



THE INVESTIGATION OF TOBACCO SMOKE INFLUENCE ON THE CHANGES OF INDOOR RADON AND ITS SHORT-LIVED DECAY PRODUCTS VOLUMETRIC ACTIVITIES

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Submitted 9 May 2012; accepted 29 Oct. 2012

Abstract. The changes of radon and its short-lived decay products were investigated in accommodations under natural living conditions and in tobacco smoke-filled premises. The measured radon and its short-lived decay products volumetric activities, aerosol particle concentration in the air, radioactive equilibrium factor, unattached fraction factor values are presented. It was identified that the increase of aerosol particles concentration in the air (in smoke-filled premises) determines the increase of the unattached nuclides of radon short-lived decay products attached to aerosol particles (the average values of radioactive equilibrium factor varied from 0.35 to 0.72). In this case, larger volumetric activity of the alpha particles is registered. Therefore larger amount of radon progeny is inhaled in smoke-filled premises and there is an increased possibility of damaging the organism. Positive correlation ($r = 0.9$) between the radioactive equilibrium factor and aerosol particle concentration in the air of accommodation, as well as negative correlation ($r = -0.64$) between the radioactive equilibrium factor and unattached fraction factor have been determined. Seasonal changes of the radioactive equilibrium factor are presented.

Keywords: radon, short-term decay products, volumetric activity, aerosol particle, tobacco smoke, radioactive equilibrium factor, unattached fraction factor.

Introduction

The ^{222}Rn is radioactive scentless and colorless inert gas. It is formed in the result of decaying of ^{226}Ra , which is the member of decaying chain of ^{238}U isotope. Radon and its decay products compose most of ionizing radiation (Girgždys, Ulevičius 1985; Gesell 1983). The average annual irradiance dose of Lithuanian inhabitants, aroused by natural sources, is about 2.2 mSv, about 1 mSv is conditioned by radon and its decay products' ionizing radiation (UNSCEAR 2000).

Radon is inert gas (half-life duration 3.824 days), therefore humans' health is more effected by its non-gas short-lived decay products: ^{218}Po (half-life 3 minutes), ^{214}Pb (27 minutes), ^{214}Bi (20 minutes), ^{214}Po (164 μs).

Most of radon radiation and its progeny is easily absorbed by human's skin surface, therefore human's inner irradiance is the most important, that is the irradiance due to inhaled radon decay products in the organism (Hess *et al.* 1983; Mohammed 1999). Having inhaled the air filled with radon short-lived decay products, they settle and decay on breathing organs. Then exuded alpha particles penetrate and damage the tissue. Due to that lungs or breathing organs cancer can develop (Lubin 2003; Ishikawa *et al.* 2003) (Fig. 1).

In Lithuania as in many other countries the largest source of radon is soil (Clavensjo *et al.* 1999; Mastauskas, Morkūnas 1996). The radon inert gas easily enters the premises through the gaps from the soil, which the house is built on, as well it is exuded from construction materials or even from the water. Thus, the big amount of radon is stored in the premises and after its decaying the hazardous short-lived products volumetric activities are formed, especially if the premises are not winded (Morkūnas 2000; El-Hussein *et al.* 1999; Andersen *et al.* 1997).

The larger the amount of aerosol particles in the air, the larger part of radon progeny settles on these particles but not on furniture, walls, or curtains. Thus, there are more radon short-lived decay products in a smoke-filled room than in a "clean" room with the same concentration of radon in the air (Dwiwedi *et al.* 2001; Mohamed 2005).

The influence of radon progeny on health depends on its behavior in the accommodations. There are two types of decay products: "unattached to aerosol particles" (the diameter of their diffusion equivalent is from 0.5 nm to 5 nm), and "attached to aerosol particles" (the diameter of particle is from 5 to 3 000 nm) (Hopke *et al.* 1992; Reineking *et al.* 1994).

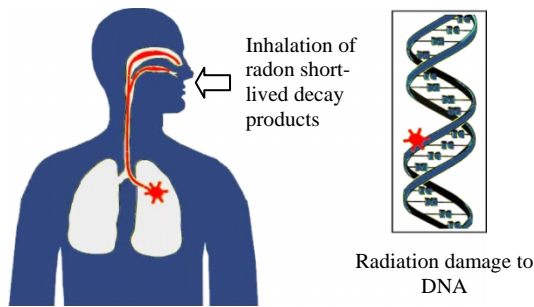


Fig. 1. The influence of radon short-lived decay products on human's organism

The nuclides of attached and unattached to aerosol particles are not separated while carrying different research on radon progeny out. It is important to evaluate the amount of attached and free nuclides in premises because due to different diffusion of molecules the short-lived decay products settle in different places of the airway and in different amounts (Castren *et al.* 1985; Swedjemark 1990).

It is proven by research that smoking strengthens the negative effect of radon. A smoker, who is together with a non-smoker in premises with the same amount of radon, risks to get a cancer even more than a non-smoker because radon progeny settle on tiny particles of tobacco smoke (i.e. aerosol), and newly appeared particles affect the airways much more. Though general effect of smoking and radon is still under investigation, there are some data showing that if the amount of radon in the premises increases 4 times, the possibility for a smoker to get lungs cancer is about 9 times higher than for a non-smoker (Radiation Protection Centre newsletter 2008).

The concentration of radon progeny and their location in the environment depend on the "behavior" of the first radon decay product ^{218}Po (Ho *et al.* 1982; Leung *et al.* 1998). This "behavior" is conditioned by such processes as cluster formation, neutralization of electric charge, attachment to the aerosol particles, and deposition on the surfaces. The electric charge of particles with ^{218}Po nuclides has a big impact on these processes and on the spectrum of radon short-lived decay products bearers (i.e. aerosol particles) (Porstendorfer 1994).

About 90% of ^{218}Po ions appeared after the decay of radon are positively charged. Part of ^{218}Po ions is neutralized; therefore another part of ions is neutral. ^{218}Po attaches to the aerosol particles, which are always in the air in natural environment, or clusters. Some of ^{218}Po ions, attached to clusters or aerosol particles, deposit on the surface. These processes of attachment to aerosol particles and surfaces depend on clusters and aerosol diffusions, and electric charges (Porstendorfer *et al.* 2005; Li, Hopke 1992).

The amount ratio of radon to its decay products is defined by radioactive equilibrium factor, usually

named as F factor (Mohamed 2005; Baixeras *et al.* 1999):

$$F = \frac{(0,105C_1 + 0,516C_2 + 0,379C_3)}{C_0}, \quad (1)$$

where C_0 is radon volumetric activity; C_1 , C_2 , C_3 are ^{218}Po , ^{214}Pb and $^{214}\text{Bi}/^{214}\text{Po}$ volumetric activities in the air ($\text{Bq}\cdot\text{m}^{-3}$).

This factor shows radioactive balance between radon and its progeny. The radioactive equilibrium factor depends mostly on winding intensity and concentration of aerosol particles in the air (Clavensjo, Akerblom 1994; Ramola *et al.* 2003).

Radioactive equilibrium factor is used as a factor to proceed from radon volumetric activity to its decay products volumetric activity in the premises, or vice versa. Radioactive equilibrium factor makes it possible to calculate the conditional effecting dose of radon in the premises (Baeza *et al.* 2003).

Another important parameter, which characterizes radon progeny indoors, is unattached fraction factor, which shows which part of radon decay products is not settled on aerosol particles in accommodations (Yu *et al.* 1996):

$$f_i = \frac{C_i^{eq}}{C_i}, \quad (2)$$

where C_i^{eq} is balanced equal activity. C_i is volumetric activity of each radon decay product unattached nuclides.

Equivalent equilibrium concentration is expressed (Mohamed 2005; Yu *et al.* 1996):

$$C_{eq} = 0.105C_1 + 0.516C_2 + 0.379C_3, \quad (3)$$

where C_1 , C_2 , C_3 are radon short-term decay products (^{218}Po , ^{214}Pb , ^{214}Bi) volumetric activities in the air.

The aim of this work is to investigate the influence of tobacco smoke on changes of indoor radon and its short-lived decay products volumetric activities, evaluate the dependence of unattached fraction factor and radioactive equilibrium factor on concentration of aerosol particles in the air.

1. Materials and methods

Measurements were carried out applying invented equipment and method for constant measurements of radon short-lived decay products volumetric activities in the air (Jasaitis, Girždys 2007).

The measuring equipment consists of optical aerosol particles counter, filtration device with a radiometer and air volume meter with a pump (Fig. 2).

The metal frame of filtration device has inlet and outlet. Inlet is used for air suction. A pump is attached to the outlet. The diameter of the air inlet is 20 mm.

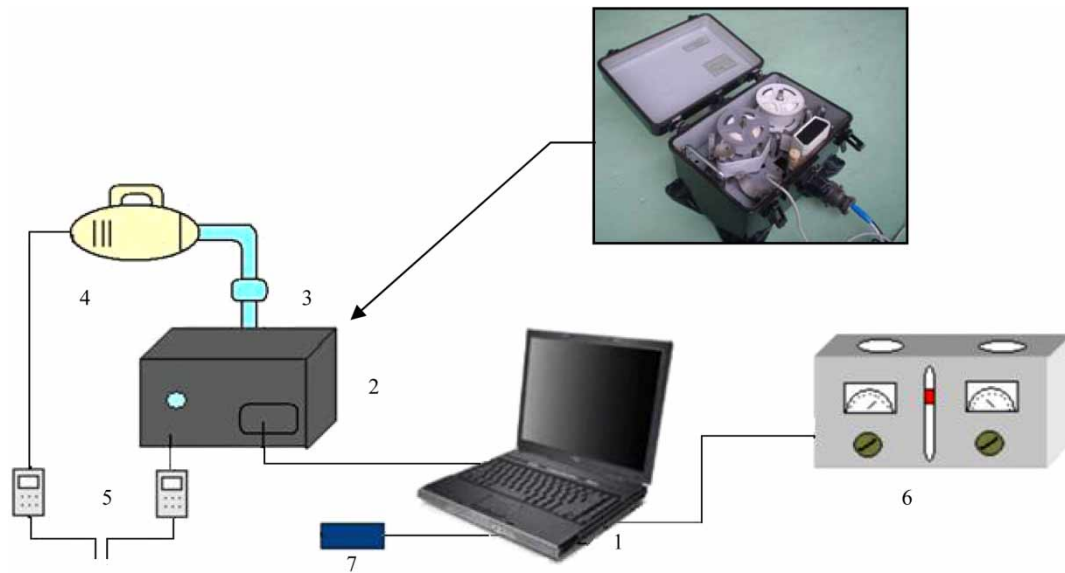


Fig. 2. Measurement scheme: 1 – computer; 2 – filtration device; 3 – air volume meter; 4 – air pump; 5 – stopwatches; 6 – optical aerosol particle counter; 7– multifunctional data logger

The air suction rate is 40 l/min. and it is controlled by air flow meter. The air in the device frame changes within 25 seconds. The 800 W powered air pump sucks the air through filter band “Fiberglaz” which diameter is 50 mm. The air is sucked through the part of filter band which is between a suction channel and a radiometer. The band is stopped during the measurement period.

When the measurement is over, the band is overwound so that the “clean” (i.e. without radon progenies) part of the filter appears above the radiometer and the air suction channel of the pump. The radiation of the particles, settled on the filter, is measured with the radiometer GM-45.

The radiometer GM-45 is small, light and extremely sensitive detector of the ionizing radiation. It contains a Geiger - Müller counter, which is sensitive to alpha, beta and gamma radiation (alpha above 3 MeV, beta above 50 keV, gamma above 7 keV). The diameter of isinglass is 42 mm. The RAD (Radiation Acquisition and Display) programme is included in this device. The usage of RAD programme enabled data to be stored and transferred into MS Excel programme, where they could be processed. During the measurement data were automatically recorded and saved.

The radiometer is fixed so that the filter band through which the air is sucked moves beside the radiometer measurement window (Fig. 3).

Electronic stopwatches TS-ED1 are set, and the time of suction and when the filter band is turned are programmed. Having set the hours when the measurements are automatically carried out, the device can operate permanently.

The concentration of particles attached to aerosol and “free” nuclides of radon progeny and their small

clusters in the air are assessed by plugging in and then unplugging the collector of “free” nuclides and clusters to the entrance of the air flow in the control device. This collector is made of 10 layers of a wire (the diameter is 0.2 mm) net bundle. The net bundle is connected to soil so that the charged nuclides can deposit on the surface of the wire more efficiently. Free positively charged nuclides of ^{218}Po and other short-lived decay products and small clusters deposit on the surface of the net wire due to high mobility. The nuclides attached to aerosol particles pass the net bundle; they are collected by the filter band “Fiberglaz” where after the radioactive decay they are registered with the radiometer.

The aerosol particle concentration is measured with an optical aerosol particle counter AZ-5. The aerosol counter measures the light scatter of separate

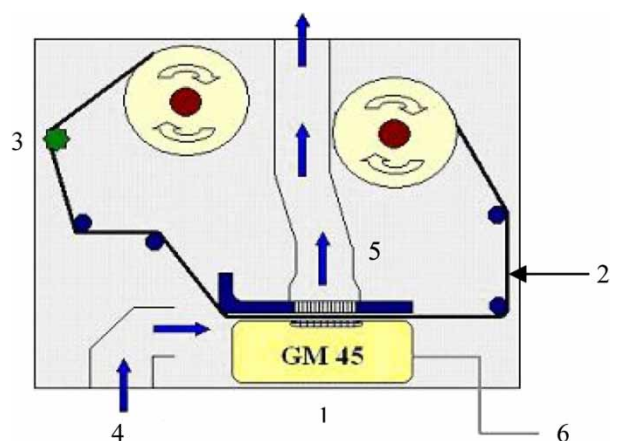


Fig. 3. Filtration device: 1 – radiometer; 2 – filter band; 3 – filter band speed regulator; 4 – air flow pumped from outside; 5 – air flow pumped by pump; 6 – data sent to the computer

aerosol particles and can rate aerosol particles sized from 0.4 to 10 μm .

The aerosol counter AZ-5 consists of an optical sensor, a pump and an electrical part. The optical sensor creates an electric pulse for each particle. The amplitude depends on the size of particles. The pump sucks the air through the measuring zone of the optical sensor (capacity 1.2 l/min). AZ-5 can measure the concentration of aerosol particles in the air from 0 to 310^6 particles per liter. Possible systematic bias is $\pm 20\%$.

In this measuring method the aerosol particle counter is connected to the computer, to which data are sent and recorded. With the help of a data logger ADC-16 the data of continuous measurements are accumulated in the computer. The concentration of aerosol particles and its alterations are constantly controlled.

The bias of radon short-lived decay products measuring method was evaluated. It depends on the efficiency changes of the filter band, the speed of air flow through it, exposition time, and the concentration of progeny in the air. The established bias was not higher than 6%.

2. The Results of Measurement

The research was carried out in the rooms of an individual house under natural living conditions and having increased the concentration of aerosol particles in the air, i.e. after having smoked in the room.

The average radon short-lived decay products volumetric activities were measured under natural living conditions on all the floors. The results of the measurements are presented in Fig. 4.

Considering the measurement results in the cellar, on the ground and first floors, it can be stated that the main source of radon progeny in the investigated house is the soil under the house, as it is usual in Lithuania and other countries (Clavensjo *et al.* 1999), which conditions the highest level of radon and its decay

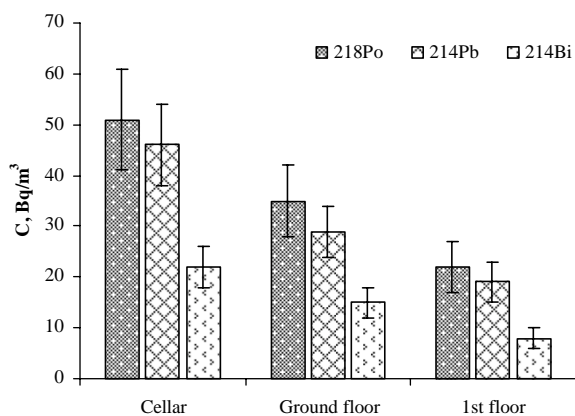


Fig. 4. Average radon short-lived decay products volumetric activities on different floors of the house

products volumetric activities in the cellars or on the ground floors (if there is no cellar).

Further measurements were carried out on the ground floor of the house. It was chosen because inhabitants of the house usually spend most of their time there, and the first floor is used for bedrooms. The volumetric activity of radon progeny in bedrooms is low (Fig. 4), what is more, these premises are usually winded before sleep. The plan of the house is presented in Fig. 5.

Radon volumetric activities (C), unattached fraction factor (f_{Po}), radioactive equilibrium factor (F) and aerosol particle concentration (N) in the air under natural living conditions were measured in all rooms of the investigated house. The results of measurements are presented in Table 1.

The values of radon volumetric activities altered from 16 to $45 \text{ Bq}\cdot\text{m}^{-3}$. The statistical bias of the measurement reached up till 15%. It was determined that the average radon volumetric activity on the ground floor of the investigated house was $34 \pm 4 \text{ Bq}\cdot\text{m}^{-3}$. To compare the average volumetric activity of individual houses in Lithuania is about $56 \text{ Bq}\cdot\text{m}^{-3}$ (Clavensjo *et al.* 1999).

According to valid hygienic norms in Lithuania the volumetric activity of radon should not be higher than $200 \text{ Bq}\cdot\text{m}^{-3}$ in newly built premises, and $400 \text{ Bq}\cdot\text{m}^{-3}$ in already existing premises. As it is seen in Table 1, radon volumetric activities are lower in those premises, which are winded better and which are less hermetic (Fig. 5).

The values of the unattached fraction factor altered from 0.05 to 0.11, calculated average value is 0.08 ± 0.02 . It was determined that the increase of aerosol particles concentration in the air of accommodations conditions the decrease of the unattached fraction factor. Also the inverse dependence between unattached fraction factor and radioactive equilibrium factor was determined.

The values of radioactive equilibrium factor altered from 0.33 to 0.65, calculated average value is 0.39 ± 0.03 . A tight correlation (correlation coefficient $r = 0.85$) between the radioactive equilibrium factor and aerosol particles concentration in the air was



Fig. 5. The plan of the ground floor of the investigated house: 1 – anteroom; 2 – hall; 3 – toilet; 4 – kitchen; 5 – office room; 6 – living-room

Table 1. Measured values of radon volumetric activities (C), unattached fraction factor (f_{Po}), radioactive equilibrium factor (F) and aerosol concentration (N) in the air on the ground floor of the investigated house under natural living conditions

Nr.	$C, \text{Bq} \cdot \text{m}^{-3}$	f_{Po}	F	$N \times 10^3, \text{cm}^{-3}$
1	16 ± 3	0.11 ± 0.02	0.33 ± 0.03	1.6 ± 0.19
2	29 ± 4	0.08 ± 0.02	0.34 ± 0.03	1.9 ± 0.20
3	32 ± 4	0.08 ± 0.02	0.37 ± 0.03	2.4 ± 0.20
4	45 ± 6	0.05 ± 0.01	0.65 ± 0.05	3.4 ± 0.24
5	40 ± 5	0.07 ± 0.02	0.37 ± 0.03	2.8 ± 0.22
6	42 ± 5	0.07 ± 0.02	0.38 ± 0.03	3.2 ± 0.22

determined. The higher aerosol particle concentration, the higher is the radioactive equilibrium factor, as the process of attachment becomes faster than the deposition of unattached particles on the surface.

The average absolute bias of measured aerosol particles concentration in the air reached 8%.

Further research was carried out to investigate what impact the increased concentration of aerosol particles has on indoor radon short-lived decay products alterations.

The measurements were carried out having smoked in the rooms of an individual house.

During the research the temperature in premises and relative humidity was about $21 \text{ }^\circ\text{C}$ and 47%, the average aerosol particles concentration reached $1.9 \cdot 10^3 \text{ cm}^{-3}$, the average radon volumetric activity was $32 \text{ Bq} \cdot \text{m}^{-3}$. The speed of winding in the premises was about 0.2 h^{-1} . Having smoked in the premises the aerosol particles concentration had increased till $95 \cdot 10^3 \text{ cm}^{-3}$. The results of measurement are presented in Table 2. Statistical averages of the measurements are presented there. In total 45 measurements were carried out.

Under natural conditions indoors the values of unattached fraction factor varied from 0.05 to 0.20,

Table 2. Aerosol concentration in the air (N), unattached fraction factors (f_{Po}) and radioactive equilibrium factors (F) in smoke-filled and non-smoked premises

	Under natural conditions	Smoke-filled premises
$N \times 10^3, \text{cm}^{-3}$	1.90 ± 0.20	92.5 ± 6.5
F	0.35 ± 0.03	0.72 ± 0.06
f_{Po}	0.09 ± 0.02	0.03 ± 0.01

and the average value was 0.09 ± 0.02 . The values of radioactive equilibrium factor altered from 0.15 to 0.50, and the average value was 0.35 ± 0.03 .

Usually the indoor aerosol particles concentration is low, therefore only very small amount of radon short-lived decay products attaches to the particles. It means that most of radon progeny appeared after the decay are free (unattached), and they settle on different surfaces. Facts mentioned above condition the radioactive disbalance between radon and its short-lived decay products. As it was mentioned before with the increase of aerosol particles concentration the radioactive equilibrium factor increases too.

Having smoked in the accommodation the aerosol particles concentration in the air has increased, which conditioned the increase of average value of radioactive equilibrium factor till 0.72 ± 0.06 and the increase of average value of unattached fraction factor till 0.03 ± 0.01 .

Figure 6 shows a typical example of changes of radioactive equilibrium factor and unattached fraction factor during day and night period, where the concentration of indoor aerosol particles is changing under natural conditions and in smoke-filled premises.

It was noticed that with the increase of aerosol particles concentration in the air the unattached fraction factor was decreasing – the values of factors were only reaching 0.03–0.05. At the same time the

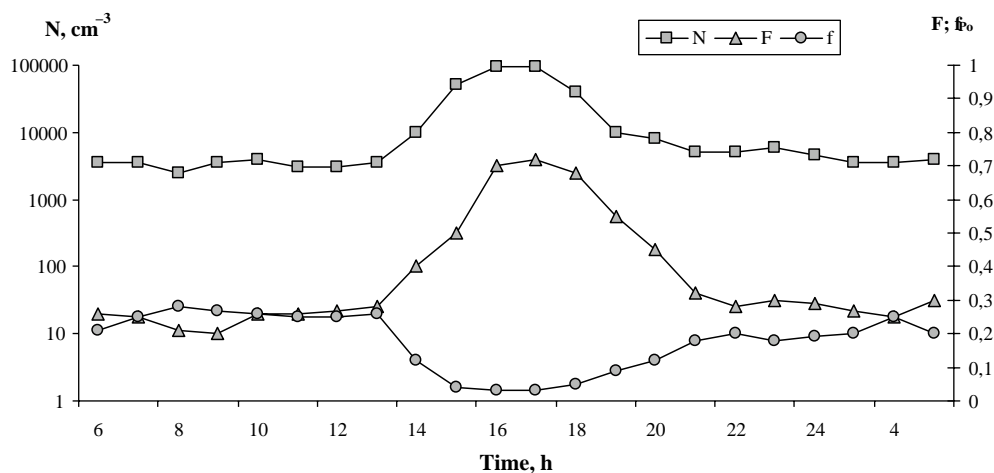


Fig. 6. Changes of unattached fraction factor and radioactive equilibrium factor when the concentration of aerosol particles in the air alters due to the smoke in the premises

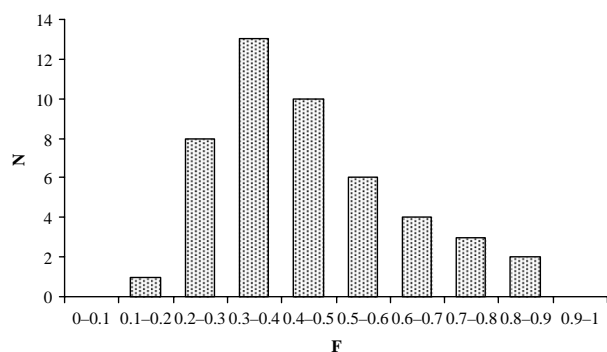


Fig. 7. Repeated frequency of indoor radioactive equilibrium factor

radioactive equilibrium factor was increasing, its values reached 0.6–0.7, and in some cases even about 0.8–0.9.

As it is seen in figure 6, there is an inverse dependence between radioactive equilibrium factor and unattached fraction factor. The negative correlation between the unattached fraction factor and the radioactive equilibrium factor was obtained, which can be expressed as $f = 0.06 \cdot F^{-0.64}$. Such dependence was also determined during other measurements (Politova, Jasaitis 2010). What is more, a clear correlation between radioactive equilibrium factor and indoor aerosol particles concentration was determined, the correlation coefficient $r = 0.9$.

Figure 7 presents the repeated frequency of calculated radioactive equilibrium factors in different investigated accommodations.

Usual repeated value of radioactive equilibrium factor is 0.3–0.4.

The seasonal average values of the radioactive equilibrium factor in the premises are presented in Figure 8.

It was determined that the seasonal values of the radioactive equilibrium factor in the investigated house varied from 0.43 to 0.52 (Jasaitis, Girgždys 2011).

Figure 8 shows, that the values of radioactive equilibrium factor are the highest in winter and the lowest in summer. In winter the accommodations are

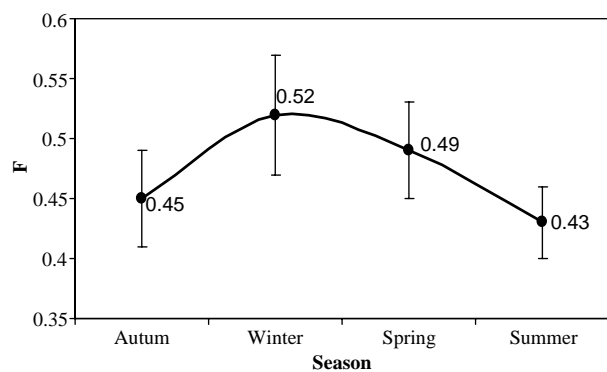


Fig. 8. Seasonal variations of radioactive equilibrium factor in the investigated accommodations

less winded and the house is more hermetic than during other seasons. This allows radon and its progeny to accumulate. Therefore the radioactive equilibrium factor between radon and its progeny increases. In summer accommodations are well ventilated and this lowers the radioactive equilibrium factor between radon and its progeny (Porstendorfer *et al.* 1994).

The obtained measurement values of radioactive equilibrium factor are similar to the values calculated in other European countries. The received values are similar to the measurements in Holland and Germany (Cavallo 2000; Kreuzer *et al.* 2003). It is noticed that the radioactive equilibrium factor is higher in cold countries than in tropical countries. However, having in mind a wide range of radioactive equilibrium factor variations in different places, it is recommended to calculate it separately for specific regions.

Conclusions

1. The measured average radon volumetric activity in the investigated house was $34 \pm 4 \text{ Bq} \cdot \text{m}^{-3}$. It is lower than average volumetric activity in individual houses in Lithuania (about $56 \text{ Bq} \cdot \text{m}^{-3}$). It was determined that radon volumetric activities were lower in better winded, less hermetic accommodations of the investigated house.

2. Considering the results of volumetric activities measurements in the cellar, on the ground and first floors of the house, it has been determined that the main source of radon progeny in the investigated house is the soil under the house.

3. Under natural conditions the average values of unattached fraction factor and aerosol particles concentration were 0.09 ± 0.02 and $(1.90 \pm 0.20) \cdot 10^3 \text{ cm}^{-3}$. In smoked premises respectively – 0.03 ± 0.01 and $(92.5 \pm 6.5) \cdot 10^3 \text{ cm}^{-3}$. Having increased aerosol particles concentration, the amount of attached free nuclides of radon short-lived decay products to the aerosol particles increases, therefore higher volumetric activity of alpha particles is registered.

4. The average value of measured radioactive equilibrium factor under natural conditions was 0.35 ± 0.03 , at the same time the value in smoked premises often reached 0.7–0.8, therefore there is a higher possibility to inhale radon progeny and damage organism in the smoked premises.

5. It was determined that with the increase of indoor aerosol particles concentration unattached fraction factor decreased. What is more, the invert dependence between unattached fraction factor and radioactive equilibrium factor was determined.

6. Clear positive correlation between the radioactive equilibrium factor and aerosol particles concentration in the premises was determined, correlation coefficient $r = 0.9$. Correlation between radioactive

equilibrium factor and unattached fraction factor was negative.

7. The most frequently repeated value of the radioactive equilibrium factor is 0.3–0.4. It has been determined that radioactive equilibrium factor is lower in summer than in other seasons. Such seasonal change of the radioactive equilibrium factor due to better ventilated accommodations during warm season.

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