

ORIGINAL ARTICLE

Measurement of radon concentration in soil gas of the city of Rivne (Ukraine)

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This work presents the results of determining the content of radioactive gas Radon-222 in a soil gas. The measurement was carried out in 48 points which are deployed uniformly on the territory of Rivne city. The city is situated within boundaries of Volyn-Podillya plate at the western slope of the Ukrainian crystalline massif. This region is characterized by the considerable content of uranium in granites. Studies were conducted by the express method with the help of a radonimeter "Alpharad PLUS" from June to October 2018. It was established that the radon concentration in a soil gas was low in correspondence with criteria determined by Akerman et al., 1984. The radon content at the depth of 1 m varies within boundaries of 1.4-17.6 kBk/cub.m., with the arithmetic mean value of 11.13 kBk/cub.m., the root-mean square deviation-4.8 kBk/cub.m. It is determined that the radon concentration in a soil gas correlates with the Radium-226 content in a soil; the possibility of transporting radon with a soil gas is established by the soil permeability and climatic conditions - temperature, moisture, pressure, wind velocity; a soil gas which contains radon passes into buildings via cracks or apertures in the foundation due to much lower pressure of air in premises in comparison with the pressure outside buildings; the radon content in a soil gas usually is sufficient to support in the inner building air the radon concentration higher than that established for Ukraine level of action (100 Bk/cub.m.); radon in a soil gas is produced both by upper soil layers, and as a result of its transportation from depths along the system of fractures. It is possible to state that there exists a certain correlation between radon levels in a soil gas, inner building air and death rate from lung cancer of the population in different districts of Rivne city. Therefore, it is expedient to continue the research into the regularities of the distribution of soil radon not only in Rivne city and its suburbs, but also in the whole region with the aim of protecting the population from irradiation on radon dangerous territories and plots.

Keywords: Radon; soil gas; volumetric activity; concentration; risks for population

Introduction

It is known that radon and associated products of its decomposition (APD) in the air of premises stipulate the increased risk of lung cancer diseases for the total population because of its entering human being organs from air during inhaling and inner irradiation of lungs (Jacobi, 1996; Peterson et al., 2013; Truta et al., 2014). Besides, the combined action of radon on organism, its APD and a number of factors of non-radiation nature (dust, exhaust gases of engines, products of tobacco combustion) strengthens unfavourable effects stipulated by these factors:

1. the development is speeded up of silicotonic process in lungs;
2. with greater probability emerge tumours of bronchi even in those beings for whom the probability of appearing cancer illnesses under natural conditions is very small;
3. simultaneously with tobacco smoke oncological effect of radon influence and its APD increase by 2-10 times and what is important diminishes the latent period of lung cancer development (with miners who smoke during 3-12 years) (Sevalnov et al., 2009).

The detailed research into causes of lung cancer formation showed:

1. in bronchi of human beings there exist "cells of risk" which receive the highest dose during the impact of a-particles which are emitted by radon and its APD;
2. delayed effects of irradiation of respiratory organs by radon and its APD are determined, first of all, by a summary dose irrespective of the time during which it was accumulated;

3. because of the unevenness of the distribution of radionuclides in lungs it is possible to have the massive irradiation of separate segments of bronchi-lung system and the development of lung cancer in them under mean absorbed doses on lungs approaching maximum admissible ones for professionals (Sevalnov et al., 2009).

Soil is a basic source of radon admission into buildings. It forms 90% of radon exposition in a dwelling (Nazaroff, 1988). The concentration of Rn-222 as a basic component of radon in atmospheric air and in premises depends on the content of U-238 and Ra-226 in mountain rocks, soils and underground waters, the presence of zones and areas with uranium ores mineralization, the presence of fissures in mountain rocks, weathering, the emanation coefficient Rn-222 from soil, on properties and structure of soil (Table 1). High uranium concentrations are characterized for the magmatic eruptions of rocks, in particular granites, sedimentary rocks which contain phosphates and for metamorphic rocks created from such deposits. It is by such parameters that is characterized the Ukrainian crystalline shield and Volyn-Podillya plate on which Rivne city is located.

Table 1. Exhalation of radon from various underlying surfaces.

Types of emanating surfaces	Values of density of radon flow, Bk	$\cdot \text{m}^{-2} \text{second}^{-1}$
Zones containing uranium secondary	3.7-11.1	
Different granite types	(1.85-11.1)	$\cdot 10$
Present-day deposits (soils, sands, clay)	(1.85-11.1)	$\cdot 10$
Water surfaces	less 3.7	$\cdot 10$

A mean volumetric activity (VA) of radon in Earth soil at the depth of 10 cm is assessed by 136-158 kBk/cub.m. (Abdulayeva, 2013). However, radon in Earth entrails is spread very unevenly. This is connected with the fact that it is accumulated in tectonic disturbances where it admits by a system of micro-fissures from mountain rocks. In practice of geological studies many cases were registered when low-radioactive rocks contain in their voids and fractures radon in quantities by hundreds and thousands larger than in more radioactive rocks.

In water saturated rocks which lie below the level of ground waters the migration dominates of radon with the flow of underground waters in dissolved form, therefore, rocks which occur below the level of ground waters may be called as the zone of water radon migration. Within the boundaries of incomplete water supply (from land surface to the level of ground waters) dominates the migration of radon in a gaseous phase as a component of soil gas and therefore such zone may be called a zone of air radon migration. Radon migration in these two zones takes place, more often, absolutely independently. In the zone of water migration dominates the horizontal transfer of radon while in the region of air migration radon transfer takes place mostly in a vertical direction.

As a result of vertical migration of radon in soil there happens its emission into atmosphere. The speed of radon entrance from soils into atmosphere depends on physic-geometrical properties of soils, the hour of day and night, seasonal temperature variations, pressure and moisture (Grodzinskiy, 1987). The decreased atmospheric pressure causes the increased content of radon in over-soil air; the increase-diminishes this concentration. There exist contradictory observations of seasonal variations of the level of radon in soil, in some-higher concentration was observed in winter or summer (Schumann & Gundersen, 1996; Hutter, 1996; King, 1994), in others were approximately the same during a year (Fleischer et al., 1980).

Along with this, with increased humidity is increased the degree of filling ground pores by water (moisture saturation) and, correspondingly, is decreased the radon exhalation of radon at the expense of dissolving its atoms in water. With the saturation over 70% the air movement in pore channels of soils is practically absent. The admission of air into atmosphere in this case is possible only at the expense of the diffusion from the surface of weak permeable blocks. Under non-substantial decreased humidity micro pores are drained, and gas penetration is substantially increased. Maximum values are obtained under the filling degree of pores by water less than 15-20% (Miklyayev & Ziangirov, 2001).

On the basis of researching the levels of radon concentration in a soil gas a subsequent classification was proposed of volumetric activity (VA) in soil: low (<25 kBk/cub.m. (mean (25-100 kBk/cub.m.), increased (100-250 kBk/cub.m.) and high (>250 kBk/cub.m.) (Akerblom et al., 1984; Kemski et al., 1998).

The investigation of the relationship between measured radon concentration in premises and the gas in soil under it showed that between them is revealed a very weak dependence (Albering et al., 1996; Varley and Flowers, 1998). Naturally, if radon concentration in soil is "high", that is, >250 kBk/cub.m., the admissible level of radon action in premises is 250 kBk/cub.m. (Germany), was exceeded in all cases (Kemski et al., 1998). On another hand, it is informed that radon concentration in soil to 10 kBk/cub.m. may cause the concentration of this gas in premises higher the admissible level, in particular, in Great Britain this is 200 kBk/cub.m. (Varley & Flowers, 1998).

Our preliminary studies showed that in dwelling premises in Rivne were observed significant variations in radon concentration, including the fact that there occurred such ones which greatly exceed the established levels for Ukraine (Klymenko & Lebed, 2017).

Thus, considering all above said, the task of our researches will be the determination of statistic characteristics of the distribution of radon VA in a soil gas in different districts of Rivne city.

Object and methods of research

Geological parameters of the domain studied. Rivne city is located within the boundaries of Volyn-Podillya plate on Rivne forest plateau which by depth fractures separates Male and Volyn Polissya (Klymenko et al., 2014). Into this plate the Ustya river is cut which divides it in submeridian direction into two parts with flood land and over-flood land terraces. The geological structure on the city territory is represented by Proterozoic, Mesozoic and Cainozoic deposits. This Volyn-Podillya plate is a western slope of the Ukrainian crystalline shield destroyed by a complicated system of fractures. In its turn this slope falls by steep fractures-steps (blocks) to Halyxh-Volyn depressions. The mountain systems of the Ukrainian crystalline shield which were formed before Cambria period, during all the Phanerosis remained a dry valley, and territories of Volyn-Podillya plate adjoining them were periodically flooded by the sea during transgressions.

All territory of Rivne city is covered by a significant thickness of upper chalk rocks of Senoman, Turon, Santon layers. The most spread on the territory of the city are deposits of Turon layer which are represented by white writing chalk, greenlike-grey marls and chalk-like limestone with ingredients of silicon of total capacity of 20-45 m. Paleogenic deposits are spread in the southern and western environs of the city and are represented, in the main, by formations of Kyiv layer from grey and blue-like alevrites and green-like quartz-glaucconitic sands. Neogenic deposits in the south-west and east of the city are represented by deposits of Sarmate layer with the capacity of up to 30 m. They are sands, limestone and clays. The specifics of lithologic composition of upper-quatarnary deposits for Rivne city consists in the substantial spread of loess complex of beech layer. Deposits are represented by micro-porous, carbonate loesses and similar to loesses loams of pale-yellow-gray colour with the capacity of 6-10 m. which are underlaid by grey gley fossilized soils.

The bases of present-day relief are upper-chalk deposits. The relief of the surface is wavelike-hilly, hills are with flat tops, their slopes are flat and gradually transform into borders of valleys and gullies. Steep slopes are characterized by precipice plots, landslides.

Main soil forming in Rivne city are loesses. Soils of the Ustya river terrace developed on alluvial-diluvia and pluvio-glacial deposits. Therefore, there exists a distinct connection between the deployment of certain types of soils with relief and geological specifics of the city. Within the boundaries of watershed plots soils are represented by black soils usually carbonate with the low content of humus, black soils, meadow, mean loam, gley, dark-gray degraded turf-carbonate soils. The territory of the city is characterized by the diversity of deployment of soil varieties; they have island localization which is stipulated by crosscut, variegated relief of the city. This influences the nature of washed-out, air and water regimes, and hence, the intensiveness of radon exits from soil.

The right bank of the Ustya river is characterised by loess soils-grey and black soils of podzolic type, on the left bank developed leached black soils. The soils of the flooded part of the territory in the city are less strong and less variegated and represented in the southern part by sludge-swamp, and in the northern - by turf-swamp ones. This is explained by relatively uniform conditions of their formation. During the long history of the city they transformed many times, were artificially filled, drained. As researches testify in some places of Rivne natural soils of flooded land and outside the flooded land are covered by a layer of artificial filled soils with the capacity of up to 4.5 m. City soils have also a strong cultural layer which contains a substantial part of building and household rubbish. Soils of artificial street plants and grass lawns contain less humus. The most hardness, density, filtration capability have massifs of new buildings (saturated by building rubbish) and places of mass concentration of population. In such places the hardness of soil varies within boundaries of 35-44 kg/cub.cm. and more, the density- 1.1-1.9 g/cub.cm.

Surface waters of city territory are represented by the Ustya river, water reservoirs and ponds in its valley. Hydrological the city territory is within the boundary of Volyn-Podillya artesian basin. Climatic conditions of the territory are characterized by substantial quantity of atmospheric precipitation, moderate temperatures and increased air humidity which under conditions of good permeability of covered deposits stipulates the permanent replenishment of underground waters reserves forming zones of active water exchange to a significant depth (up to 100 m.).The absence of held in plan and in the cross section of water firm horizons conditions the close hydraulic connection between water carrying horizons.

Measurement of radon in soil gas. The majority of experiments on measuring radon concentrations in premises air, water, soil gas is based on using integral methods of measurement on the basis of solid body nuclear trek detectors (SSNTD on the basis of films LR-115) (Tabar et al., 2013) or on using "instantaneous" measurement methods - ionization (Vaupotic et al., 2010; Kunovska et al., 2012; Everlise G. Lara et al., 2011), electrostatic (Lebed & Myslinchuk et al., 2018; Lebed & Klymenko et al., 2018) or aspiration (Winkler et al., 2001). Integral ways measure, integrate and average the value of radon VA during the period from several weeks to a year. Films LR-115 have some positive characteristic:

1. very sensitive to alpha-particles;
2. may be used for a short-term measurement of during minimum 10-30 days of exposition, and also for long-term measurement, from 3 months to a year;
3. not sensitive to changing parameters of environment such as humidity, availability of water in the zone of measurement and values of soil temperature to 60 °C;
4. suit to measure radon in stagnant soil zones or water (Dwaikat et al., 2007).

However, the measurement of Radon concentration in a soil gas with the help of solid body nuclear trek detectors has one substantial shortcoming which is seldom paid attention to. Along with the determination of Radon-222 concentration in soil air they register treks of Radon-222 (Toron) which influence substantially the value of summary radon concentration in soil. As Toron has very small period of half-decay (55.6 s.) it practically does not enter the premises from soil and does not influence the radon concentration in it. This shortcoming is absent in measurements on the basis of "instantaneous" methods.

Our measurements of radon VA in soil air were based on electro-static method. The time choice was based on maximum dry period (June-September 2018), when soil pores by our observations were maximum open. The selection of samples of soil air was carried out from a drilled blast-hole in soil by a special screw into a sample collector with the subsequent determination of radon VA in a sample collector by radonometer "Alpharad PLUS" (Figure 1) by way of mixing the sample between air volumes of a sample collector and a measuring chamber with the subsequent measurement of radon VA in a measuring chamber of a block measuring VA. In researched points of soil blast-holes were drilled of 3-5 cm diameter and depth of 70-100 cm into which were lowered sample collectors by a rope. The orifices of blast-holes were dusted with soil. The time of the sample collector exposition in a blast-hole necessary for measuring VA of a sample collector and a blast-hole accounted to not less than 12 hours. The time between lifting the sample collector from a blast-hole and the air isolation in it amounted not more than 1 min. The time from the air isolation in a sample collector and its joining to a radonometer amounted to not less than 1 hour for a guaranteed decomposition of radon in a sample. The time between lifting the sample from a blast-hole and the measurement of radon VA by a device was taken into account during the calculations for considering Radon-222 decomposing.

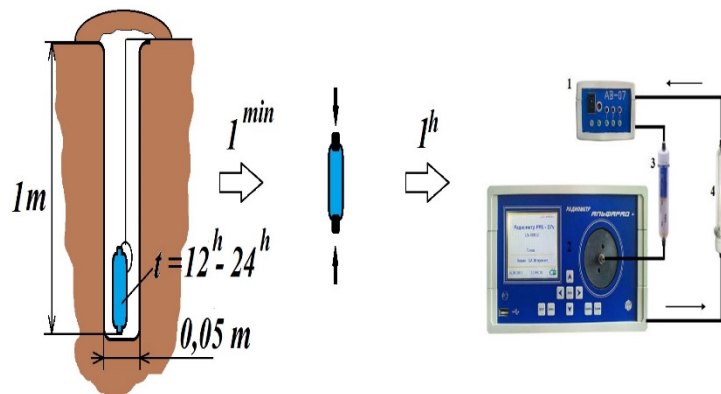


Figure 1. Methods of measuring radon in soil gas: 1-autonomous air blow; 2-measurement block of VA; 3-dryer-cartridge; 4-sample collector of soil air.

The principle of determining Radon-222 VA is based on electro-static sedimentation of charged ions Polonium-218 from the selected sample of air to the surface of a-detector (semi-conductor detector). The nucleus of Radon-222 which is decomposed inside the chamber leaves the product of its decomposition nucleus Polonium-218 as a positively charged ion. The electric field inside the chamber drives into movement this positively charged ion in the direction of the detector to which it is electrostatically drawn. VA of Rn-222 is determined by the quantity of registered α -particles during the decomposition of Polonium-218 atoms which stayed on the detector.

Results and discussion

Measurements were carried out in 48 points of Rivne city from June to October 2018 at the depth of 1 m. (Figure 2). In the researched period the mean temperature of air was about 25 °C, and precipitations were insignificant. Studies give the reason to consider that even Radon concentrations in a soil gas for this locality are low 1.4-17.7 kBk/cub.m. with the arithmetic mean of 11.13 kBk/cub.m. (Table 2), but sufficient for creating radon VA for buildings at the level of up to 1420 Bk/cub.m. registered in investigations in 2017 (Klymenko & Lebed, 2017). Obtained values are comparisons with Radon VA in soils of countries represented in Table 3. The preliminary careful conclusions say that, there exists a certain correlation between radon levels in a soil gas, inner building air and death rate from lung cancer of the population in Rivne city. The juxtaposition of these data shows that, for example, during the period of 2014-2016 no inhabitant of Basiv Kut district was registered sick for lung cancer. It is here that there were registered the least values of radon VA in building air and the lowest values of the concentrations of soil radon (1412 Bk/cub.m.). Not very big values of the concentration of soil radon were observed in point's stated along the Ustyia river. Buildings in the river embankment have also not great values of VA for inner radon, and the population residing in them have low indices of diseases for lung cancer. At the same time for points near Grushevskiy str., where the number of dead from lung cancer during the same period amounted to 42 people, are observed the highest in Rivne values of radon VA in buildings and high values of the concentration of soil Radon.

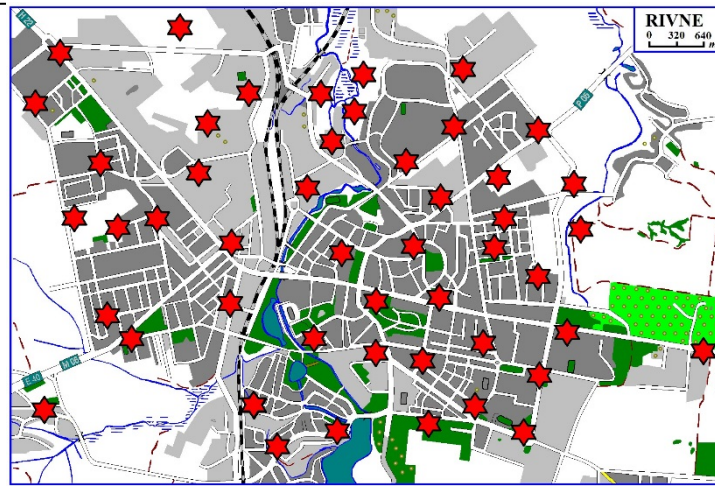


Figure 2. Points (sites) of sampling of soil gas in Rivne city.

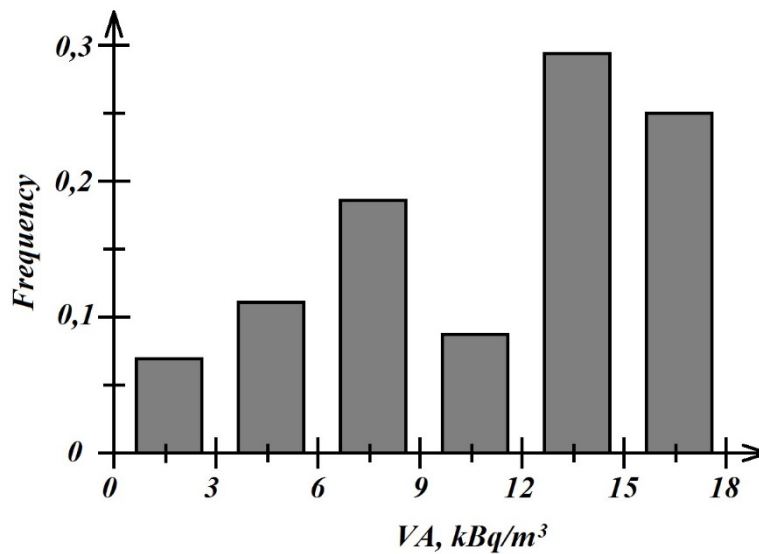


Figure 3. Frequency of distribution of radon concentration in soil gas in Rivne city.

Table 2. Statistic characteristics of measuring concentrations of Radon-222 in soil gas in Rivne city.

Type of samples	Number of samples	Arithmetic mean	Geometric mean	Minimum	Maximum	Mean square deviation	Median	Standing arithmetic deviation	Standard geometric deviation
Soil gas, kBk/cub. m.	48	11.130 ± 0.693	9.731	1.41 2	17.638	4.803	12.346	0.693	1.806

Table 3. Comparison of Rn-222 activity in soils of some localities of different countries.

#	VA, Bk/cub.m.	Country	References
1	1657-1855	India	Chaudhuri H. et al., 2010
2	1500-15900	India	Choubey V. M. et al., 2007
3	800-26700	Jordan	Abumurad K. M. et al., 2005
4	1700-24000	Russia	Iakovleva V.S. et al., 2003
5	9910-42100	Ghana	Amponsah P. et al., 2008
6	5500-8700	Cameroon	Ngachin M. et al., 2008
7	6800-74700	Canada	Chen J. et al., 2008
8	209-7389	Turkey	Erees et al., 2006
9	4300-9800	Turkey	Inceoz et al., 2006
10	98-8594	Turkey	Tabar et al., 2013
11	4000-97000	Bulgaria	Kunovska et al., 2012
12	7000-93000	Brazil	Lara E.G. et al., 2011
13	900-32900	Slovenia	Vaupotič et al., 2010

14	17340-72520	Pakistan (Islamabad)	Ali et al., 2010
15	690-3890	Pakistan (Murri)	Ali et al., 2010
16	1400-17600	Rivne city (Ukraine)	Our studies

Figure 3 shows the frequency distribution of radon concentration in a soil gas in Rivne city based on 48 measured points. Along with this, obtained in general low values of radon concentration in a soil gas of Rivne do not agree with results of our preceding measurements of radon volumetric activity inner building air. By our data (Lebed et al., 2018) in 5.5% of city buildings the concentration of inner radon is over 400 Bk/cub.m. and the exposition which a human being gets during all life is near the level which miners uranium mines obtain, though a share of population living there does not exceed 10% of all inhabitants of the town. It is evident that it is necessary to conduct larger number of measurements with less step between experimental blast-holes because, perhaps, we did not manage in most measurements to find the system of fractures along which radon enters into the near-surface layer of soil.

Conclusion

Results of our studies show that the content of Radon-222 in a soil gas correlates with the content of Radium-226 in soil; the possibility of transporting radon with soil gas is determined by the permeability of soil and climatic conditions-temperature, humidity, pressure, wind velocity also; a soil gas containing Radon enters buildings via cracks or apertures in the foundation through lower air pressure in premises in comparison with the pressure outside buildings; the content of Radon in a soil gas usually is sufficient to support in inner building air the concentration of radon higher the established in Ukraine level of action (100 Bk/cub.m.); Radon in a soil gas is produced both in upper soil layers, and as a result of its transportation from depths by a system of fractures.

The levels of Radon concentrations in a soil gas for Rivne city are low by the classification of Akerblom and amount to 1.4-17.7 kBk/cub.m. with arithmetic mean of 11.13 kBk/cub.m., standing deviation of 4.803 kBk/cub.m., and mean geometric of 9.731 kBk/cub.m. Hence, it is possible to state that a certain correlation exists between Radon levels in a soil gas in inner building air and death rate from lung cancer for the population in different districts of Rivne city.

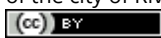
Actual is the problem of determining regularities of the soil Radon distribution not only in Rivne city and its outskirts but also on the whole territory of the region with the aim of prophylaxis and protection of population from irradiation on Radon dangerous territories and plots.

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