-spectrometer, suitable to distinguish radon and thoron. Precise measurement is provided by constant fresh air supply into an active chamber, after removing the moisture by a drying unit connected to the device inlet.

AlphaGuard and RAD7 instruments were calibrated at the accredited trial metrological Lab. SUJCHBO Kamenna, Czech Republic, in 2015 and SARAD instruments were calibrated by the manufacturer in 2018. All instruments participated with satisfactory results in the 2018 NRPI Inter-comparisons of Radon gas Continuous Monitors and 2015 NRPI Intercomparisons of Radon gas Measurement Instruments at SURO v.v.i. Institute, Prague, Czech Republic within the IAEA Technical Cooperation Projects RER/9/153 and RER 9127.

RESULTS AND DISCUSSIONS

Descriptive statistics of continuous measurements of: radon concentrations C_{Rn}, thoron concentrations C_{Tn}, equilibrium equivalent concentrations for radon EERC and thoron EETC, respectively and equilibrium factors for radon F_{Rn} and thoron F_{Tn}, respectively, are given in tab. 1. All data were collected on an hourly basis and for statistical evaluations the first three records have not been taken into account.

Exceeded theoretical value of 1 for thoron equilibrium factor (tab. 1) is probably due to the fact that in certain measurements, thoron concentration was measured in the central position of the room, at a height of 1 m from the floor, which will lead to a small thoron concentration, due to its short half-life and therefore, to a large measurement uncertainty. Consequently, this will introduce a large uncertainty in the assessment of the equilibrium factor as well, and directly affect the arithmetic mean of thoron equilibrium factor, which exceeds even the radon equilibrium factor in this study. In order to avoid that and obtain more realistic thoron concentrations, measurement should be carried out in close proximity to the possible source of indoor thoron (walls, floor or cracks).

Comparison of indoor radon concentrations continuously monitored by AlphaGuard, RTM1688-2 and RAD7 are shown in fig. 1. Elevated arithmetic mean of radon concentration, obtained by RAD7 in-

Table 1. Descriptive statistics (minimum and maximum value and arithmetic mean (AM) with standard deviation) of C_{Rn}, C_{Tn}, *EERC*, *EETC*, F_{Rn}, and F_{Tn} for the used active monitors

Monitor		C_{Rn} [Bqm^{-3}]	C_{Tn} [Bqm^{-3}]	*EERC* [Bqm^{-3}]	*EETC* [Bqm^{-3}]	F_{Rn}	F_{Tn}
AlphaGuard PQ2000 PRO with AlphaPM	Min	167	–	26	–	0.07	–
	Max	766	–	266	–	0.41	–
	AM	473 136	–	130 50	–	0.28 0.05	–
RTM 1688-2 with RPM2200	Min	153	18	22	3.3	0.07	0.046
	Max	844	219	323	94.2	0.60	1.04
	AM	430 153	64 35	125 64	23 18	0.24 0.09	0.32 0.26
RAD7	Min	171	2.17	–	–	–	–
	Max	870	178	–	–	–	–
	AM	549 163	39 40	–	–	–	–

strument, could be explained by the fact that RAD7 was operational only for the first 170 hours from the beginning of a measurement, when the indoor radon concentrations were higher, fig. 1. Ambient conditions such as: pressure, relative humidity and temperature in the investigated room, are shown in fig. 2. From the graphics, it can be concluded that during the test period there were no major deviations in temperature and relative humidity of indoor air, significant deviations were registered only for air pressure. Air pressure ranged from 100.017 kPa to 102.159 kPa, fig. 2. Recent publications show close relationship between radon concentrations and indoor vapor pressure variations [12].

A Pearson's correlation was run to estimate the relationships between indoor radon concentration and three independent variables (*i. e*., temperature, pressure, and humidity). Preliminary analyses show that the relationship is linear with all variables normally distributed, as assessed by Shapiro-Wilk's test ($p > 0.05$), and there were no outliers. Moderate positive, but statistically significant correlations are found between: radon concentration and temperature, $r = 0.532$, $p < 0.0005$; radon concentration and pressure, $r = 0.304$, $p < 0.0005$,

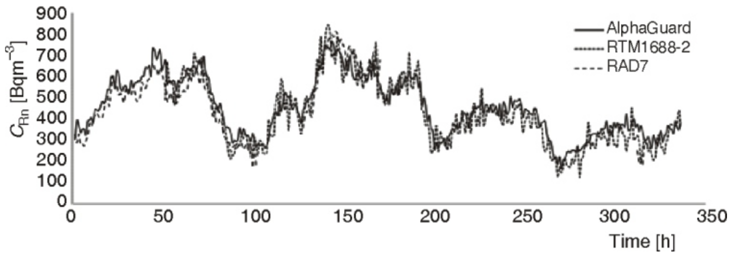


Figure 1. Time variation of C_{Rn} continuously monitored by three active detectors

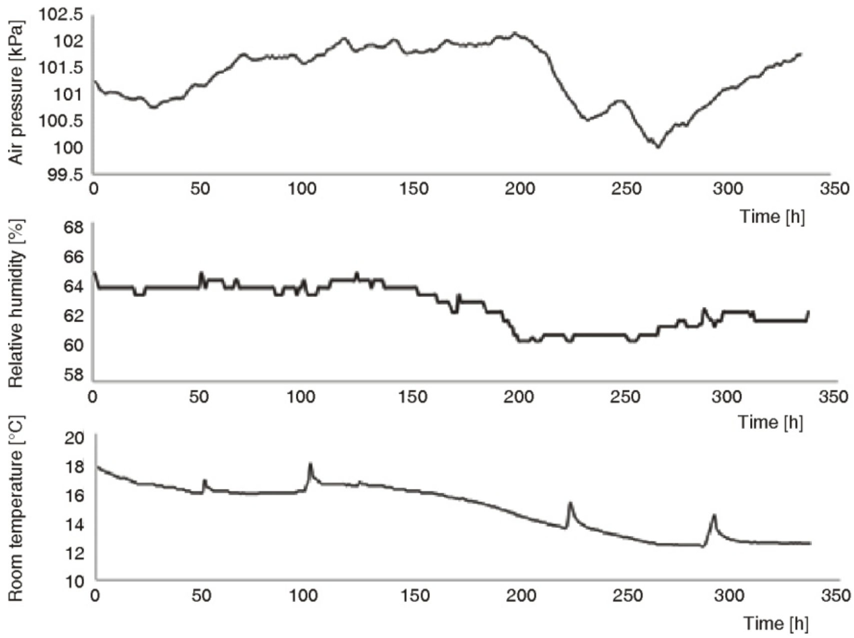


Figure 2. Time variation of pressure, relative humidity, and air temperature during the measurement

and radon concentration and humidity, $r = 0.497$, $p < 0.0005$. To predict the value of indoor radon from temperature, pressure and humidity, a multiple regression was run. There was linearity, as assessed by partial regression plots and a plot of standardized residuals, against predicted values. There was no evidence of multi-collinearity, as assessed by tolerance values greater than 0.1. The assumption of normality was met, as assessed by a *Q-Q* Plot. The multiple regression model statistically significantly predicted radon concentration, F(3.329) = 46.275, $p < 0.0005$, explained 30 % of the variability of radon concentration. All three variables added statistically significantly to the prediction, $p < 0.05$. Regression coefficients and standard errors can be found in tab. 2.

Since there were no repeated measurements in the comparison, the results can be compared graphically using a modified Youden's plot, figs. 3-5, that easily identifies the systematic and random errors of particular instruments [13]. The inner square of the plot represents 2σ and the outer square represents 3σ. All the measured values fall within the outer rectangle and are thus considered acceptable. Points that lie near the 45° reference line, but outside 2 square, indicate a systematic error, probably caused by different sensitivity of instruments on the change of environmental conditions. As it can be noticed, there are no random errors (points that lie far from the 45 ° reference line).

The RAD7 and RTM1688-2 devices have the ability to measure indoor thoron concentration, C_{Tn}, as well as radon concentration, fig. 6. Discrepancy of thoron concentrations, in these measurements, is a consequence of seven different experimental set-ups of active monitors, at five positions in the examined room, during the measurement:

– from 0 h to 48 h: only RAD7 was operational one meter above the floor and 30 cm close to the wall 1 – position 1.
– from 50 h to 97 h: two devices operated simultaneously (RAD7 at the same position – 1 and RTM1688-2 at the central position in the room, one meter above the floor – position 2),
– from 98 h to 121 h: only RTM1688-2 was operational but on the floor with minimal possible distance from the wall 1 (about 15 cm), 1 m from the corner of the room – position 3,
– from 122 h to 170 h: RAD7 and RTM1688-2 operated side-by-side on the floor with minimal possible distance from the wall 1 depending on instrument inlet set-up (5 cm for RAD7 and 15 cm for RTM1688-2), 1 m from the corner of the room – position 3,
– from 171 h to 218 h: RAD7 and RTM1688-2 operated side-by-side on the floor close to the wall 2, 1.5 m from the corner of the room – position 4,
– from 219 h to 260 h: only RTM1688-2 was operational at the central position in the room, 1 m above the floor – position 2, and

Table 2. Summary of multiple regression analysis, $p < 0.05$

Model	Unstandardized coefficients		Standardized coefficients
	B	Std.Error	Beta
1 (Constant)	–3208.288	1355.893	
Temperature	28.216	7.363	0.347
Pressure	2.201	1.279	0.089
Humidity	16.033	8.150	0.171

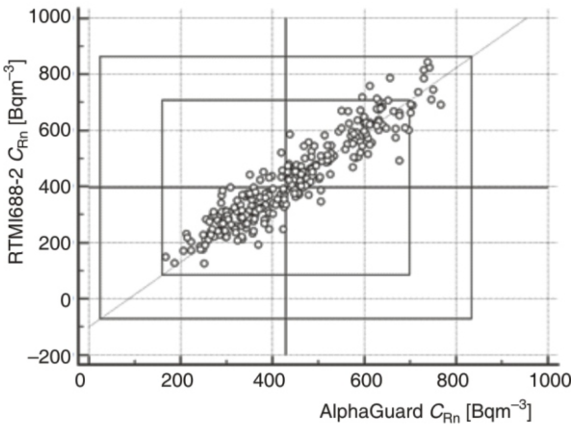


Figure 3. Youden's plot of RTM1688-2 and AlphaGuard radon concentrations

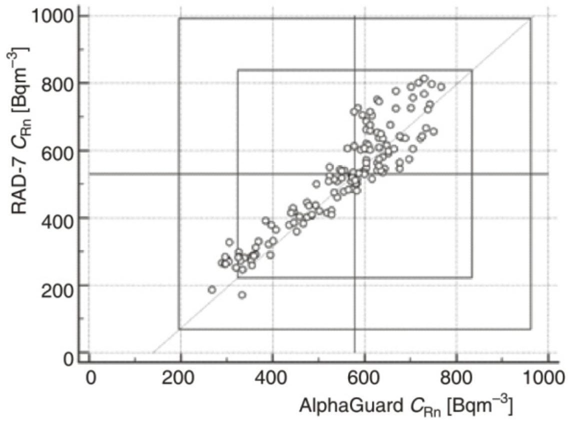


Figure 4. Youden's plot of RAD7 and AlphaGuard radon concentrations

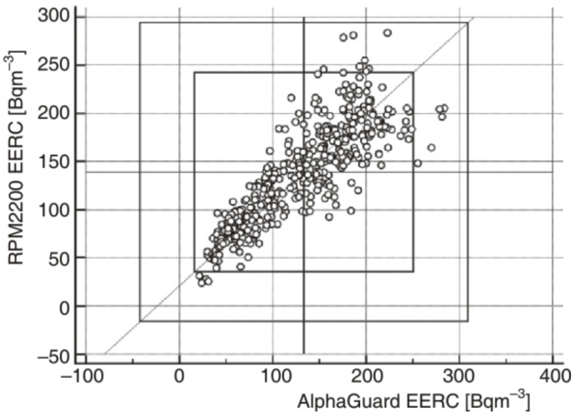
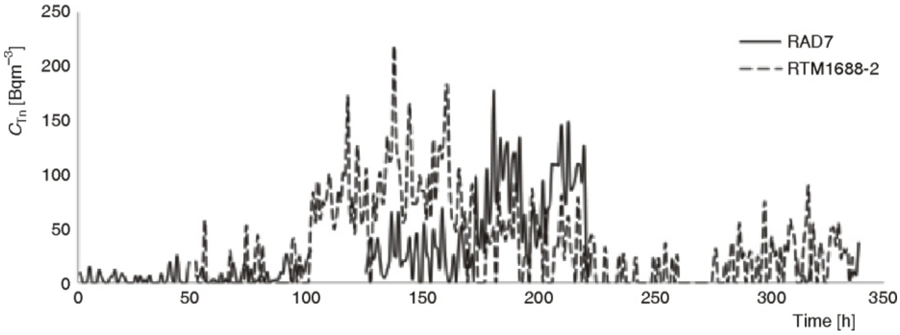


Figure 5. Youden's plot of RPM2200 and AlphaGuard EERC concentrations

Figure 6. Comparison of indoor thoron concentrations measured by RAD7 and RTM1688-2 active monitors



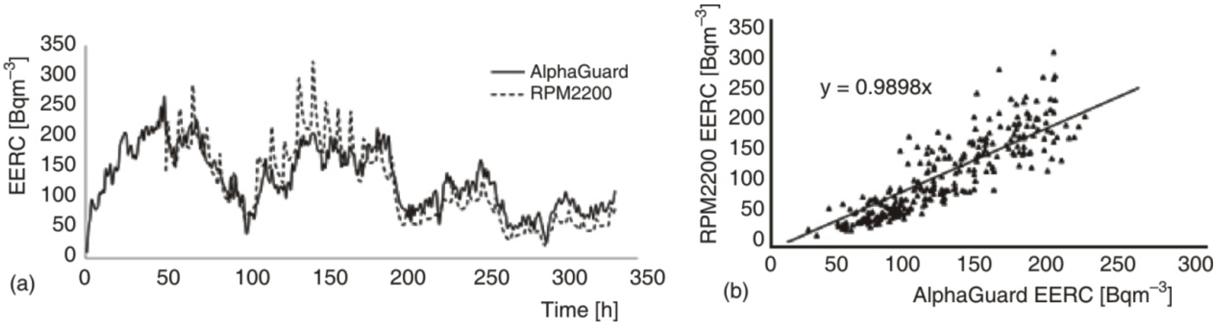


Figure 7. (a) Time variation of EERC continuously monitored by AlphaGuard and RPM2200 and (b) linear correlation of the obtained results

– from 275 h to 340 h: RAD7 and RTM1688-2 operated side-by-side at about 1.2 m above the floor, close to the wall 2, 1.5 m from the corner of the room – position 5 (fig. 6).

Due to the fast increase in Tn concentrations when the instruments were positioned on the floor (especially close to the wall 2), we concluded that the dominant source of thoron in the room is soil beneath the wooden floor. Large discrepancies between RAD7 and RTM 1688-2 results could be explained by different distances from the floor of fixed inlets as thoron sampling points (for RAD7, inlet is about 5 cm above the floor and for RTM 1688-2 instrument, inlet tube is about 30 cm above the floor). According to some authors [14, 15] if the detector system is placed closer than 10 cm to the wall, measured Tn concentration mainly represents exhalation from the wall. Measured thoron concentrations are in good agreement with the results of the first indoor thoron survey in our region, which was carried out using UFO and RADUET detectors in houses of Kosovo and Metohija, Serbia, in three series between 2003 and 2011, with wall distance from 10-30 cm [16].

Comparative measurements of EERC were done by AlphaGuard and RPM2200, and the results are shown in fig. 7. Linear coefficient of correlation between EERC, measured by AlphaGuard, and RPM2200 has the value of 0.9898, fig. 7, which shows a good agreement of the results obtained by two devices. Along with measuring of EERC, the device RPM2200 measured EETC, and the obtained measurement values are shown in fig. 8. Examining the graphics it can be concluded that EETC has the same trend as the *EERC*. However, from the graph 8, it can be noticed that there are some periodical variations of *EETC* results on every 5-6 hours. We assume that the cause of these variations is hourly-used differentiate mode for determination of *EETC.* This can be corrected by selecting a time interval of approximately 5 hours. This period is necessary to reach the saturation of ^{212}Po, thoron progeny that is alpha emitter, used for *EETC* determination. The obtained ratio of *EETC* to *EERC* with an arithmetic mean of 0.215 is in good line with a worldwide range of 0.01-0.5 [1].

Figures 9 and 10 show the comparisons of radon gas concentrations and *EERC*, monitored by AlphaGuard and by SARAD instruments (RTM1688-2 and RPM2200), respectively. It can be noticed that both concentrations exhibit the same trend of increase and decrease over time, for both instrument set-ups.

Good correlations between radon concentrations and EERC are a consequence of the stable indoor radon equilibrium. The obtained similar average values, for FRn of (0.28 0.05) for AlphaGuard and (0.24 0.09) for SARAD instruments, are in good agreement with UNSCEAR value of 0.4 [1]. Since the examined house is located in a non-urban district, even better agreement is achieved with typical published values for houses in Europe (0.40 for towns and 0.32 for villages) [10].

A multiple Pearson's correlation was run to estimate the relationships between indoor radon C_{Rn}, indoor thoron C_{Tn} and their progenies, *EERC* and *EETC*, for AlphaGuard and SARAD's instruments, tab. 3. All correlations are statistically significant, but, there is a weak correlation between indoor thoron and EETC.

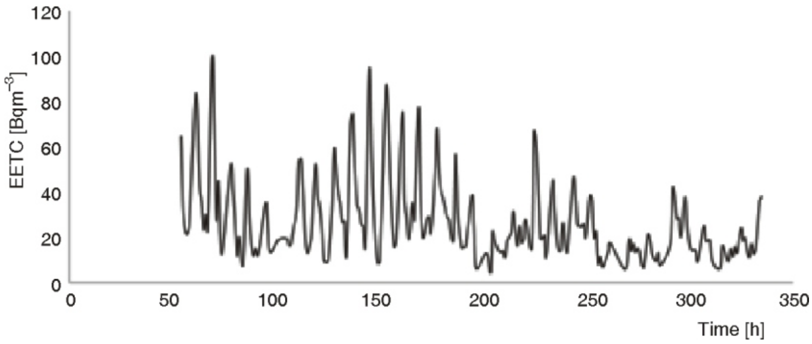


Figure 8. Time variation of EETC measured by RPM2200 active monitor

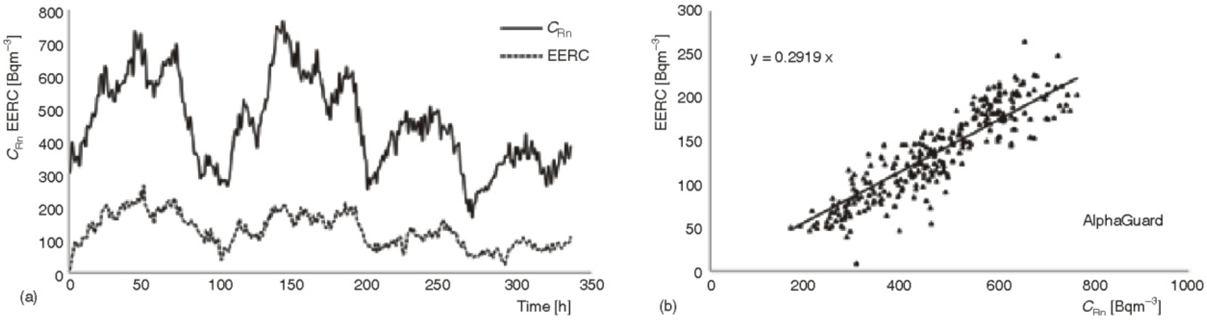


Figure 9. (a) Time variation of radon gas concentration and EERC monitored by AlphaGuard instrument and (b) linear correlation of the obtained results

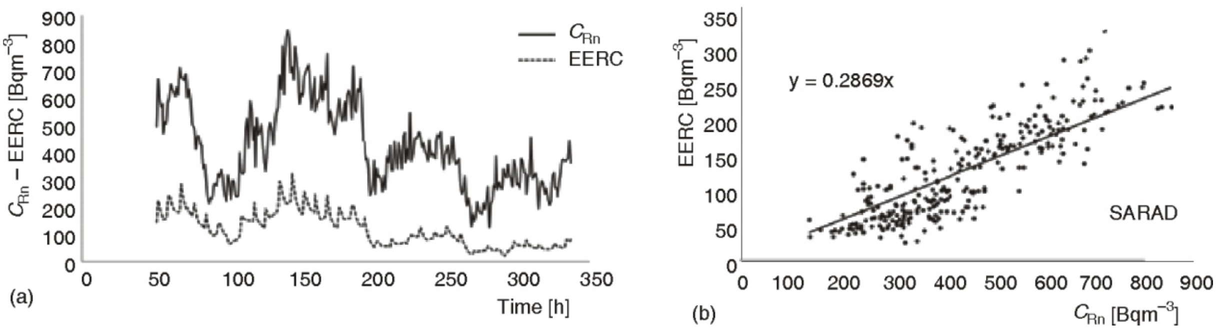


Figure 10. (a) Time variation of radon gas concentration and EERC monitored by SARAD instruments (RTM1688-2 and RPM2200) and (b) linear correlation of the obtained results

This confirms previously introduced assertion that thoron determination is strongly affected by the position of the monitor in the room, which is not the case for measurement of homogeneously distributed thoron progenies. Otherwise, correlation coefficients, for the data obtained in cases when monitors were operated side by side (EERC and radon concentration for AlphaGuard and EERC and radon concentration for SARAD instruments – see tab. 3), are close to 1.

CONCLUSIONS

Indoor radon, thoron, *EERC*, and *EETC* variation in a single room, in the same period (November 2018), was investigated for two weeks. Long term measurement, with passive detectors, is appropriate for general assessment, but, more detailed research must be performed in the case of elevated radon levels. The present study proved that with active radon/thoron detectors, it is possible to monitor daily and weekly variations of radon and thoron concentrations and to search radon and thoron equilibrium, based on the behavior of their progenies.

On the contrary to passive detectors, application of active monitors allows measurement of thoron concentrations in more than one position in the room and therefore, enables finding of thoron potential sources. Once the sources of thoron in the room are identified, special attention should be paid to the distance of in-

Table 3. Pearson correlations for studied variables – AlphaGuard instrument and SARAD instruments (RTM1688-2 and RPM2200)

	C_{Rn} (AlphaGuard)	C_{Rn} (RTM1688-2)	*EERC* (AlphaGuard)	*EERC* (RPM2200)	*EETC* (RPM2200)	C_{Tn} (RTM1688-2)
C_{Rn} (AlphaGuard)	1					
C_{Rn} (RTM1688-2)	0.960**	1				
EERC (AlphaGuard)	0.896**	0.874**	1			
EERC (RPM2200)	0.864**	0.850**	0.874**	1		
EETC (RPM2200)	0.406**	0.411**	0.355**	0.402**	1	
C_{Tn} (RTM1688-2)	0.386**	0.421**	0.264**	0.435**	0.124*	1

**correlation is significant at the 0.01 level (2-tailed), * correlation is significant at the 0.05 level (2-tailed)

strument inlet from the walls or the floor, which should be set to approximately 10 cm.

Good agreement of correlations given in tab. 3, for two different active monitors, is another confirmation of their reliability, and suitability for investigation of radon and thoron equilibrium in indoor air.

Despite the numerous problems and limitations that are associated with thoron and their progeny behavior, this study shows that active measurements of thoron are important, but still difficult for comprehension, and need further investigations. It would be convenient for national legislators to consider the continuous measurements as obligatory before and after remediation, which should be proposed in the future national action plan.

ACKNOWLEDGMENT

This work was supported by Ministry of Education Science and Technological Development of the Republic of Serbia, within the projects Nuclear Methods Investigations of Rare Processes and Cosmic Rays No171002, Biosensing Technologies and Global System for Continuous Research and Integrated Management No 43002 and Nuclear Physics, Methods and Applications No 171018.

AUTHORS' CONTRIBUTIONS

Manuscript was written by R. Z. Lakatoš, S. M. Forkapić and S. D. Samardžić. Figures were prepared by K. I. Bikit-Schroeder and R. Z. Lakatoš. All authors analyzed and discussed the results and reviewed the manuscript. Experimental data were obtained by R. Z. Lakatoš, S. M. Forkapić, I. T. Čeliković and V. Radolić. Theoretical analysis was carried out by D. S. Mrdja and S. M. Forkapić. Statistical analysis was carried out by S. D. Samardžić.

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Received on January 28, 2019
Accepted on April, 22, 2019

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СИМУЛТАНА ИСПИТИВАЊА РАДОНА, ТОРОНА И РАВНОТЕЖНИХ ЕКВИВАЛЕНТНИХ КОНЦЕНТРАЦИЈА ЊИХОВИХ ПОТОМАКА У ПОРОДИЧНОЈ КУЋИ – СТУДИЈА ЈЕДНОГ СЛУЧАЈА

Симултана испитивања радона, торона и равнотежних еквивалентних концентрација њихових потомака коришћењем три различита активна радонска монитора спроведена су и упоређена у реалним амбијенталним условима, у временском периоду од две недеље. Концентрације радона су варирале од 153 Bqm^{-3} до 870 Bqm^{-3}, са варијацијом концентрација торона у опсегу (2.17-219) Bqm^{-3}. Добијене су аритметичке средине равнотежних еквивалентних концентрација радона и торона од (130 50) Bqm^{-3} и (23 18) Bqm^{-3}, респективно. Интеркомпарација активних уређаја за мерење радона и торона је била фокусирана на конзистентност међусобно добијених резултат, као и на утицај спољашњих услова на понашање сваког детекторског система. Добијена статистички значајна корелација међу резултатима коришћених активних монитора потврђује њихову примену за детекцију радона у току ремедијације објеката са повишеном концентрацијом радона и у прорачуну примљене дозе.

Кључне речи: радон, торон, равнотежна еквивалентна концентрација радона, равнотежна еквивалентна концентрација торона