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Investigation of the influence of chamber construction parameters on radon exhalation rate

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Abstract. Radon exhalation from ground is a process dependent on the emanation and migration of radon through ambient air. Most studies on radon exhalation from soil were performed regarding the influence of meteorological and soil parameters. As radon exhalation rate can be affected by the internal properties of the sample, it may also be influenced by the exhalation chamber geometry such as volume-to-area (V/S) ratio or other construction parameters. The measurements of radon exhalation from soil were made using different constructions of accumulation chamber and two types of radon monitors: RAD7 (Durridge) and AlphaGUARD PQ2000PRO (Genitron). The measurements were performed on one site in two locations and approximately at the same time. The first tests did not show the correlations of exhalation rate values and the chamber's construction parameters and their geometrical dimensions. However, when examining the results, it seems that there are still too many factors that might have affected the process of radon exhalation. The future experiments are planned to be conducted in controlled laboratory conditions.

Key words: accumulation chambers • exhalation rate • measurement techniques • radon

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Introduction

Radon (^{222}Rn) is a naturally occurring radioactive gas that originates from the decay of radium (^{226}Ra) in the primordial uranium (^{238}U) decay series. As a noble gas, it easily migrates through inter-granular spaces; then, because of advection, diffusion and convection with other gasses or with water, it moves through fissures and faults until it reaches the outdoor atmosphere. Its half-life is long enough that it can migrate even to quite long distances from the place of its decay into buildings raised in a certain area. According to Nazaroff and Nero [1], this distance in soil and rocks can be as long as a few metres. This is the reason that radon and its decay products are the main source of ionizing radiation in non-industrial buildings. They are responsible for about half of the radiation dose people in Poland are exposed to [2].

The effects leading to the presence of radon in flats and buildings are emanation, transport (mainly diffusion-advection) and exhalation. All these processes were already extensively examined and they are well understood in theory ([1] and references herein). However, in practice, the parameters such as emanation coefficient, diffusion coefficient and exhalation rate that describe the mentioned effects are affected by many environmental factors [3, 4].

Radon exhalation is the complex process of its escaping from the ground or other materials (e.g. building materials) into the outdoor or indoor air. Radon exhalation rate is defined as the radon activity released from an area unit during a time unit [Bq·m^{-2}·s^{-1}]. This process is interesting and depends mainly on the factors that influence radon emanation coefficient and radon transport. These are radium (^{226}Ra) concentration in soil or rock grains, soil properties (porosity, permeability, humidity, temperature) and meteorological conditions (air pressure, air temperature, wind speed, precipitation). These relationships are not yet completely explained and are still the subject of research. The soil properties influence directly the dynamics of radon exhalation. Meteorological parameters are factors that indirectly affect the process of radon exhalation from soil.

The investigation of radon exhalation rate from soil using different techniques was presented by several researchers. Many papers describe the 'laboratory techniques' for the determination of mass radon exhalation rate from soil samples and/or from building materials samples [5–9]. The most common methods for radon registration are the solid-state nuclear track detector (SSNTD) technique, charcoal canisters, Lucas cells and the active techniques using e.g. AlphaGUARD or RAD7 radon monitors. The *in situ* measurements are also performed, and they are more interesting in terms of looking for relationships between radon exhalation rate and different parameters. Some authors described this kind of measurements but the presented results regarded only a single or short-term measurements of radon exhalation rate from soil [10–14]. The study of the relationship between radon exhalation rate from soil and meteorological conditions has been carried out by Mazur and Kozak [15] and Ferry *et al.* [16] The authors used an automatic device for long-term measurements of radon exhalation rate.

In this study, the different types of accumulation chambers have been tested for measurements and the results have been compared aiming at examining whether the construction and geometry of the chamber (especially volume-to-area (V/S) ratio or internal partition of the chamber) influence the result of the measurement.

Materials and methods

Different techniques to measure radon exhalation rate from soil were described by Mazur [17]. All of them are based on the measurement of the concentration of radon gas accumulating in a certain time inside a chamber installed on the ground. The experimental setup used in the present experiments is shown in Fig. 1. It consists of a closed circuit system with the following components: (1) the accumulation chamber; (2) radon monitor; (3) additional elements such as the air pump, the desiccant or the filter; and (4) connecting pipes. The authors used different types of the accumulation chambers (Fig. 2a–d) and two different radon monitors, that is,

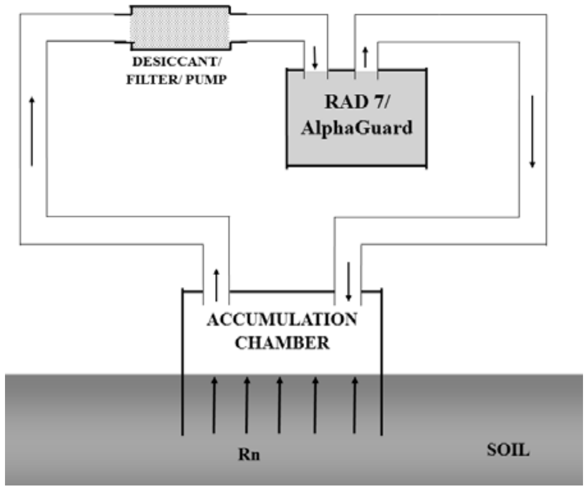

Fig. 1. The schematic diagram of radon exhalation measurement.

Fig. 2. Different accumulation chambers used in the present studies: (a) from the University of Silesia (Durridge, USA chamber) with the partition with V/S = 0.110 m; (b) from the University of Silesia with and without the partition with V/S = 0.129 m; (c) from the Institute of Nuclear Physics PAN without the partition with V/S = 0.120 m, and (d) from the Institute of Nuclear Physics PAN without the partition with V/S = 0.15 m.

RAD7 with a semiconductor detector from Durridge, USA, and the ionization chamber AlphaGUARD PQ2000PRO from Genitron, Germany. The accumulation chambers had different geometry and construction. The diameter and height of a certain chamber depend on the cost, the strength and the measured signal. The differences of our chambers were in the volumes, heights and covered areas and also in the introduction of the internal partition in two of them. The test measurements were performed on two 'study fields':

- at the Institute of Physics of the University of Silesia in Katowice (using RAD7),
- at the Institute of Nuclear Physics PAN in Kraków (using AlphaGUARD).

The radon exhalation rate E_{Rn} [$Bq \cdot m^{-2} \cdot s^{-1}$] was calculated according to the formula [18]:

$$E_{Rn} = (V/S) \cdot a \tag{1}$$

where: V – volume of the system [m^3], S – soil area covered by the accumulation chamber [m^2], V/S – a constant value, different for each chamber, a – the slope of the line fitted to the experimental points of the increasing radon concentration inside the accumulation chamber.

The uncertainty $u(E_{Rn})$ was calculated as a square root of the sum of the uncertainties in all quantities in quadrate:

$$u(E_{Rn}) = \sqrt{\left(\frac{V}{S}\right)^2 \cdot u^2 + \left(\frac{a}{S}\right)^2 \cdot u^2(V) + \left(\frac{V \cdot a}{S^2}\right)^2 \cdot u^2(S)} \tag{2}$$

Measurements were made during stable meteorological conditions and stable soil temperature in May 2015 (Katowice) and July and September 2015 (Kraków). Other meteorological parameters such as wind speed or soil humidity were not recorded because of the lack of a special equipment.

On the study field in Katowice, five experiments were performed within 5 h. All available chambers were used in each measurement day and the measurement with each chamber last for 1 h. Chambers were placed next to each other in bare ground of area of approximately 1 m^2, one by one. The same placement was repeated in the next days. The authors always chose 5 h time in the middle of the day between 10 a.m. and 3 p.m.

On the study field in Kraków, two experiments have been performed at the same time. Air temperature was recorded to ensure the lack of its changes during the experiment. In one setup, the accumulation chamber without the internal partition was used, whereas in the second setup, the accumulation chamber had the internal partition. The value of the V/S ratio was the same in both chambers. Two AlphaGUARD monitors were applied in the measurements.

The increase in radon activity concentration was registered in 15 min (Katowice) and 10 min (Kraków) time intervals within 1 h time just after the accumulation chamber has been installed on the ground. The experimental values of radon concentration [$Bq \cdot m^{-3}$] in time [s] were plotted for each experiment to obtain the factors a representing the slope of the line fitted (Eq. (1)). The examples of two graphs of build-up times of radon concentrations inside two different accumulation chambers are shown in Fig. 3.

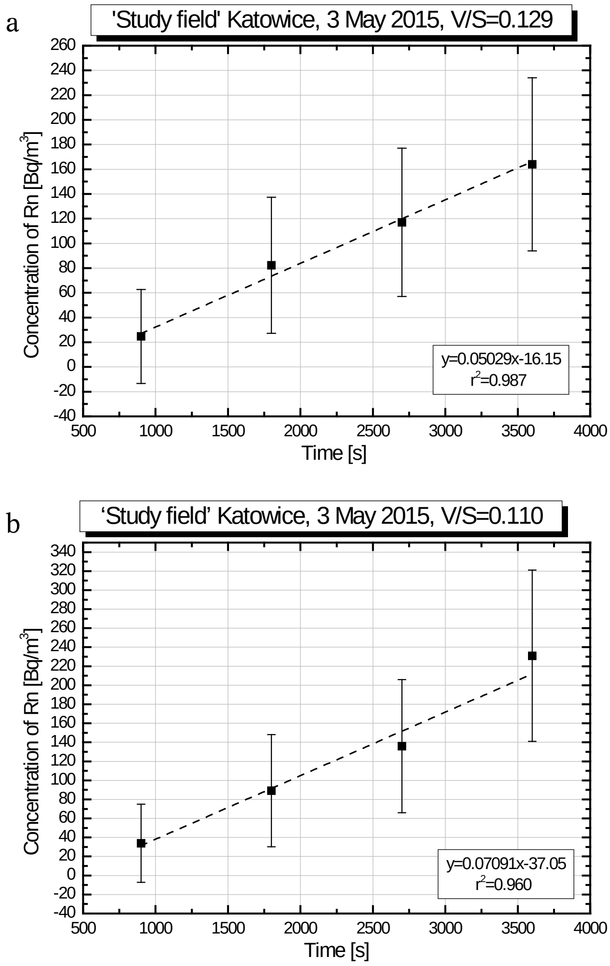

Fig. 3. Typical graphs of build-up times of radon concentration inside two different accumulation chambers. Experiments performed on the 3rd of May in Katowice, (a) with blue chamber with the partition, $a = 0.0503 \pm 0.0033$; (b) with Durridge chamber, $a = 0.0709 \pm 0.0082$. r^2 in both cases is close to 1.

Results and discussion

Having the detailed characteristics of the studied ground, one can assess the expected value of radon exhalation rate E_{Rn} [$Bq \cdot m^{-2} \cdot s^{-1}$] using the following formula [15]:

$$E_{Rn} = \varepsilon A_{Ra} \rho \sqrt{\lambda_{Rn} D} \tag{3}$$

where: ε – the radon emanation coefficient in soil [–], A_{Ra} – the radium concentration in the soil [$Bq \cdot kg^{-1}$], ρ – the average soil density [$kg \cdot m^{-3}$], λ_{Rn} – the radon (^{222}Rn) decay constant, 2.1×10^{-6} s^{-1}, D – the radon diffusion coefficient in soil [$m^2 \cdot s^{-1}$].

Concentrations of gamma radionuclide radium ^{226}Ra in soil in Katowice and Kraków were determined by the authors several times with the use of gamma spectrometry systems. The measurements showed that the activity concentrations A_{Ra} in both regions were in a range from 20 ± 2 to 30 ± 2 $Bq \cdot kg^{-1}$. These values were in agreement with the data published in Radiation Atlas of Poland [2]. Studies performed by Mazur [17] showed that investigated regions are covered by silty-sandy kind of soil; thus for this type of soil, other necessary parameters were adopted. For soil grains from 0.6 to 1 mm, one can assume the value of the radon emanation coefficient in soil ε equal to 0.1–0.15 ([17] and references herein). Radon diffusion coefficient D varies in a range (4.8×10^{-6})–(8.5×10^{-6}) $m^2 \cdot s^{-1}$ [3, 17] and the average soil density ρ was assumed as 1.6×10^3 $kg \cdot m^{-3}$ [17]. Theoretically calculated E_{Rn} were in a range from 8.4 to 14 $mBq \cdot m^{-2} \cdot s^{-1}$ for $\varepsilon = 0.1$ and from 12.5 to 20.7 $mBq \cdot m^{-2} \cdot s^{-1}$ for $\varepsilon = 0.15$. The results of this order of magnitude were expected in the studied regions, although the values in a range from 0.2 to 50 $mBq \cdot m^{-2} \cdot s^{-1}$ were obtained for soils in the southern Poland [17, 19] and from 2×10^{-3} to 5×10^4 $mBq \cdot m^{-2} \cdot s^{-1}$ for soils worldwide [3].

Exhalation rate studies concern soil and rocks *in situ* measurements and building materials measurements performed in laboratories. For the latter, there are theoretical publications examining radon diffusion length and samples geometries (volumes, masses, thickness of layers, etc.) in several materials [3] and possible sources of uncertainties [20]. During *in situ* measurements, other factors play a role, such as meteorological conditions (air temperature, air pressure) [4, 17] and a precise and repeatable mounting of the accumulation chamber on studied soil during measurements.

The authors performed measurements during days with ensured stable weather conditions. The aim was to test accumulation chambers of different constructions and study whether chamber's parameters may influence the result of radon exhalation measurement. Five chambers were examined. Four of them had different values of V/S ratio V/S equal to 0.11, 0.12, 0.129 and 0.15 m (Fig. 2). Two of them (V/S = 0.11 and 0.129 m) had the partition mounted inside. The aim of the partition was to avoid mixing of gasses exhaling from ground and forced into the chamber from a close loop. For the same reason, two chambers had the outer valves mounted on side walls (not on the top) of the containers. Figure 4 presents the results obtained for measurements performed in the 'study field' of the Institute of Physics (Katowice) in May 2015. Concerning the radon exhalation rate E_{Rn} values for soils in the southern Poland (0.2–50 $mBq \cdot m^{-2} \cdot s^{-1}$ [20]), the obtained values in a range from 2.4 ± 0.4 to 12.9 ± 1.8 $mBq \cdot m^{-2} \cdot s^{-1}$ were all in low limits. No correlations with construction parameters or with increasing values of V/S were visible.

The second experiments were performed at the Institute of Nuclear Physics PAN in Kraków during nice weather days in July and September 2015. The authors used two chambers with V/S equal to 0.129 m, with and without the internal partition (Fig. 2b). Figure 5 shows the graph with exhalation rate values obtained during consecutive days. The values scattered from 6.2 ± 2.9 to 27.5 ± 8.7 $mBq \cdot m^{-2} \cdot s^{-1}$ with no correlation with a different construction of the chamber.

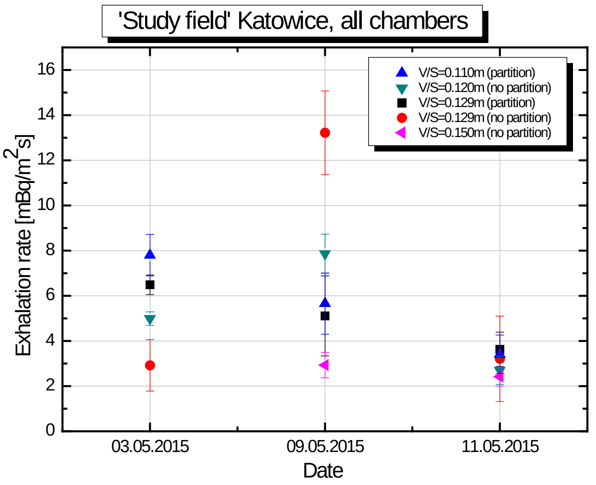

Fig. 4. The exhalation rate values obtained for measurements with different types of accumulation chambers performed at the 'study field' of the Institute of Physics, Katowice in the following days: the 3rd of May 2015, the 9th of May 2015 and the 11th of May 2015.

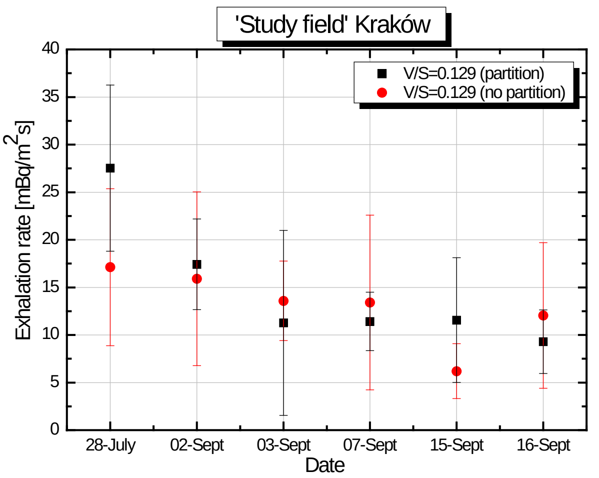

Fig. 5. The exhalation rate values obtained for measurements with the use of two accumulation chambers (with and without a partition, V/S = 0.129) performed at the Institute of Nuclear Physics PAN in Krakow in July and September 2015.

Conclusions

Radon exhalation from ground is a process dependent on the emanation and migration of radon through ambient air. The true and precise measurement is not easy because it must be performed in natural environmental conditions where a lot of factors such as air temperature, air pressure or wind power changes may play a role. The measurement of the exhalation rate from building materials performed in the laboratory is simpler in that sense.

In this publication, the authors raised a question whether the construction and the geometry of the chamber may influence the result of the measurement. As it was mentioned earlier, accumulation chambers available on the market or constructed by researchers always have different geometry and construction, being the engineering compromise amongst the cost of the device, the strength and the measured signal. Our experiments were performed at two testing grounds with similar characteristics for which the theoretically calculated radon exhalation rate was in a range from 8.4 to 20.7 $mBq \cdot m^{-2} \cdot s^{-1}$. The results obtained from measurements were of the order of magnitude of the calculated values allowing to conclude that chosen measurement method is generally correct.

The first tests performed in field conditions did not show the correlations of exhalation rate values and the chambers construction parameters and their geometrical dimensions. The measured values of radon exhalation rate varied from 2.4 ± 0.4 to 12.9 ± 1.8 $mBq \cdot m^{-2} \cdot s^{-1}$ in the 'study field' in Katowice and from 6.2 ± 2.9 to 27.5 ± 8.7 $mBq \cdot m^{-2} \cdot s^{-1}$ in the 'study field' in Kraków. The results were consistent within the errors range, which means that, as expected, the idea of the measurement method has been confirmed. The variability of V/S ratios was not very wide (V/S from 0.11 to 0.15) and this also could be the reason for such results. However, the process of radon exhalation from soil is complex and depends, amongst others, on meteorological factors. It seems that there are still too many factors that might have affected the results. Although the experiments were made within a few hours and during nearly the same meteorological conditions, we cannot exclude some impact to the obtained results, e.g. soil humidity and wind, which were not registered. Thus, the next measurements are planned to be performed in laboratory conditions without the influence of temperature/pressure/wind changing conditions. Apart from that, new accumulation chambers will be constructed with larger V/S ratios differences. This hopefully will make possible to compare the results only in terms of 'chambers' construction excluding or confirming the idea that if the chamber volume is large in comparison with the covered area, this may cause the underestimated registration of radon accumulated in the chamber and, in turn, the improper result of radon exhalation rate.

Trying to avoid disturbing factors the following future experiments are planned:

– the laboratory experiments with the well-known radon concentration value and without the influence of temperature/pressure changing conditions are being prepared,
– new chambers with higher V/S ratio differences are constructed.

References

1. Nazaroff, W. W., Nero, A. V. (Eds.). (1988). *Radon and its decay products in indoor air.* New York: Wiley Interscience.
2. Isajenko, K., Piotrowska, B., Fujak, M., & Kardaś, M. (Eds.). (2012). *Radiation atlas of Poland 2011*. (Biblioteka Monitoringu Środowiska). Warszawa: Centralne Laboratorium Ochrony Środowiska (in Polish).
3. Hassan, M., Hosoda, M., Ishikawa, T., Sorimachi, A., Sahoo, S. K., Tokonami, S., & Fukushi, M. (2009). Radon migration process and its influence factors; Review. *Jpn. J. Health Phys*., *44*, 218–231.
4. Kojima, H., & Nagano, K. (1999). The influence of meteorological and soil parameters on radon exhalation. In Proceedings of the International Conference "Radon in the living environment", 19–23 April 1999, Athens, Greece.
5. Sroor, A., El-Bahi, S. M., Ahmed, F., & Abdel-Haleem, A. S. (2001). Natural radioactivity and radon exhalation rate of soil in southern Egypt. *Appl. Radiat. Isot*., *55*, 873–879.
6. Saad, A. F., Abdallah, R. M., & Hussein, N. A. (2013). Radon exhalation from Libyan soil samples measured with the SSNTD technique. *Appl. Radiat. Isot.*, *72*, 163–168.
7. Singh, H., Singh, J., Singh, S., & Bajwa, B. S. (2008). Radon exhalation rate and uranium estimation study of some soil and rock samples from Tusham ring complex, India using SSNTD technique. *Radiat. Meas*., *43*(Suppl. 1), S459–S462.
8. Faheem, M., & Matiullah. (2008). Radon exhalation and its dependence on moisture content from samples of soil and building materials. *Radiat. Meas*., *43*, 1458–1462.
9. Shafi-ur-Rehman, Matiullah, Shakeel-ur-Rehman, & Rahman, S. (2006). Studying ^{222}Rn exhalation rate from soil and sand samples using CR-39 detector. *Radiat. Meas*., *41*(6), 708–713.
10. Shweikani, R., & Hushari, M. (2005). The correlations between radon in soil gas and its exhalation and concentration in air in the southern part of Syria. *Radiat. Meas*., *40*, 699–703.
11. Dueñas, C., Fernández, M. C., Carretero, J., Liger, E., & Pérez, M. (1997). Release of ^{222}Rn from some soils. *Ann. Geophys*., *15*, 124–133.
12. Dueñas, C., Liger, E., Canete, S., Perez, M., & Bolivar, J. P. (2007). Exhalation of ^{222}Rn from phosphogypsum piles located at the Southwest of Spain. *J. Environ. Radioact*., *95*(2/3), 63–74.
13. Oberstedt, S. H., & Vanmarcke, A. (1996). A radon exhalation monitor. *Radiat. Prot. Dosim*., *63*(1), 69–72.
14. Iimoto, T., Akasaka, Y., Koike, Y., & Kosako, T. (2008). Development of a technique for the measurement of the radon exhalation rate using an activated charcoal collector. *J. Environ. Radioact.*, *99*, 587–595.
15. Mazur, J., & Kozak, K. (2014). Complementary system for long term measurements of radon exhalation rate from soil. *Rev. Sci. Instrum*., *85*, 022104-1-7. DOI: 10.1063/1.4865156.
16. Ferry, C., Beneito, A., Richon, P., & Robe, M. -C. (2001). An automatic device for measuring the effect of meteorological factors on radon-222 flux from soils in the long term. *Radiat. Prot. Dosim*., *93*(3), 271–274.
17. Mazur, J. (2008). *Dynamika procesu ekshalacji radonu z gruntu a parametry meteorologiczne i własności gleby*. Ph.D. thesis, Instytut Fizyki Jądrowej PAN, Kraków, Poland.
18. ISO. (2012). ISO Standard: Measurement of radioactivity in the environment – Air: radon-222 –Part 7: Accumulation method for estimating surface exhalation rate. ISO 11665-7:2012(E).
19. Wysocka, M. (2011). *Wpływ górnictwa na migrację radonu w środowisku geologicznym*. Habilitation thesis, GIG, Katowice.
20. Samuelsson, C., & Pettersson, H. (1984). Exhalation of ^{222}Rn from porous materials. *Radiat. Prot. Dosim*., *7*, 95–100.