



# Measurement of Exhalation of Radon-222 from Soil of City of Rivne, Ukraine

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**Abstract:** These studies show that the radon content in the air of buildings of Rivne city is mainly influenced by the factors radiological characteristics of the underlying soils under the buildings and building materials (first of all, the radon emanation coefficient in soil, soils gas permeability, etc.), characteristics of the ventilation systems of buildings and modes of ventilation, design features of buildings (number of floors, type of foundation, available aerodynamic connection between floors, location of communication systems within outlines of building, etc.), the type of glazing (the degree of tightness, available ventilation valves, etc.), the type of heating (stove, electric, gas or central) and the level of floor in spaces: basements, semi-basements, first floors regarding land surface. Considering the given factors, can conclude that essential reduction in radon content in indoor air of buildings can be achieved by) choosing a site for building, where radon release from soil is low, applying building envelopes that effectively prevent entering of radon from the soil into building, removing of radon from indoor air. In order to comply with these requirements it is necessary to monitor radiation safety parameters according to volumetric activity in indoor air and radon flux density (RDF) from soil under buildings. The results thereof should be used for sanitary and epidemiological assessment of land sites for building and environmental safety in living spaces. We consider that the main parameter of the anti-radon pre-design stage of construction is the determination of RDF from soil in building site and mapping of RDF from soil in modern cities will allow to develop plans for ecological building and ensure reduction of the ecological radon risk for the citizens. Mapping is based on the analysis of the spatial distribution of values of radon flux density, which is released from surface of city soils. Such approach allows to use the map in urban planning, in pre-design surveys for building, in hygiene studies, for estimating of the doses that are received by citizens due to radon exposure and, finally, as a resource for informing the citizens.

**Keywords:** Radon, Exhalation, Radon flux density, Lung cancer

Recently, in connection with the adoption of a number of documents by world organizations, wherein new and more severe provisions regarding radiation contamination of the inhabited territories in Europe and the world, which are prescribed in the Council Directive 2013/59 / EURATOM (Council of the European Union 2014) and WHO (Zeeb and Shannoun 2009), are generated. The study of radon fields of cities with a large cluster of people located in platform areas that are characterized by a calm geodynamic environment is the subject of interest. Currently, the national legislations of the countries of the world have a number of regulations that govern the radon concentration in residential and industrial spaces and the radon RFD from soil surface in the built-up area or the area that is planned to be built-up [DBN 1.4-2.01-97 (Ukrainian national construction regulation), 1997; SP 2.6.1.2612-10 2010 (Set of Rules, etc.)]. The limiting RFD rate is the value 80 mBq/(m<sup>2</sup>·c). Despite the fact that studies of radon fields from geo-ecological points have been actively carried out by both Ukrainian and foreign experts in recent decades, the unified theoretical and methodological base of these studies has been developed insufficiently. This is partly connected with divergence of views among researchers

regarding formation mechanisms of the radon field of soil massifs (Mykliav 2015). According to the general opinion of researchers, its transfer in rocks, soils, and the further exhalation from the surface are complex processes subject to the influence of numerous natural factors. We note that <sup>222</sup>Rn (formed during the decay of <sup>226</sup>Ra) rises from the depth of rocks to soil surface, which is facilitated (or obstructed) by diffusion due to gradient of radon concentration; effusion due to pressure gradient in the rock mass; thermal, liquid and gas convection arising upon availability of geothermal gradient; gaseous cavitation and raising of geogas bubbles in pores filled with water; filtration and infiltration of water flows; turbulent effects in the soil air upon changes of external meteorological conditions; fluctuations of the hydro-geode formation field of the Earth; seismic activity, etc.

All these processes, except for molecular diffusion, are difficult to differentiate and quantify (Ivanova 1999). Amidst such uncertainty in the range of impact of the above factors on the mechanisms of formation of radon fields in soil, atmosphere, spaces, there is a discussion regarding parameters that characterize radon field and they are to be measured. Some scientists believe that basic information

about causative bodies in geological structures is carried by volumetric activity (VA) in the soil (subsoil) and / or atmospheric air, others consider that only RFD through the surface can provide necessary and reliable information about radon sources in deep structures that allow radon to pass through and generate radon anomalies. The purpose of this study - mapping of density of Radon-222 flux from soil of Rivne city (Ukraine). Object of study: indicators of density of Radon-222 flux from soil of Rivne city. Subject of research: samples of air that comes due to exhalation of Radon-222 from the soil of Rivne city.

## MATERIAL AND METHODS

**Geology of Rivne city:** Rivne is located within the Volyn-Podilia plate in the Rivne loess plateau, which divides Male and Volyn Polessia with deep faults (Klymenko et al 2014). Yustia river runs into this plate that divides the river in submeridional direction into two parts with the floodplain and above floodplain terraces. The geological structure in the city is introduced by Proterozoic, Paleozoic, Mesozoic and Cenozoic sediments (Herenchuk 1976, Zaleskyi 2005, Klymenko et al 2001). This Volyn-Podilia plate is the western slope of the Ukrainian crystalline shield (rich in uranium ore deposits), divided into a complex system of faults. Almost the entire territory of the Rivne region, including Rivne city, is covered with a significant layer of Upper Cretaceous rocks of the Cenomanian, Turonian, and Santonian layers. Deposits of the Turonian age are the most wide-spread on the territory of the city. They are introduced by white writing chalk, greenish-gray marls and chalk-like lime stone with flint of depth from 20 to 45 m. The Neogene deposits in the southwest and east of the city are introduced by deposits of the Sarmatian stage with depth of 30 m. These are sands, limestone and clay.

**Soil-forming rock of Rivne city:** Right bank of Yustia river is characterized by loess soils - gray and podzolized chernozem, leached chernozems were developed on the left bank. The soils of the floodplain area of the city are not so deep and not so diverse. The silty-marsh soils are located in the southern part and the peat-boggy soils are in the northern part. This is due to the relative sameness of the conditions of their creation. Over the long history of the city they were repeatedly transformed, artificially filled up and dried out. Studies of individual spots of Rivne city show that the natural soils of the floodplain and above floodplain terraces are covered with a layer of artificial filled up soils with depth of up to 4.5 m.

**Sampling points, methods and instruments of measurement:** The territory of Rivne city was divided into 48 tested sites. At each site the RFD was measured at three

points (7 times at each point with further averaging), total number of measurements is 144 in the spring-summer periods 2016-2017. The radon flux density from the soil was measured by the complex "Alfarad plus" to monitor radon, thoron and their daughter products. This complex can carry out rapid measurements and continuous monitoring of the RFD of Radon-222 from the soil surface (within  $(20 \cdot 10^3)$  mBq/(m<sup>2</sup>·c)), soil air (within  $(10^3 \cdot 10^6)$  Bq/m<sup>3</sup>), with a relative error  $\leq 30\%$  (Fig. 1).

The principle of determination of the VA of Radon-222 is based on electrostatic deposition of charged ions of <sup>218</sup>Po from the air sample taken onto the surface of the  $\alpha$ -detector (semiconductor detector). The core of <sup>222</sup>Rn, which decays inside the chamber, leaves the product of its decay, <sup>218</sup>Po core, as a positively charged ion. The electric field inside the chamber drives this positively charged ion in the direction of the detector, to which it is being electrostatically attracted. The flux density of <sup>222</sup>Rn is determined by the number of  $\alpha$ -particles detected during decaying of <sup>218</sup>Po atoms that deposited on the detector. After measuring is completed, the results are shown on the monitor. The results include date, measurement completion time, number of impulses, operating mode, value of radon RFD from soil, measurement error, pressure, temperature, humidity, and they are shown on the complex screen (Fig. 3).

## RESULTS AND DISCUSSION

The Table 1 and the histogram (Fig. 2) show the results of determination of the density of radon flux from the soil of Rivne city (Ukraine). The measured RFD values are within quite wide limits: from 16 mBq/(m<sup>2</sup>·s) (Fig. 3) to 173 mBq/(m<sup>2</sup>·s) (Fig. 4).

The obtained values of the RFD indicate a complex radon situation in the city, as proved by studies of radon VA in basements, semi-basement rooms and spaces of the first floors of residential buildings (Klymenko & Lebed, 2017, Lebed and others 2018). A significant range of RFD (from 16 to 173 mBq/(m<sup>2</sup>·s) is obviously determined by meteorological, climatic and biotic factors. The obtained statistics shows that radon fluxes from the city's soil are comparable to generally accepted radon-hazardous zones of the world, such as the Caucasus, Irkutsk Region, Tatarstan, Krasnoyarsk krai (Russia), Illinois (USA) (Table 1). The average continental value of radon exhalation in Europe is considered to be 21 mBq/(m<sup>2</sup>·c), which is 1 atom/(cm<sup>2</sup>·s), however, they vary considerably in space and time. Average values of the flux of <sup>222</sup>Rn from soil to atmosphere for different regions of the world are shown in Table 2.

The distribution of the frequency of falling of the measured RFD value within the appropriate range has

lognormal nature. The statistical characteristics were calculated according to the obtained values (Table 2).

Today, it can be argued that the radon flux density field of platform territories has a discrete spatial structure and it is divided into background and anomalous components

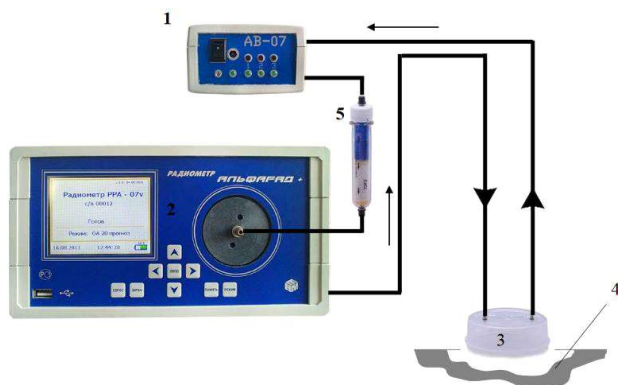


Fig. 1. Scheme of air sampling for density of radon flux from the soil (1. Self-contained blower; 2. RFD measurement unit; 3-Accumulation chamber; 4. Soil; 5. Dehydrator plug)

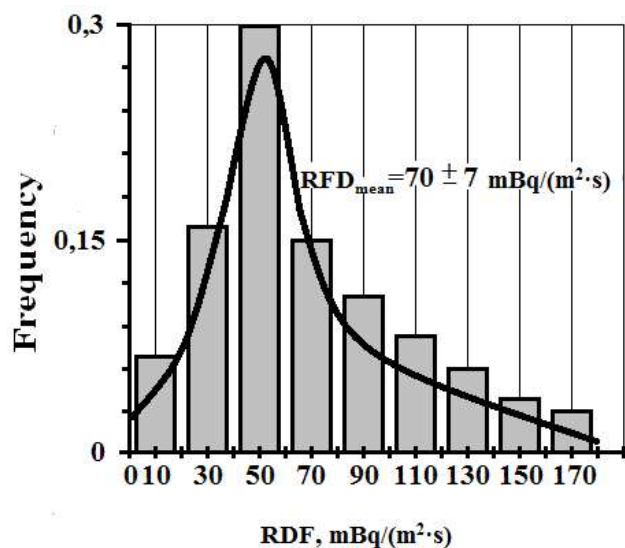


Fig. 2. Frequency of falling of density flux values (RFD) into corresponding range

(Mykhaiev 2001). These parameters are constantly changing in time (Rogalis et al 2001). Radon anomalies are associated with geodynamic active zones and connected with anomalous deformations of near-surface soils. The average continental value of radon exhalation in Europe is considered to be 21 mBq/(m<sup>2</sup>c), which is 1 atom/(cm<sup>2</sup>·s), however, they vary considerably in space and time. Average values of the flux of <sup>222</sup>Rn from soil to atmosphere for different regions of the world are shown in Table 3.



Fig. 3. Values of the meter when measuring RFD in soil on Vynnychenko Str.



Fig. 4. Values of the meter when measuring RFD in soil near "Avanhard" stadium

Table 1. Radon flux density (RFD, in) from soils of the city

RFD (mBq/(m <sup>2</sup> ·s))	0-20	20-40	40-60	60-80	80-100	100-120	120-140	140-160	160-180
Number of sampling spots	10	23	42	21	16	12	9	6	5
Repetition rate	0.07	0.16	0.30	0.15	0.11	0.08	0.06	0.04	0.03

Table 2. RFD statistics in mBq/(m<sup>2</sup>·s) from soils of Rive city

Type of determination	Number	Arithmetic mean	Geometrical mean	Standard deviation	Median	Max.	Min.
RFD from soil	144	707±7	60	41	62	173	16

The RFD parameter is more sensitive to changes in the stress-strain behavior of the geological environment than the VA value. It can be used as an independent or additional prognostic parameter. Simultaneous use of two values – VA of soil radon and RFD from the soil surface allows to increase reliability of predicted evaluation in dynamics of radon entry indoors (Korshunov et al 2012). The high concentrations of radon were recorded in clay soils and in very rotted and old

rocks. Low radon concentrations are observed in pure limestone and sand. The values of exhalation depending on the type and condition of the soil are shown in Table 4.

On average, in loose homogeneous sedimentary rocks the maximum volumetric concentration of soil radon is reached at a depth of about 5 m. As a result of the vertical migration of radon in soil, it is released into the atmosphere. For the model of expansion of gas - emanation in non-

**Table 3.** Average values of flux of  $^{222}\text{Rn}$  from soil to atmosphere for different regions of the world

Region	RFD of $^{222}\text{Rn}$ from soil, mBq/(m <sup>2</sup> ·c)	Source
Australia	22	Schery et al (1989)
Innsbruck (Austria)	9-16	Zeilingner (1935)
Nepal	39	
China	29.7±9.4	Zhuo et al (2018)
Seclin (France)	15	Servant (1964)
Aachen (Germany)	17	Israël (1970)
Heidelberg (Germany)	19	Dörr et al (1989)
Sakarya (Turkey)	0.2-1.4	Tabar et al (2018)
Osaka (Japan)	8-11	Megumi and Mamuro, (1972,1973), Tojo ( 1989)
New Zealand	4	Rosen 1957)
Gunma (Japan)	2-11	Prasad et al ( 2012)
Warsaw (Poland)	7	UNSCEAR (1982)
Socorro (New-Mexico, USA)	34	Wilkening and Hand (1960)
Uttar Pradesh (India)	0.1-0.3	Zubair et al (2012)
Yucca Flat (Nevada, USA)	21	Wilkening et al (1972)
Linkoln (Massachusetts, USA)	50	Wilkening et al (1972)
Chapman County (Illinois, USA)	53	Pearson and Jones (1966)
Argonne (Illinois, USA)	21	Pearson and Jones (1966)
Central Europe	7	(Milin et al (1968)
Kazan (Russia)	17-149	Apkin (2016)
Northern Russian Federation	38	Kirichenko (1970)
Caucasus	73	Kirichenko (1970)
Middle Asia	19	Kirichenko (1970)
Moscow (Russia):		
Clay soils;	38	Milin et a (1968)
Sandy soils;	21	Milin et al (1968)
Krasnoyarsk (Russia)	40	Berezina ( 2014)
Krasnoyarsk (Russia)	32	Kurguz (2002)
Irkutsk (Russia)	50-100	Berezina (2009)
Keiga (India)	3	Berezina (2014)
Malaga(Spain)	10-25	Berezina (2014)
Vilnius(Lithuania):		
Clay soils;	38	Berezina ( 2014)
Sandy soils	25	Berezina ( 2014)
Rivne (Ukraine)	70	Our studies

radioactive soil layer upon availability of convective transfer from emanation source, the solution of the radon transfer equation is as follows (Shuleikin 2010):

$$Q = Q_0 \cdot \exp\left(z \cdot \left[\frac{v}{2r} - \sqrt{\frac{v^2}{4r^2} + \frac{\lambda}{r}}\right]\right) \quad (1)$$

where  $r$  is the diffusion coefficient ( $\text{cm}^2/\text{s}$ ),  $Q$ ,  $Q_0$  is the amount of gas-emanation in one cubic centimeter of soil air at a depth of  $z$  and  $z = 0$ , respectively;  $\lambda$  is the radioactive constant ( $1/\text{s}$ );  $v$  is the radon transfer rate ( $\text{cm}/\text{s}$ , for sedimentary rocks  $v = 5 \cdot 10^{-4}$   $\text{cm}/\text{s}$ );  $z$  is the vertical coordinate, the beginning of which is located on the border of the emanation layer ( $\text{cm}$ ).

This equation quite fully covers real soil situations; it allows to estimate the depth of radon entry into the near-surface soil layers for cases of diffuse and convective transport mechanisms. Meteorological and climatic factors (Pichura et al 2019) are one of the determining factors for the RFD size from the soils of cities located in platforms of calm geodynamic conditions. It has been established (Hrozdynskyi 1965) that during the day due to solar radiation the increased turbulence over the soil surface leads to intensive air mixing; at night and in the morning temperature inversion suppresses radon transfer. This dependence in the daily variations of lightness can lead to a double change in the rate of exhalation.

The impact of biotic factors leads to loosening of soil by the roots of plants, occurrence of channels where rotten roots used to take place. The activity of burrowing animals, earthworms collectively leads to a complete restructuring of the parent rock, changes in its structural features and permeability. The soil layer is characterized by an intense development of macropores-large secondary openings and channels with sizes more than 1 mm, the occurrence of which is caused by the processes listed above, so as cracks that occur with changes in humidity, temperature and mechanical (man-made) effects. The depth of the soil layer that contains macropores and cracks is determined by the type of soil, climatic and other conditions and usually it is 25-50 cm, very

rarely it is 1 m. Soil compaction (for example, due to trampling) leads to the collapse of macropores and, accordingly, deterioration of soil aeration, which contributes in the accumulation of radon in the soil air. Radon anomalies that are connected with soil over compaction are well known in uranium exploration geology and they are called the "path effect" (Miklyaev 2001). It should be noted that the radon exhalation is influenced by soil moisture, its porosity and permeability, but fluctuations in the values thereof can be considered insignificant for several days and at distances of several kilometers.

The degree of filling of soil pores with water (moisture saturation) is increased together with humidity and, accordingly, the exhalation of radon is decreased due to the dissolution of its atoms in water. When saturation is more than 70%, the air movement in pore channels of soil is practically absent. The release of gases into the atmosphere in this case is possible only due to diffusion from the surface of weakly permeable blocks. When moisture is slightly decreased, the macropores are dried and the gas permeability is significantly increased. It reaches its maximum values when the degree of pore filling with water is less than 15–20% (Shestakov 1982, Miklyaev 2001). Humidity and, accordingly, the gas permeability of soils and rocks in zone of incomplete water saturation (from ground surface to groundwater level) are subject to significant temporal fluctuations. The nature and amplitude of fluctuations changes together with depth. The highest amplitude and aperiodic moisture fluctuations associated with changes in meteorological conditions, precipitation regime, water consumption for evaporation and transpiration (moisture consumption by plant roots) are observed in the upper layer of soil that contain macropores. Seasonal nature is taken with depth of moisture fluctuations and the amplitude dry out. The depth, at which moisture fluctuations completely dry out, ranges from (2-3 m) for clay soils up to (5-10) m for sands and sandy loams (Shestakov 1982).

Natural and artificial land cover (ice, snow, asphalt, concrete, etc.) also affect the decrease in radon exhalation. So, snowing on the ground, freezing of underlying surfaces, heavy showers facilitate in accumulation of radon in soil. Intensive infiltration of precipitation can cause a decrease in radon concentrations in the upper areas or throughout the zone profile. Therefore, in the marshy lands the zone of air migration of radon is almost completely absent (Miklyaev 2001). The reason for interseasonal variations of RFD from soil is the active so-called "restructuring" of the soil, when during 24 hours the daily surface is unevenly affected by significant temperature differences, resulting in short-term conditions for formation of crack network in frozen soil layer

**Table 4.** Radon exhalation from the underlying surfaces of various types

Type of surface subject to emanation	RFD, $\text{mBq}/(\text{m}^2 \cdot \text{c})$
Recent deposits (soils, sands, clays)	1.85–11.1
Various types of granite*	18.5–111
Zones, that contain secondary uranium minerals	3700–11100
Water surface	$<3.7 \cdot 10^{-3}$

\* - in spots with high radioactivity of soil, for example, in areas of granite release the content of radon in the air and RFD is significantly increased

and intensive migration of moisture in soil and radon. The water content of the soil in this case plays the screening role for soil radon in warm time of the day, and it acts as a factor of initial formation of surface cracks when the soil moisture freezes during cold time of day (Kurguz 2002).

Provided that reliability is sufficient, the RFD from soil and temperature relationship can be approximated by the function of the form:

$$RFD = 34.47 + 0.53 \cdot t \tag{2}$$

It is obvious that the RFD from soil increases on average by 5.3 mBq/(m<sup>2</sup>·c) at every 10 ° C (Kurguz 2002). The increase in atmospheric pressure also reduces radon intake in the surface air. The anomalous component of the RFD is typical for territories of cities with clear deformations of soil, which were formed due to tectonic activity. It appears during earthquakes that can occur even over a distance of tens of

thousands of kilometers from the places of registration of RFD from soil and VA in the spaces. The value of the equilibrium volumetric activity (EEVA) of radon in the spaces of such cities may exceed permissible levels by tens and hundreds of times (Pavlov et al 2003).

The highest RFD values are associated to territories, the surface of which is composed of clay deposits (110-170 mBq/(m<sup>2</sup> s)) (Fig. 5). Areas that are composed mainly of sandy soils are characterized by reduced average RFD values in general [20-40 mBq/(m<sup>2</sup>s)].

The morbidity and mortality statistics of Rivne city in relation tracheal, bronchial and lung cancer in the period 2014-2016 was analused (Golovne Upravlinnja 2016). The obtained data was indicated on the map of tested sites according to the addresses of the sick and dead people (Fig. 6). High correlation between the RFD values from the soil and

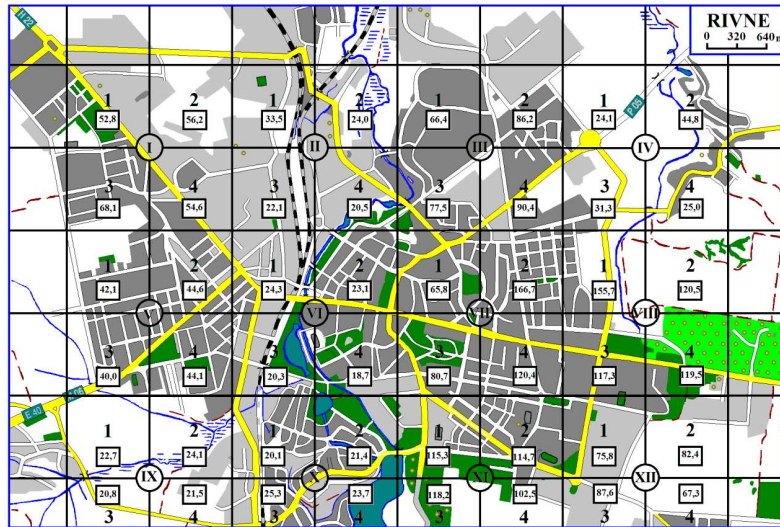


Fig. 5. Average values of RFD from soils of tested sites of Rivne city

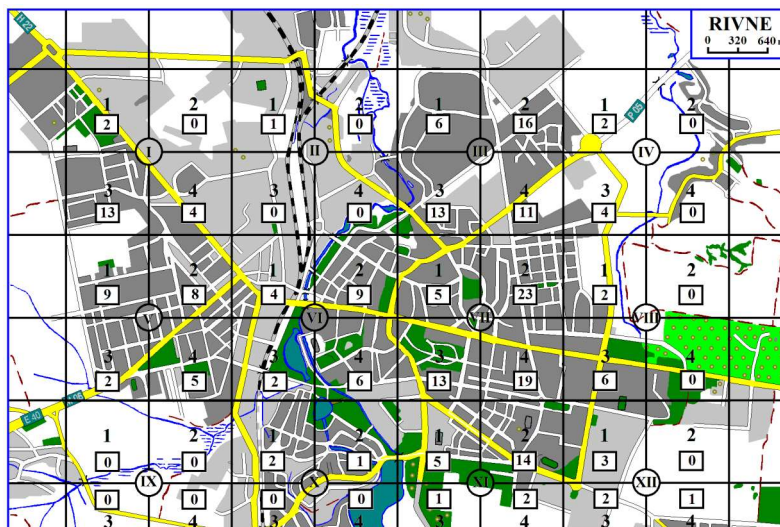


Fig. 6. Number of citizens who have or died due to lung cancer in 2014-2016 in tested sites of Rivne city



the mortality rate due to lung cancer in corresponding tested sites may be noted. The lowest RFD values from the soil were recorded in plot X. For the VII.2 and VII.4 tested sites, where the number of deaths due to lung cancer during the specified period was 23 and 19, respectively, the highest values of RFD are observed in Rivne city: 166.7 and 120.4 mBq/(m<sup>2</sup>s), respectively.

### CONCLUSIONS

The measured characteristics of the radon field (RFD) are, obviously, a sensitive indicator of the spatial and temporal changes in natural factors, including the difference in atmospheric temperature, pressure and precipitation; changes in the radiological and physico-mechanical properties of rocks; available structural features of rocks (zones of fragmentation, karst); geodynamic state of rocks (preparing for earthquakes and landslide processes). The analysis of spatial distribution of the RFD in the territory of Rivne city shows that the radon flux density field has discrete spatial structure and it is divided into background and anomalous components. The spatial distribution of the radon flux density is, obviously, log normally described, the value of the RFD varies widely, but the overwhelming majority (99%) falls into the 3-sigma interval for lognormal distribution. The interval is 10-180 mBq/(m<sup>2</sup>c) for the average values for tested sites with the arithmetic average of 70±7 mBq/(m<sup>2</sup>c) and geometric average 60 mBq/(m<sup>2</sup>c). These values may be considered as regional RFD background for the territory of Rivne city (background radon field). Comparison of the spatial distribution of the background radon field and the field of morbidity and mortality of citizens due to lung cancer in 2014-2016 indicates clear correlation between them.

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