Journal of Environmental Radioactivity 183 (2018) 59–72

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Indoor radon regulation using tabulated values of temporal radon variation

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A B S T R A C T

Mass measurements of indoor radon concentrations have been conducted for about 30 years. In most of the countries, a national reference/action/limit level is adopted, limiting the annual average indoor radon (AAIR) concentration. However, until now, there is no single and generally accepted international protocol for determining the AAIR with a known confidence interval, based on measurements of different durations. Obviously, as the duration of measurements increases, the uncertainty of the AAIR estimation decreases. The lack of the information about the confidence interval of the determined AAIR level does not allow correct comparison with the radon reference level. This greatly complicates development of an effective indoor radon measurement protocol and strategy.

The paper proposes a general principle of indoor radon regulation, based on the simple criteria widely used in metrology, and introduces a new parameter – coefficient of temporal radon variation $K_V(t)$ that depends on the measurement duration and determines the uncertainty of the AAIR. An algorithm for determining $K_V(t)$ based on the results of annual continuous radon monitoring in experimental rooms is proposed. Included are indoor radon activity concentrations and equilibrium equivalent concentration (EEC) of radon progeny. The monitoring was conducted in 10 selected experimental rooms located in 7 buildings, mainly in the Moscow region (Russia), from 2006 to 2013. The experimental and tabulated values of $K_V(t)$ and also the values of the coefficient of temporal EEC variation depending on the mode and duration of the measurements were obtained. The recommendations to improve the efficiency and reliability of indoor radon regulation are given. The importance of taking into account the geological factors is discussed. The representativity of the results of the study is estimated and the approach for their verification is proposed.

1. Introduction

Radon is the main source of radioactive exposure for humans. On average, the radiation dose from radon is about 42% (1.2 mSv/year) of all known sources of natural and artificial radioactivity (UNSCEAR, 2008). According to the World Health Organization, up to 14% of all lung cancers are caused by radon (WHO, 2016). In contrast to the action, for example, of cosmic radiation, the exposure from radon can be regulated and mitigated. Therefore, in many countries there are limits set for the radon concentrations in buildings. For this purpose, measurements are taken to estimate the annual average indoor radon (AAIR) concentration and to compare it with the control levels, which range from 74 Bq m^{-3}, which serves as reference level in USA (ANSI/AARST MAH, 2014), to 4000 Bq m^{-3}, which is a limit level in Czech Republic (IAEA, 2017). If the national reference/action/limit level (in further for the simplicity: reference level) is exceeded, radon-protective measures should be carried out and then the AAIR should be determined again in order to decide about mitigation efficiency.

Mass measurements of indoor radon concentrations began in USA, United Kingdom and Sweden about 25 years ago (EPA, 1992). However, despite the extensive international experience, there is still no unified standard or measurement protocol to determine AAIR with a known accuracy.

The problem with AAIR is that indoor radon concentrations have significant temporal variation - diurnal, weekly and seasonal. Obviously, the most accurate estimate of AAIR would be obtained if the measurements were performed throughout the entire year. It is also clear that shortening the measurement duration increases the AAIR uncertainty. However, in the overwhelming majority of cases, the determination of AAIR is based on short-term measurements. For example, over the last 25 years more than 98% of the indoor radon measurements in the USA were performed with short-term testing devices (George, 2015). This is due to the fact that the AAIR in the majority of buildings is much lower than the reference level, because most

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https://doi.org/10.1016/j.jenvrad.2017.12.003
Received 1 October 2017; Received in revised form 4 December 2017; Accepted 7 December 2017

probable value of AAIR worldwide is ~30 Bq m^{-3} (UNSCEAR, 2006). In these cases, the high uncertainty of the AAIR (even at 100% or more) is entirely acceptable. However, the accuracy of the AAIR assessment based on short-term measurements is different due to the inconsistency in the duration of the tests, which may last minutes, days or even months (ANSI/AARST MAH, 2014; IAEA, 2017). In order to improve reliability of the results, the testing protocols require conducting the short-term measurements in special "closed-room" conditions, which are expected to provide a less intense and more stable air exchange (ANSI/AARST MAH, 2014; IAEA, 2017).

In contrast to the short-term tests, long-term measurements allow determining the AAIR more accurately. Durations from two-three months (ANSI/AARST MAH, 2014; Howarth and Miles, 2008; ISO 11665-8, 2012) to one year (IAEA, 2017) have been used, and two long-term measurements in different seasons of the year are recommended (IAEA, 2017; ISO 11665-8, 2012). However, the problem of the AAIR accuracy in long-term measurements is not solved yet, because the AAIR uncertainty depends on the measurement duration.

A review of the evolution of the radon measurement protocols in homes in the USA, from 1993 (EPA, 1993) to the present time (ANSI/AARST MAH, 2014), shows that the US EPA document "National Radon Proficiency Program" (EPA, 1997) published 20 years ago still remains the main guidance on quality assurance. According to (EPA, 1997), quality assurance regulates only the uncertainty associated solely with the procedure for measuring the activity concentration of indoor radon. At the same time, another, much more significant problem related to the estimation of the uncertainty due to radon temporal variation, has not been discussed in depth, and is not resolved yet. The national documents of the USA (ANSI/AARST MAH, 2014; EPA, 1993; EPA, 1997), the international ISO standard (ISO 11665-8, 2012), the IAEA publications (IAEA, 2013; IAEA, 2015; IAEA, 2017) – none of these documents discuss the AAIR uncertainty and how it depends on the measurement duration. The running European project "Metrology for Radon Monitoring" (MetroRADON, 2017) deals with the uncertainty of radon devices only, and also does not take into account the uncertainty of temporal radon variation.

The protocol developed in the United Kingdom (Howarth and Miles, 2008) allows estimating the AAIR uncertainty using tabulated values of the seasonal correction factor. This factor takes into account the influence of outdoor temperature and provides the upper limit of the AAIR depending on the specific month of the year or several consecutive months during which long-term measurements were carried out. In addition, this protocol provides a formula for the coefficient of temperature influence, which was proposed about two decades ago (Miles, 1998).

Indeed, many publications note that in winter, indoor radon concentration is usually higher, than in summer (Wilson et al., 1991; Papastefanou et al., 1994; Pinel et al., 1995; Vaupotic et al., 2001). However, this is not a strict rule. In some cases, an opposite trend is observed (Steck et al., 2004; Karpinska et al., 2004; Bochicchio et al., 2005; Font, 2009; Stojanovska et al., 2011). Therefore, the use of a seasonal (or any other) correction factor to reduce the AAIR uncertainty, as proposed in (Howarth and Miles, 2008), cannot be considered completely correct without control of certain conditions (meteorological parameters, building and premises operation and maintenance, controlling of HVAC system, testing mode, etc.) during the indoor radon measurement. These conditions will be called in further "testing conditions".

A large number of studies and publications (Karpinska et al., 2004; Bochicchio et al., 2005; Font, 2009; Stojanovska et al., 2011; Gillmore et al., 2005; Groves-Kirkby et al., 2006, 2009; Denman et al., 2007; Burke and Murphy, 2011; Hunter et al., 2011; Kozak et al., 2011; Barros et al., 2014, 2016) attempted to determine the values of the correction factor and its relationship with influencing parameters. It should be noted that even the existence of a reasonable correction factor and specified testing conditions does not cancel the need to estimate the AAIR uncertainty. Thus, in any case, there must be a certain algorithm for estimating this uncertainty. It is assumed that such an algorithm should be based on either: (a) the results of measurements of different durations, or (b) a calculation model that takes into account the specific features of climate and geology, building materials and building design, the existing HVAC system, composition and behavior of building occupants, the type and location of the testing room in the house, etc. The assessment of the AAIR and its uncertainty based on a calculation model, according to (b), has not been developed yet (Bossew and Lettner, 2007; Dubois et al., 2007; Friedmann et al., 2017). Therefore, a statistical analysis of the results of measurements with different durations seems to be the only practical way to formulate the algorithm for estimating the AAIR uncertainty.

No attempt to estimate the AAIR uncertainty was made in the works studied the correction factor. Among these publications, only Steck et al. (2004) give an estimate of the dependence of the radon coefficient of variation (COV) on both the measurement duration (2 and 4 days, and 1, 3, 4 and 6 months) and the operating conditions ("closed" or "normal"). The COV values, depending on the duration of the measurements, characterize the uncertainty of the AAIR. Steck et al. (2004) analyzed the results of radon surveys in 62 homes in Minnesota, the region of elevated radon and highly variable climate. The works of D. Steck and his co-authors (Steck et al., 2004; Barros et al., 2014, 2016) represent an important step towards understanding the uncertainty of indoor radon measurements. However, the data obtained by these authors are not suitable to estimate the AAIR uncertainty in a single premise, because they define COV in the traditional way – as a ratio between standard deviation to expected (mean) value, using the results of parallel short-term and long-term measurements in a large number of buildings. This approach does not take into account that frequency distributions of the radon activity concentrations in single premises are never regular (see Section 3 in more detail) and have no connection with the lognormal distribution of radon concentration in a set of buildings.

Moreover, D. Steck and his co-authors use the approaches and terms ("sensitivity", "specificity", "efficiency", "predictive value of a positive/negative test"), which are accepted in medical statistics (Mackinnon, 2000). These approaches and terminology may be not fully understood by the radon professionals involved in measurements, because cannot be used for estimating uncertainty of the measurement result in accordance with the fundamental metrological document (ISO/IEC Guide 98-3, 2008).

To conclude, the questions how to quantify the AAIR uncertainty and what is considered as a reliable (or repeatable) result of indoor radon testing are still unanswered. As a result, it is impossible to compare AAIR correctly with a reference level without an information about the AAIR uncertainty. Moreover, it is impossible to optimize the duration and strategy of the measurements and improve the measurement protocol of AAIR estimation. For example, there is no sense to use more accurate devices and carry out long-term measurements in rooms with low radon concentrations; as we know, such premises represent the overwhelming majority in the world. Finally, if the indoor radon survey results based on different test modes would have a known AAIR uncertainty, then they could be correctly averaged using the inverse-uncertainty as the weight of the determined AAIR in each particular testing object: this is especially important for building radon hazard maps, as well as for a more correct assessment of collective and individual doses from indoor radon.

The current paper attempts to solve these problems and suggests a new principle of indoor radon regulation, based on the simple criteria used in metrology, and introduces the coefficient of temporal radon variation $K_V(t)$ that determines the uncertainty of the AAIR. In addition, the original approach for determining and verification this coefficient depending on the measurement duration is proposed.

2. General principle of indoor radon regulation

The general principle of indoor radon regulation can be formulated as simple as possible: it is to determine the confidence interval of AAIR in the range from $\overline{C} - U(\overline{C})$ to $\overline{C} + U(\overline{C})$ (or range from 0 to $\overline{C} + U(\overline{C})$ if $U(\overline{C}) > \overline{C}$), and compare the boundaries of this interval with the reference level, according to the following three criteria.

Criterion 1. The AAIR does not exceed the reference level, if the condition (1) is met; then the measurements are stopped, and radon-protective measures are not needed.

$$\overline{C} + U(\overline{C}) \le C_{RL} \text{ or}$$
$$k \cdot C(t) \cdot [1 + \sqrt{K_V(t)^2 + U_D^2}] \le C_{RL}$$
$$[U_{rel}(\overline{C}) = U(\overline{C})/\overline{C} = \sqrt{K_V(t)^2 + U_D^2}], \tag{1}$$

where

$\overline{C}$ is the measured or calculated AAIR value, Bq·m^{-3};
C_{RL} is the reference level, Bq·m^{-3};
$U(\overline{C})$ is the uncertainty of the AAIR, Bq·m^{-3};
U_{rel} is the relative uncertainty of the AAIR, rel;
$C(t)$ is the measured average indoor radon concentration over the time period of t, Bq m^{-3};
k is a correction factor (rel), taking into account the influence of environmental factors on the "predictable" behavior of indoor radon under certain testing conditions, in which radon behaves with a given probability according to a known (experimentally determined) law; if the signs of the predictable behavior of radon are not established, then $k = 1$;
$K_V(t)$ is the coefficient of temporal radon variation, or the temporal radon uncertainty (rel); it depends on the mode and duration of the measurements and varies from 0 (if $t = 1$ year, but without year-to-year variations; see Section 8) to **more than 200%** (if $t < 2$ days; see Section 7); this coefficient expresses the value of the maximum deviation of $C(t)$ from the AAIR and is determined by the algorithm given in Section 3;
U_D is the uncertainty of the radon device (rel) or the relative uncertainty of the value of $C(t)$, which is usually in the range **(10–30)%**; we would like to remind that the document (EPA, 1997) and the project (MetroRADON, 2017) focus on controlling this kind of uncertainty only.

Criterion 2. The AAIR exceeds the reference level, if the condition (2) is met; then the measurements are stopped, and radon-protective measures are carried out.

$$\overline{C} - U(\overline{C}) > C_{RL} \text{ or}$$
$$k \cdot C(t) \cdot [1 - \sqrt{K_V(t)^2 + U_D^2}] > C_{RL} \tag{2}$$

Criterion 3. If both conditions (1) and (2) are not met, then the continuation of the measurements leads to the situation when one of these conditions will be met, because the parameters $C(t)$ and $K_V(t)$ depend on the measurements duration, while the values of $K_V(t)$ will always decrease. If, from the results of the additional measurements, the conditions (1) and (2) are still not fulfilled, then it is recommended to assume that the AAIR exceeds the reference level.

Thus, the parameter $K_V(t)$ represents the important part of AAIR uncertainty - especially during the short-term measurements, when condition $K_V(t)^2 >> U_D^2$ is usually satisfied (see Section 8). The values of $K_V(t)$ have to be determined with a satisfactory accuracy and tabulated for practical use. It must also be taken into account that under certain testing conditions the values $K_V(t)$ can be reduced by applying coefficient k.

The proposed principle is based on the recommendations of (ISO/IEC Guide 98-3, 2008) and guarantees reliable quality control for indoor radon tests from the metrological point of view, regardless the devices and methods of measuring the radon activity concentration. This simple and clear principle is widely used for quality control in scientific and industrial measurements, but it is still not used in testing indoor radon yet. In our opinion, it is due to the lack of: (a) extensive discussion of approaches for assessing the AAIR uncertainty, (b) understanding the relationship between the AAIR uncertainty and temporal variations of indoor radon, and (c) a method for determining the coefficient of temporal radon variation, which will be discussed in the next section.

3. Determination of the coefficient of temporal radon variation

To determine the values $K_V(t)$ we propose an approach, which is fundamentally different from earlier attempts to quantify the temporal variations of indoor radon (Steck et al., 2004; Karpinska et al., 2004; Groves-Kirkby et al., 2006, 2009; Denman et al., 2007; Burke and Murphy, 2011; Hunter et al., 2011; Kozak et al., 2011 Barros et al., 2014; Barros et al., 2016). The suggested approach is based on an analysis of the results of annual radon monitoring in representative experimental rooms with a high radon content (see Section 4). The concept of "representative rooms" refers to the premises of buildings that are most common within a large region or small country in a specific climate and geological environment. However, the algorithm for processing monitoring results does not depend on the type of premises and the building location. This concept allows classification of the premises, that can be used in the future analysis (when the statistical array of data of various annual monitoring is large enough), if a reliable connection between $K_V(t)$ and the type of premises is found.

We propose for practical use to tabulate the maximum values of $K_V(t)$ (called "tabulated values of temporal radon variation" in further) obtained from all experimental rooms (of a certain type, if the statistical array is large enough), and to take into account the dependence of this coefficient on the duration and mode of measurements, which are carried out either as one continuous measurement or as several measurements in different seasons of the year.

It has to be emphasized that the coefficient $K_V(t)$ cannot be determined as a conventional coefficient of variation (COV), since the distributions of the values of radon concentration $C(t)$ in premises have never a regular pattern (normal, lognormal or other; it can be clearly seen from Section 6). In this case, it is difficult to assess, and even more so to guarantee a reliable confidence level for COV-values. Therefore, in order to ensure that the confidence level of the confidence interval of the determined AAIR is at 95% for a particular premise or experimental room, we propose to determine the value $K_V(t)$ for a given measurement duration t directly from the analysis of the $C(t)$ distribution obtained from the continuous annual monitoring of radon concentration in experimental room with a period of registration (data averaging) equal to t.

Hence, according to (3)–(6), the value $K_V(t)$ will correspond to the maximal deviation of the lower or upper limits of the distribution of values $C_i(t)$ from the experimentally determined AAIR level $\overline{C_E}$, as shown in Fig. 1 (at $t = 4$ days, for example).

$$K_V(t) = \max[K_V^L(t); \quad K_V^U(t)] \tag{3}$$

at

$$|K_V^L(t) - K_V^U(t)| \to \min, \tag{4}$$

$$K_V^L(t) = \max[|\overline{C_E}/C^L(t) - 1|; \ |C^L(t)/\overline{C_E} - 1|], \tag{5}$$

$$K_V^U(t) = \max[|\overline{C_E}/C^U(t) - 1|; \ |C^U(t)/\overline{C_E} - 1|], \tag{6}$$

where $C^L(t)$ and $C^U(t)$ is the lower and upper limits, respectively.

The location of the lower and upper limits of this distribution (Fig. 1) is chosen so that the fraction of values of $C_i(t)$ in the cut-off "tails" does not exceed 5%, and satisfies the condition (4).

The values of the function $K_V(t)$ for a particular experimental room

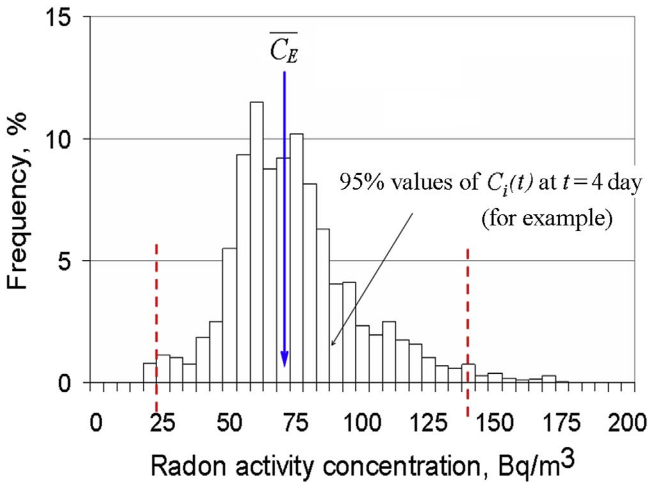


Fig. 1. Example of the radon activity concentration distribution and location of the lower and upper limits (the dashed lines).

over the measurement period, for example, from 1 day to 12 months, can be determined using Equation (3), based on the data of one annual monitoring in this room, if the registration period (averaging) does not exceed 1 day. However, in order to accumulate a statistically significant array based on the results of annual monitoring, for example, including 8760, 4380 or 2920 values, the optimal device registration period should be 1, 2 or 3 h, respectively. This allows to convert the original dataset consisting of the short hourly periods into the array with daily, weekly or monthly durations of measurement with the same interval (1, 2 or 3 h) by calculating the moving average. In this case, the number of values in the new (converted) array is reduced slightly.

In the case of conducting several measurements within one year, the values of the function $K_V(t)$ can be also determined by converting the same original dataset based on the results of annual monitoring in the same experimental room. Particular value of K_V in the case of 2 (or 4) measurements, each of duration t^* (from one day to several months), with an interval between the measurement starts of 6 (± 1) or 3 (± 0.5) months, respectively, is determined in the following sequence of steps:

(a) the original data set (with a registration interval of 1, 2 or 3 h) is divided into two (or four) equal parts, in which the data with the same sequential numbers correspond to the time of start measurements with the interval equal to 6 (or 3) months;
(b) the data of each array is separately converted by calculating a moving average over a period t^* with interval equal 1, 2 or 3 h, at that the size of the new array decreasing by a number of values equal to t^* divided by 1, 2 or 3 h, respectively;
(c) the resulting two (or four) arrays are converted into one by calculating the average of two (or four) values with the same sequential numbers;
(d) the minimum value $C_{\min}$ and the maximum value $C_{\max}$ of radon concentration are selected from the combined array, and then the K_V is calculating by the following formula:

$$K_V = \max\begin{bmatrix} |\overline{C_E}/C_{\min} - 1|;\ |C_{\min}/\overline{C_E} - 1| \\ |\overline{C_E}/C_{\max} - 1|;\ |C_{\max}/\overline{C_E} - 1| \end{bmatrix}. \tag{7}$$

The algorithm described before allows determining the dependence of $K_V(t)$ on the mode and duration of the indoor radon measurement in a particular experimental room. However, it should be taken into account that the satisfactory reliability of the tabulated values of the radon temporal variation coefficient can be obtained only from the analysis of the time dependence $K_V(t)$ covering a set of representative experimental rooms with characteristic behavior of radon, taking into account the influence of environmental factors under certain testing conditions. In some cases, the influence of environmental factors can be expressed quantitatively, through the coefficient k.

Determining the structure and values of the correction factor k, as well as its relation to the coefficient $K_V(t)$ and the classification of premises, is a complex task, which will be discussed in detail in a separate article. Within the framework of this article, k = 1.

4. Experimental rooms

Average global concentration of outdoor radon is about 10 Bq m^{-3} (UNSCEAR, 2008). In some regions, the average outdoor radon may increase to 25 Bq m^{-3} (Barros et al., 2015) and even to higher levels (ISO 11665-1, 2012). Usually in the regions with increased outdoor radon, there are also increased indoor radon (due to a higher release of soil radon), so the reference level is usually also higher. Thus, the main requirement for representative experimental room is an increased indoor radon concentration, which, on average, must significantly exceed outdoor radon concentration - at least by 5 times. This requirement is explained because the indoor radon is of our concern in cases if its average concentration approaches or exceeds the reference level. Otherwise, if the outdoor radon is a significant source, then the AAIR will be much less than the reference level.

The experimental part of the study was conducted between 2006 and 2013 and covered ten rooms with a high radon concentration. These rooms were located in six buildings located in Moscow and Moscow region, and one building in the Ryazan region, i.e. within the same climatic zone. Outdoor radon is about 10 Bq m^{-3}.

The most important characteristics of the selected experimental rooms are assembled in Table 1, including the characteristics of buildings, geology and climate. These characteristics are easy to obtain when inspecting rooms and buildings, as well as from the Internet. Such a detailed description is necessary for the classification of premises and the identification of the factors (qualitative features) that most affect the temporal variations of indoor radon and the $K_V(t)$ values. Thus, the classification of premises based on simple qualitative characteristics can significantly improve the efficiency of the indoor radon measurement protocol. Such classification would be improved in the future even more, if a large array of annual radon monitoring data in different rooms of different buildings could be collected.

All experimental rooms had natural ventilation. The experimental rooms ER 1, 3, 4, 5, 6 and 10 were almost not occupied, and these rooms were always closed. The other four rooms ER 7–9, including ER 2, were operated normally and periodically occupied. The ER 2 was a laboratory, where the stable temperature conditions existed - due to the presence of additional walls inside the basement.

The ER 6 was also constructed as a separate room inside the basement floor, and served as a special radon room with very low air exchange rate, an average of 0.1 h^{-1}. It has to be noted that this very low air exchange rate can be considered minimal for different types of premises in any buildings. The air exchange value was obtained by calculation for the known AAIR level and the known radon emanation rate of the source. The same artificial radon source (liquid solution of ^{226}Ra) was used in the rooms ER 1 and ER 6. This source has a high and stable emanation rate equal to 3.0 Bq s^{-1} (or 260 and 500 Bq m^{-3} h^{-1}, taking into account the volume of the rooms ER 1 and ER 6, respectively).

The experimental room ER 3 looks especially interesting, because its entire floor is an open soil, which obviously can be considered the main source of radon in this room. Due to the uneven floor, the height of the room varies by almost two times, according to Table 1. In this regard, for the purpose of a more detailed comparative study of the influence of weather conditions on the indoor radon behavior, the annual monitoring began simultaneously in the ER 1 and ER 3 rooms, which are fundamentally different. According to Table 1, the ER 1 is a small closed room inside an attic with a stable artificial radon source, while the ER 3

Table 1
Characteristics of experimental rooms.

Room	Code		ER 1	ER 2	ER 3	ER 4	ER 5	ER 6	ER 7	ER 8	ER 9	ER 10
	Measurements started (one year duration)		20.10.2006	08.10.2009[a]	20.10.2006; 02.10.2009[a]	02.10.2009[a]	05.10.2009[a]	21.09.2012	23.11.2012; 12.11.2012[a]	02.11.2011	02.11.2011	20.10.2011; 20.10.2011[a]
	Radon device		AlphaGUARD	AlphaAERO[a]	AlphaGUARD; AlphaAERO[a]	AlphaAERO[a]	AlphaAERO[a]	AlphaGUARD	RadonSCOUT; AlphaAERO[a]	RadonSCOUT	RadonSCOUT	RadonSCOUT; AlphaAERO[a]
	AAIR level, Bq·m^{-3}		880	69.2[a]	76.4; 34.5[a]	55.0[a]	43.4[a]	4703	96.1; 40.3[a]	72.2	210	101; 41.2[a]
	Average temperature, °C		18.5	19.8	19.2; 20.4[a]	24.8	18.8	22.5	21.4	24.3	21.1	18.7
	Floor		Attic	Basement	Basement	Basement	Basement	Basement	First	Ground	Ground	Ground
	Function		Technical	Laboratory	Technical	Archive	Store	Radon room	Office	Kitchen	Library	Bedroom
	Occupancy		No	Yes	Rarely	No	No	Rarely	Yes	Yes	Yes	No
	Size, m		4 × 3.5 × h3	12 × 10 × h4	12 × 17 × h(2 ÷ 3.7)	6 × 2.5 × h3.2	5 × 4.1 × h3.1	3 × 3 × h2.4	8 × 5.6 × h3.2	3 × 5 × h4	3 × 4 × h3	4 × 3.6 × h2.4
	Heating		Yes	Yes	No	Yes	No	No	Yes	Yes	Yes	Yes
	Conditioning		No	Yes	No	No	No	No	No	No	No	No
	Number of	windows	0	0	0	0	0	0	1	2	2	3
		doors	1	1	1	1	1	1	1	1	1	1
		external walls	1	0	3	1	1	0	1	2	2	2
Building	Location		Moscow region, Mendeleevo		Moscow			Moscow region, Zelenograd		Moscow region, Ljalovo		Ryazan region
	Function		Office	Office	Office		Office	Office		Residential		Residential
	Year built		1960	1970	1980		1970	1975		2003		1926
	Storeys number		3	6	9		4	2		3		1
	Building area, m^2		1150 (big)	1900 (big)	1200 (big)		630 (middle)	2000 (big)		100 (small)		56 (small)
	Asphalt pavement [b]		No	No	Yes		Yes	Yes		No		No
	Foundation type		Slab	Slab	Slab/Band		Slab	Slab		Slab		Band
	Ventilation		Natural	Natural	Natural		Natural	Natural		Natural		Natural
	Central heating		Yes	Yes	Yes		Yes	Yes		Yes		No
	Central conditioning		No	No	No		No	No		No		No
	Material of	walls	Brick	Concrete	Concrete		Concrete	Brick		Brick		Brick
		floors	Concrete	Concrete	Concrete		Concrete	Concrete		Concrete		Wood
Geology	Territory relief		Smooth		Smooth		Smooth	Smooth				Smooth
	Soil type		Clay		Sandy loam		Clay	Clay				Sand and loam
	Permeability		Low		Middle		Low	Low				Above-middle
	Groundwater depth, m		4		20		4	4				7
	Soil freezing depth,m		1.2		1.0		1.0	1.2				1.4
Climate	Temp.,°C	Warmest month	+17.8		+19.2			+17.8				+19.5
		Annual average	+6.0		+7.1			+6.0				+4.6
		Coldest month	–9.8		–6.7			– 9.8				–11.5
	Average wind speed, m/s		2.3									2.0
	Rainfall, mm		700									500
	Clear days per year		82									130

[a] Monitoring of EEC.
[b] The asphalt or other poorly permeable coating around the building.

is a large basement room with a soil floor as a natural radon source. Besides, these experimental rooms are located in different buildings, the distance between which is about 40 km. Note that in such very different rooms, a similar and regular behavior of radon is observed; the reasons for that will be discussed in detail in one of our next publications.

The monitoring revealed that the experimental room ER 5, like the building itself, was in an emergency condition, because a groundwater flooded the basement during the frequent rains and melting of snow. The water level of several centimeters in the basement was maintained for several months.

It can be seen from Table 1, that the equilibrium equivalent concentrations (EEC) of radon progeny were monitored in the experimental rooms ER 2, 3, 4, 5, 7 and 10. The measurements of EEC, which is used for limitation of the AAIR in Russia (IAEA, 2017), were conducted either in parallel with the measurements of radon activity concentration, or instead of them.

5. Measurement devices

Measurements of radon activity concentration were performed using well-known radon monitors “AlphaGUARD” (Genitron Instruments GmbH, Germany) with a recording period of 1 h and “RadonSCOUT” (Sarad, Germany) with a recording period of 3 h.

For the EEC monitoring with a recording period of 3 h the Radon Aerosol Alpha Radiometer RAA-3-01 “AlphaAERO”, developed in STC “Amplituda” (Tsapalov, 2008), was used. Measurements and calculation of EEC in a monitor mode were performed automatically by 3 h periodical sampling of the air by its pumping through on the aerosol filter for 10 min at a rate of 8 L per minute. Weekly, the filter was changing, and detector been checking by using the reference point alpha-source included to the kit. The measurements of the current activity of the filter were performed in the alpha-spectrometry mode taking into account the residual activity after the previous period of measurements. The radiometer “AlphaAERO” successfully passed the metrological and climatic tests in Russia and has been manufactured since 2008.

There is no need to provide the characteristics of radon (and EEC) devices accuracy, since this is not required when processing the results of annual monitoring, according to the algorithm described above (Section 3). By the way, this information is available on the websites of manufacturing companies. However, we would like to clarify that in any case the statistical component of the uncertainty of $C(t)$ did not exceed 3%, since the minimum integration period in the analysis of the initial data was 24 h, especially taking into account that the measurements were carried out at an increased radon concentrations in experimental rooms. Moreover, accounting the systematic component of uncertainty did not make sense, because the coefficient $K_V(t)$ is dimensionless, according to Equations (5)–(7). The most important requirement for monitor-devices is the stability of the basic metrological parameters, which is provided by relatively stable microclimate conditions in the experimental rooms. The accumulated data was downloaded weekly. All the devices operated reliably.

6. Results of annual monitoring

The results of annual monitoring in the experimental rooms with a recording period of 3 h are shown in Fig. 2(a–c). These figures confirm that all experimental rooms had elevated radon concentrations. It can be seen that in most cases, the level of indoor radon (or EEC) in summer exceeded that in winter. Even in the absence of people in the permanently closed rooms (ER 1, 3, 4, 10), there were almost as significant temporary variations of radon (or EEC) as in rooms (ER 2, 7, 8, 9) that were occupied and were operated without restrictions. The frequency distributions of radon activity concentrations or EEC did not have well-defined patterns. The temporal variations of indoor radon (and EEC) over one year were quite different. However, despite these differences, in all cases (except for EP 5) a similar characteristic dependence of the coefficient $K_V(t)$ on the mode and duration of measurements was observed (Figs. 4 and 5). We assume that the set of experimental rooms is quite representative and similar temporal radon variability will be also observed in other buildings (with a high level of radon) located on territories with other geological and climatic conditions.

In addition, considering that in the experimental rooms ER 7, 10 the concentration of radon and EEC were monitored simultaneously, Fig. 3 shows the temporal variations of the equilibrium factor (ratio of the EEC to radon concentration) in these two rooms.

7. Estimation of temporal variations of radon and EEC

The estimation of the temporal variations of radon and EEC are shown in Fig. 4 (a,b) for continuous measurement of up to one year, and also in Fig. 5 (a,b) for the case of two or four measurements with an interval of 6 or 3 months, respectively, except for the experimental rooms ER 5. For this room, the values of the temporal EEC variation appeared incomparably high due to the flooding in the building basement (see Section 4). Nevertheless, this experience is also useful in the interpretation of the results, and will be discussed in Section 8.

The implementation of the criteria (1) and (2) is possible only on the basis of experimentally derived tabulated values of the coefficient of temporal radon variation $K_V(t)$, at a confidence level of at least 95%. Table 2 shows the experimentally determined tabulated values of the coefficient of temporal radon variation in premises with natural ventilation. However, it must be borne in mind that the tabulated values of the coefficient $K_V(t)$ were obtained on a relatively limited experimental data, so they must be verified, and clarified. This aspect is discussed in more detail in the next sections.

8. Discussion of results

The information about annual monitoring of the equilibrium factor in two different rooms (Fig. 3), most likely, is demonstrated for the first time. The temporal variations of measured equilibrium factor are extremely large, including weekly and seasonal trends, so it seems that short-term or long-term measurements of the equilibrium factor make no sense. These measurements improve neither the understanding of indoor radon behavior, nor the accuracy of the dose estimation. For a more reliable estimation of radon doses, a fixed value of the equilibrium factor of 0.4 or 0.5 (with a narrow margin) is recommended. However, this does not apply to rooms with powerful mechanical ventilation, where the equilibrium factor is significantly reduced and $K_V(t) \rightarrow 0$.

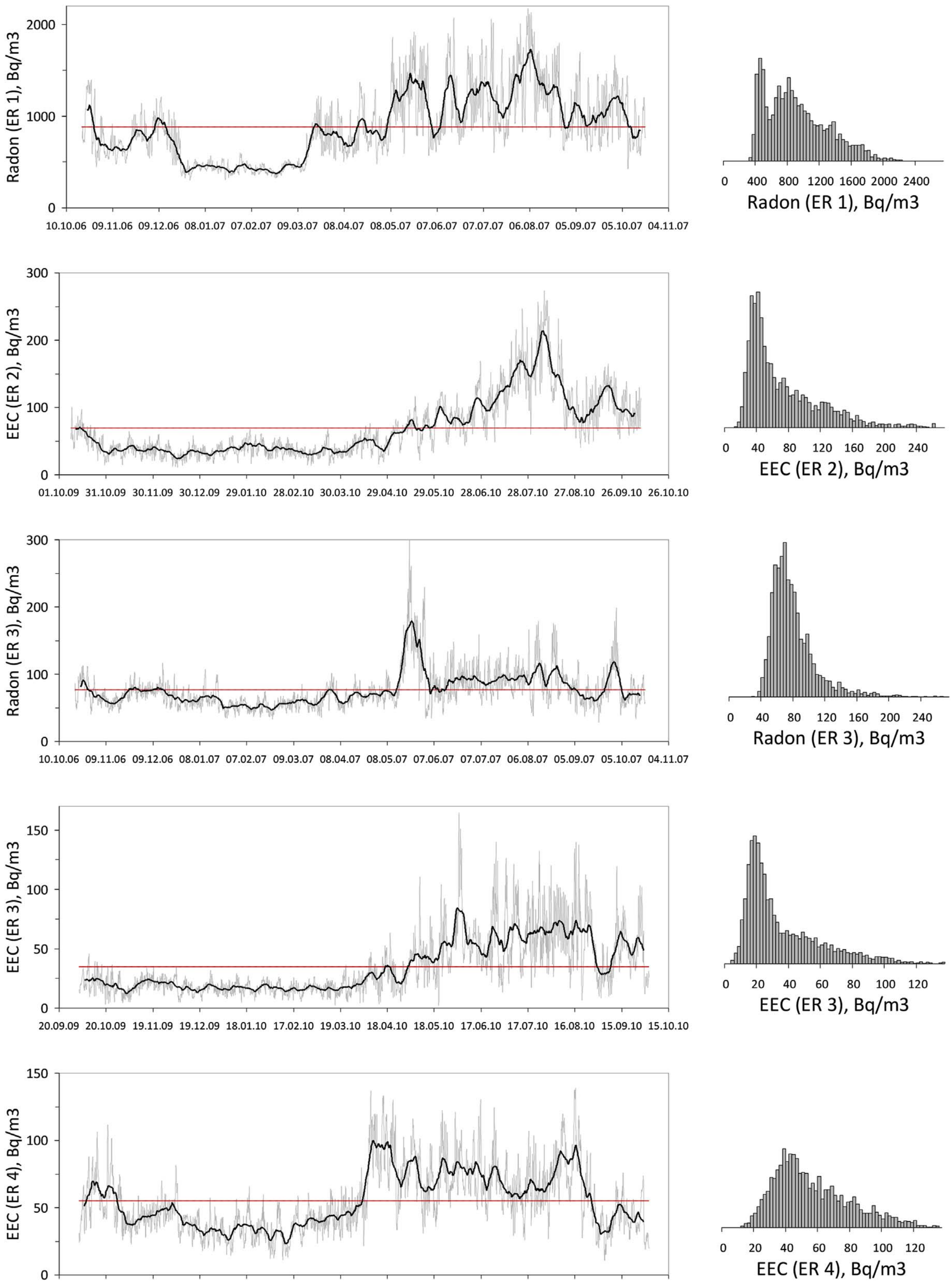
Analysis of the dependencies of the coefficient of temporal radon (or EEC) variation on the mode and duration of measurements (Table 2), leads us to the following conclusions.

The temporal variation of EEC is usually higher than the variation of radon due to the effect on EEC of an additional factor, such as precipitation of radon progeny on any surface in the room, including occupants. Another additional factor is the loss of radon progeny from the room due to ventilation.

The values of the coefficients $K_V(t)$ for radon and EEC are equal, if four measurements in different seasons are conducted.

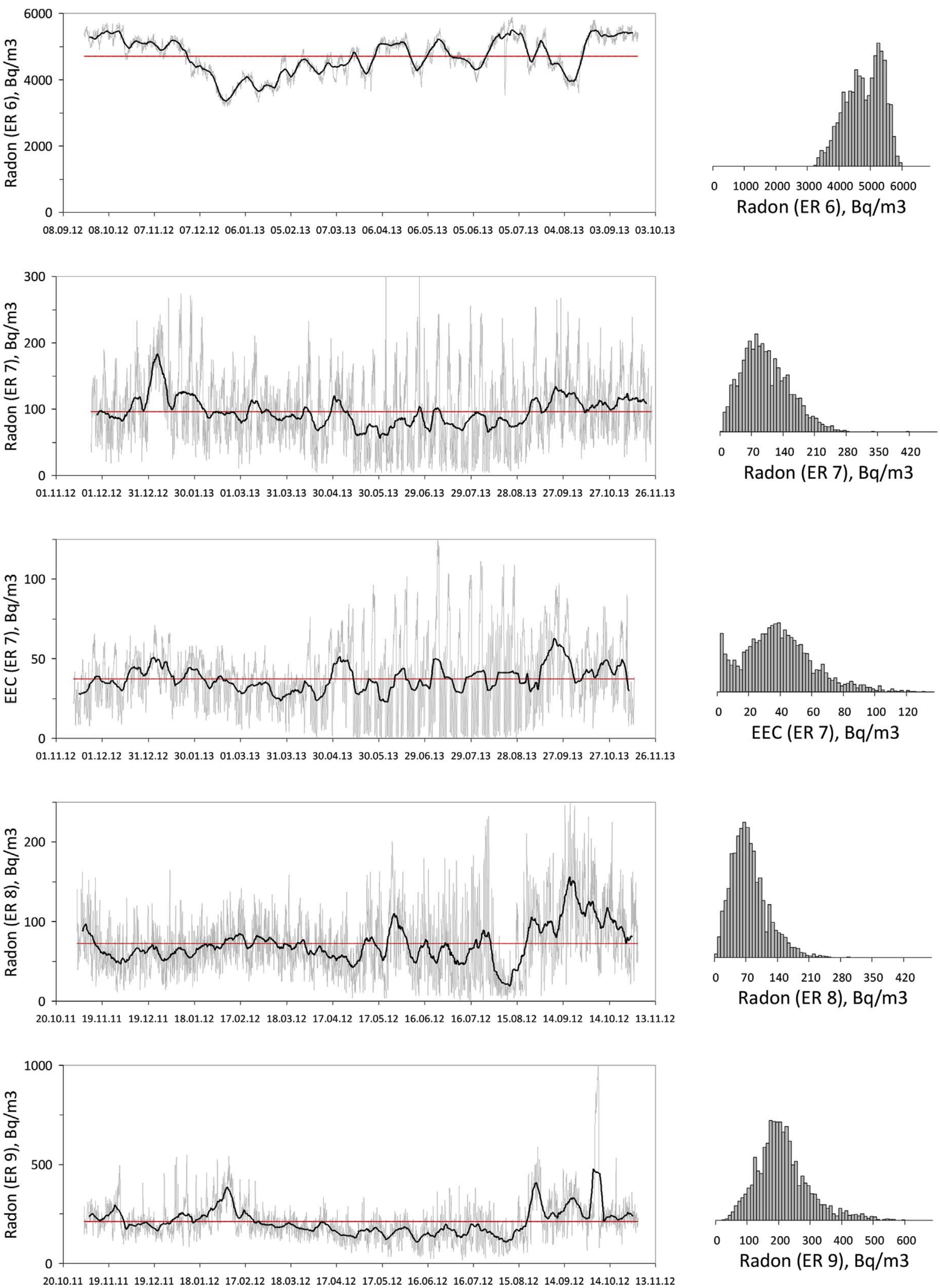
A significant, almost two-fold decrease of the coefficient $K_V(t)$ is achieved by taking into account the temperature influence, according to Fig. 6. However, according to (MG 2.6.1.037, 2015), this decrease is observed only in the short-term test and only for the closed rooms, while also taking into account the special climatic factor. Thus, due to serious limitations, the application of temperature accounting is significantly hampered.

Probably, the tabulated values of the coefficient of temporal radon variation for the continuous measurements in the period from 1 to 8 months are overestimated (Fig. 4(a) and Table 2), since the values of $K_V(t)$ for the experimental room ER 1 during this period were significantly higher than in other experimental rooms, according to



(a)

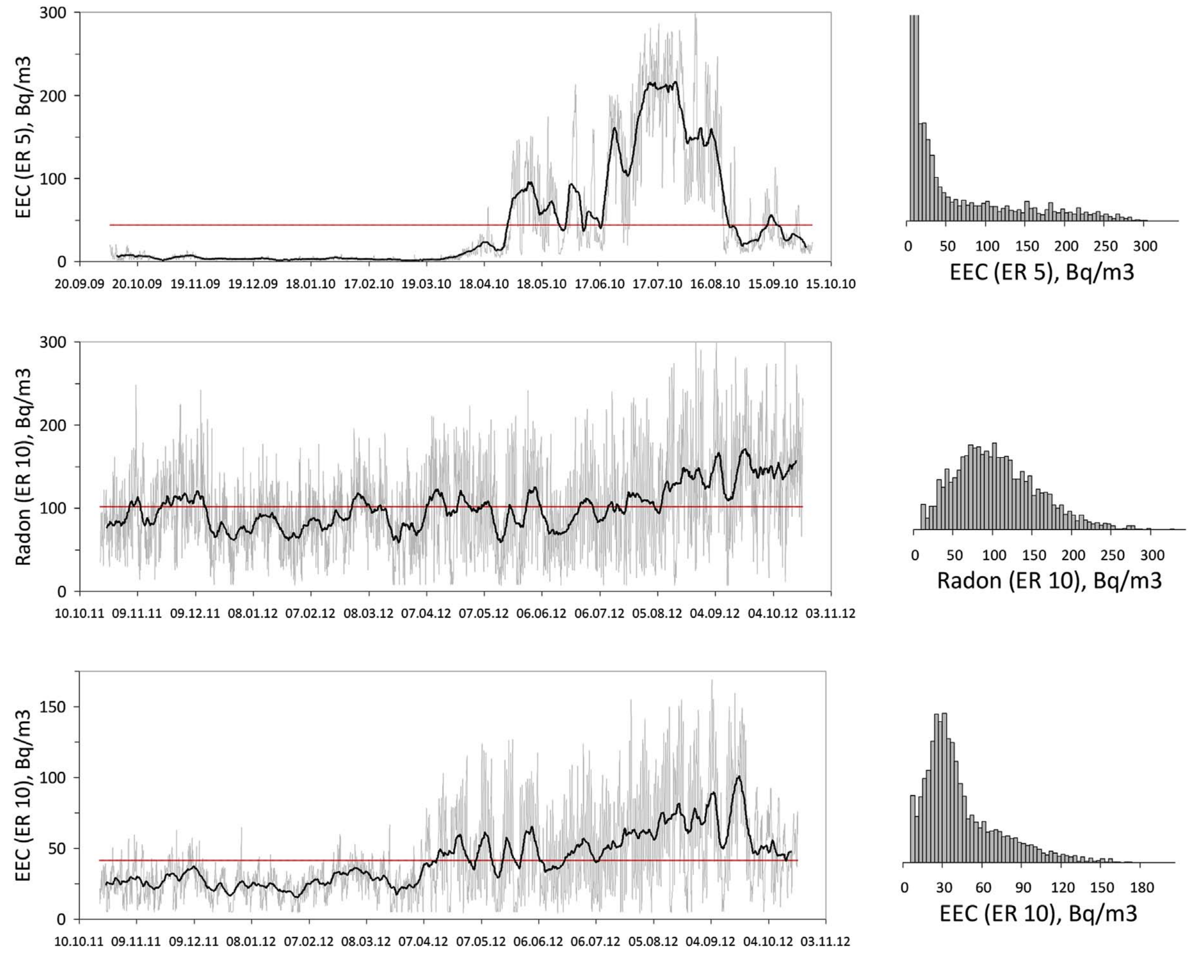
Fig. 2. The results of annual monitoring of the radon activity concentration and EEC in the experimental rooms ER 1–4 (a), ER 6–9 (b) and ER 5, 10 (c), the weekly trends (thick lines), AAIR levels (thin lines) and the patterns of the measurement results' distributions.



(b)

Fig. 2. (*continued*)

Fig. 4(a). This difference can be explained by the fact that the reduction in radon concentration in the experimental room ER 1 (heated and closed room in the attic with a stable radon source) due to higher air exchange during the cold period is not compensated by the increased exhalation of soil radon into the building. Nevertheless, we prefer a conservative approach, because the experimental data, which could



(c)

Fig. 2. (*continued*)

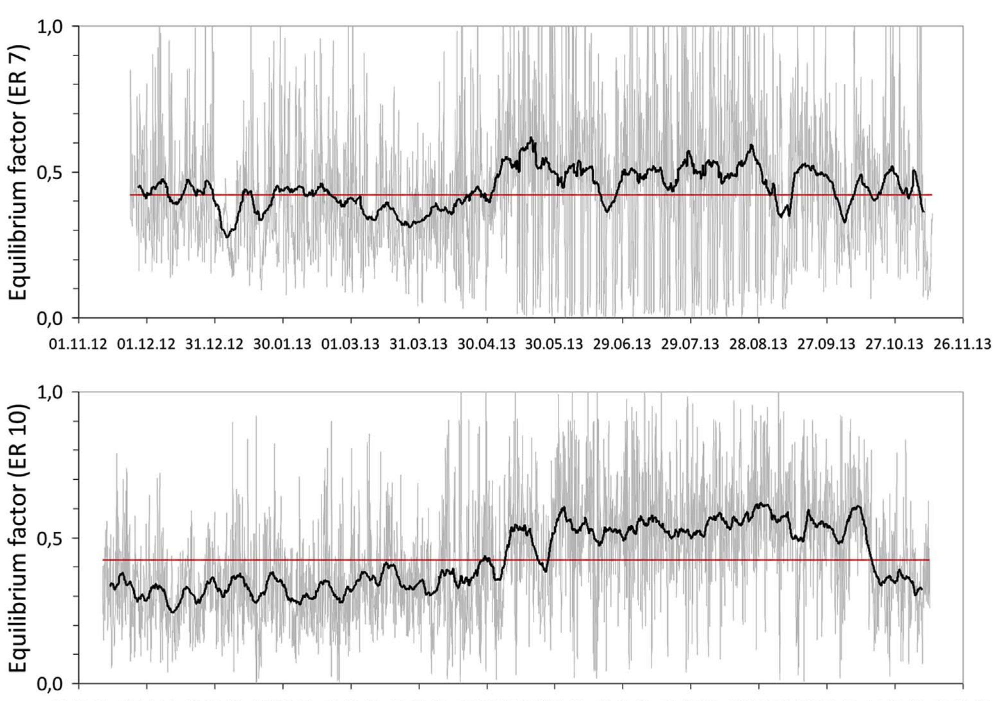
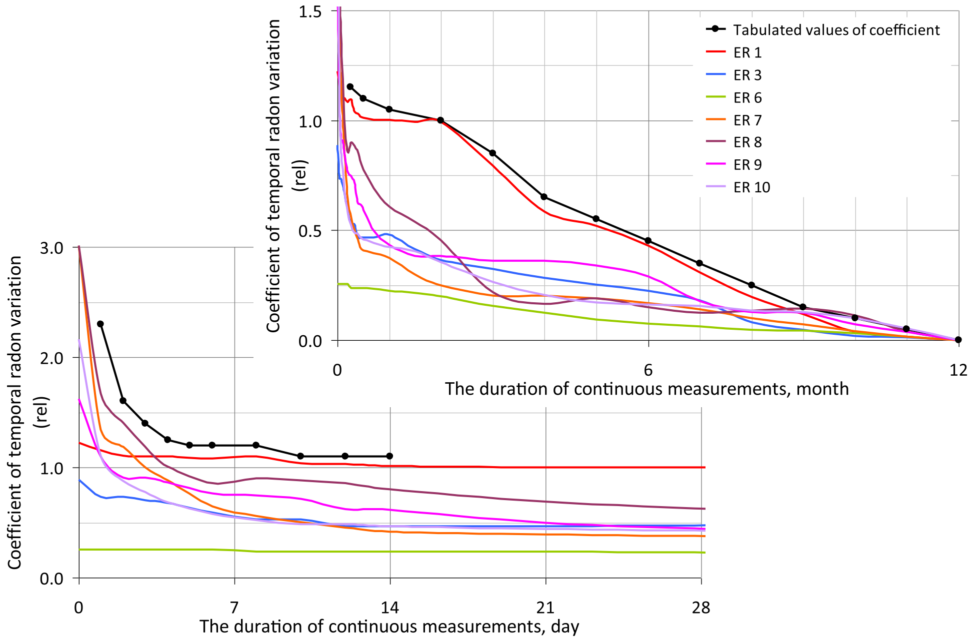
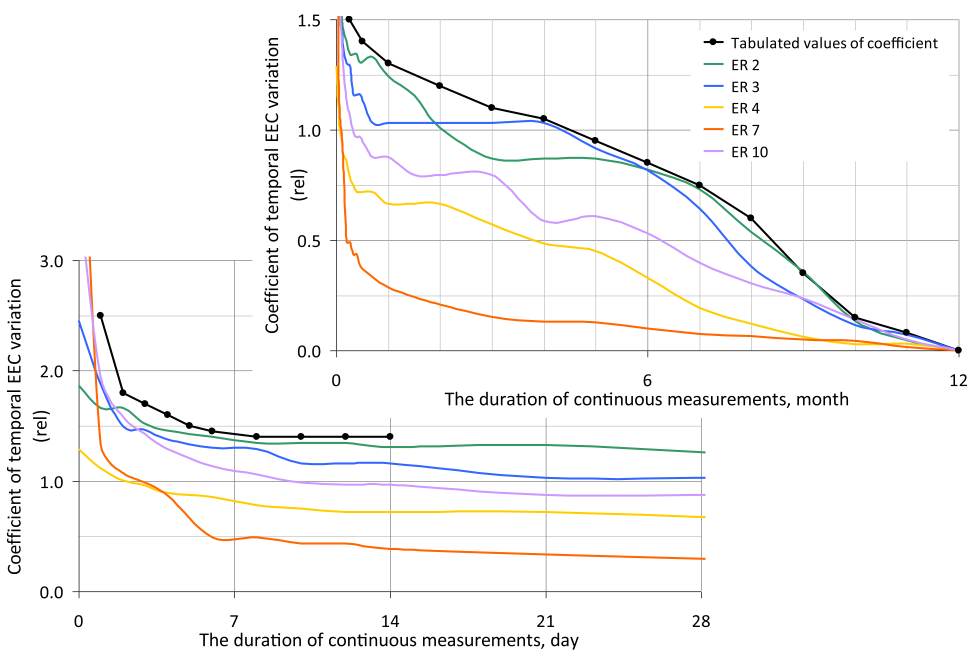


Fig. 3. Temporal variations of equilibrium factor in two experimental rooms (ER 7, 10), the weekly trends (thick lines) and annual average values (thin lines).



(a)



(b)

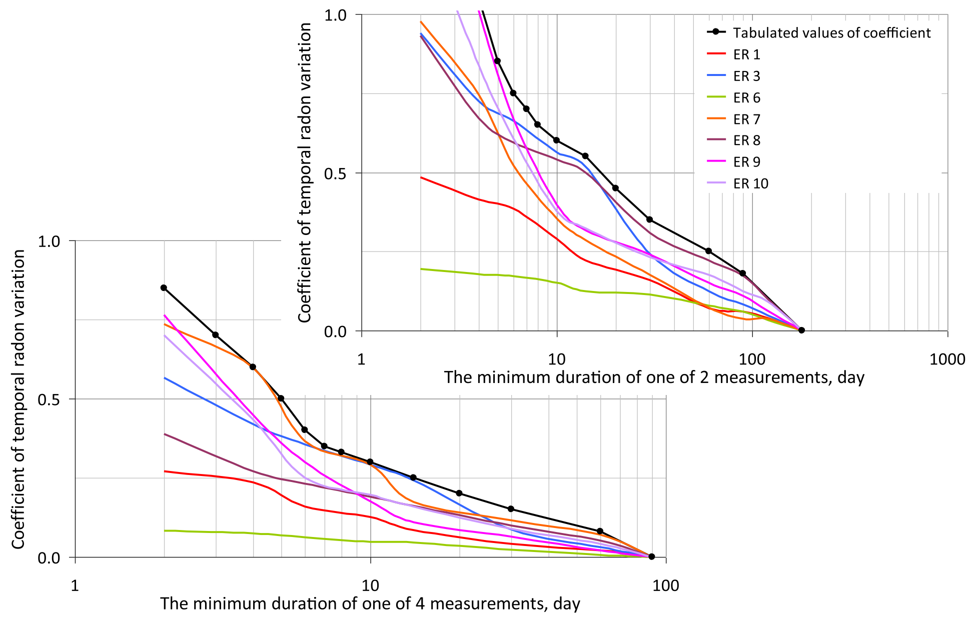
Fig. 4. Dependence of the coefficients of temporal variations of radon (a) and EEC (b) on the durations of continuous measurements by the results of annual monitoring in the experimental rooms.

confirm this assumption, are limited.

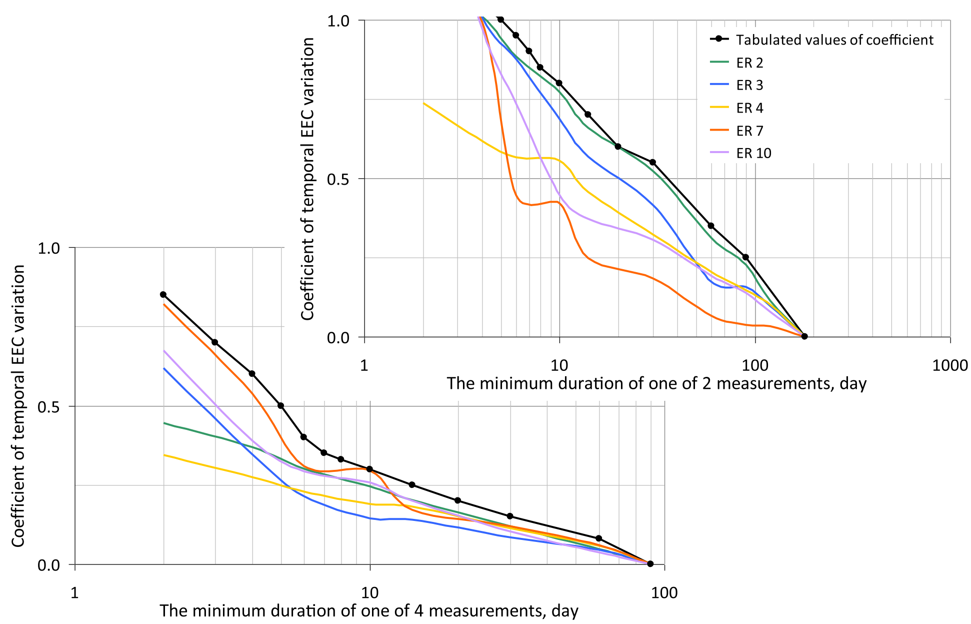
The rapid decline of $K_V(t)$ mainly occurs within first 7–10 days of continuous measurements of radon or EEC, as shown in Fig. 4 (a, b) and 6. Then, during the next few weeks and about 2 months of continuous measurements, the values of $K_V(t)$ decrease very little, for example, in range from 1.1 (or 110%) to 1.0 (or 100%) for radon. Therefore, it is advisable to take a time interval of not more than 10 days as the duration of short-term measurements, and a time interval from 10 days to 2 months as the duration of middle-term measurements. Moreover, after 10 days of measurement, the values of $K_V(t)$ showed little dependence on the state of the room (closed or normal).

It is important to note that in the specified period of short-term measurements, the values of $K_V(t)$ remain high enough, according to Fig. 6, i.e. the condition $K_V(t)^2 >> U_D^2$ is satisfied. This means that in the short-term measurement period, it makes no sense to measure indoor radon activity concentration or EEC with high accuracy (as suggested in (ANSI/AARST MAH, 2014) by conducting the parallel measurements of radon concentration at two closely spaced points).

The long-term measurements (or sampling) from 2 months to 1 year were not effective due to a slow decrease of the coefficient $K_V(t)$ over time, according to Table 2. Therefore, it is advisable to perform several measurements (2 or 4) in different seasons of the year. For example, reducing the values of $K_V(t)$ to the level of about 0.25 (or 25%) can be achieved by two or four measurements lasting 2 months or 2 weeks,



(a)



(b)

Fig. 5. Dependence of the coefficients of temporal variations of radon (a) and EEC (b) on the minimum duration of one of 2 (top) and 4 (bottom) measurements with an interval of 6 or 3 months, respectively, by the results of annual monitoring in the experimental rooms.

respectively. The same value ($K_V(t) = 0.25$) is achieved with a continuous measurement duration of about 8 months. Increasing the duration and number of the measurements in order to obtain values of $K_V(t)$ lower then 0.25 (i.e. achieve more accurate estimation of the AAIR level) is inappropriate - due to the existence of year-to-year variations of the AAIR level itself. On average, the amplitude of such variations is approximately estimated in the range of 14% (Bochicchio et al., 2009) to 26% (Steck, 2009) with a maximum year-to-year variation of about 40% (Hunter et al., 2005; Lubin et al., 2005) and even reaching 50% (Darby et al., 1998).

In our opinion, significant year-to-year variations of the AAIR may be due to the development of new defects (cracks, discontinuities, delaminations etc.) in building foundations and due to the geological factor - namely, increased seismic or local geodynamic activity of the territory. Obviously, these geological processes have a significant effect on the dynamics of radon migration in soil, enhancing the fluctuations of radon exhalation into the atmosphere, as well as the variations of radon exhalation from soil and its entry into the building. Besides, such geological processes, compared to natural air exchange in buildings, are characterized by extreme temporary instability in long-term intervals (half a year, a year and more). Such geological processes and degradation of the insulation of the building foundation are usually hidden, that creates significant problems for reliable estimation of the AAIR. Therefore, in the territory with the signs of unstable geological conditions, when conducting the indoor radon control, at least 2 long-term measurements throughout the year are probably needed. Note that with respect to Table 2, the additional 2 measurements over the next year, but performed in other seasons, can be considered, taking into account 2 measurements in the previous year, as 4 measurements in different seasons of one year.

Table 2
Tabulated values of the coefficients of temporal radon (and EEC) variation $K_V(t)$ depending on the mode and duration of measurements.

The minimum measurement duration		One continuous measurement	Two measurements with the interval of 6 months (± 1 months)	Four measurements with the interval of 3 months (± 2 weeks)
Day	1	2.30 (2.50)	–	–
	2	1.60 (1.80)	1.60 (1.60)	0.85 (0.85)
	3	1.40 (1.70)	1.30 (1.30)	0.70 (0.70)
	4	1.25 (1.60)	1.05 (1.05)	0.60 (0.60)
	5	1.20 (1.50)	0.85 (1.00)	0.50 (0.50)
	6	1.20 (1.45)	0.75 (0.95)	0.40 (0.40)
	7	1.20 (1.45)	0.70 (0.90)	0.35 (0.35)
	8	1.20 (1.40)	0.65 (0.85)	0.33 (0.33)
	10	1.10 (1.40)	0.60 (0.80)	0.30 (0.30)
	12	1.10 (1.40)	0.58 (0.75)	0.28 (0.28)
	14	1.10 (1.40)	0.55 (0.70)	0.25 (0.25)
	20	1.10 (1.40)	0.45 (0.60)	0.20 (0.20)
Month	1	1.05 (1.30)	0.35 (0.55)	0.15 (0.15)
	2	1.00 (1.20)	0.25 (0.35)	0.08 (0.08)
	3	0.85 (1.10)	0.18 (0.25)	0
	4	0.65 (1.05)	0.12 (0.18)	–
	5	0.55 (0.95)	0.06 (0.08)	–
	6	0.45 (0.85)	0	–
	7	0.35 (0.75)	–	–
	8	0.25 (0.60)	–	–
	9	0.15 (0.35)	–	–
	10	0.10 (0.15)	–	–
	11	0.05 (0.08)	–	–
	12	0	–	–

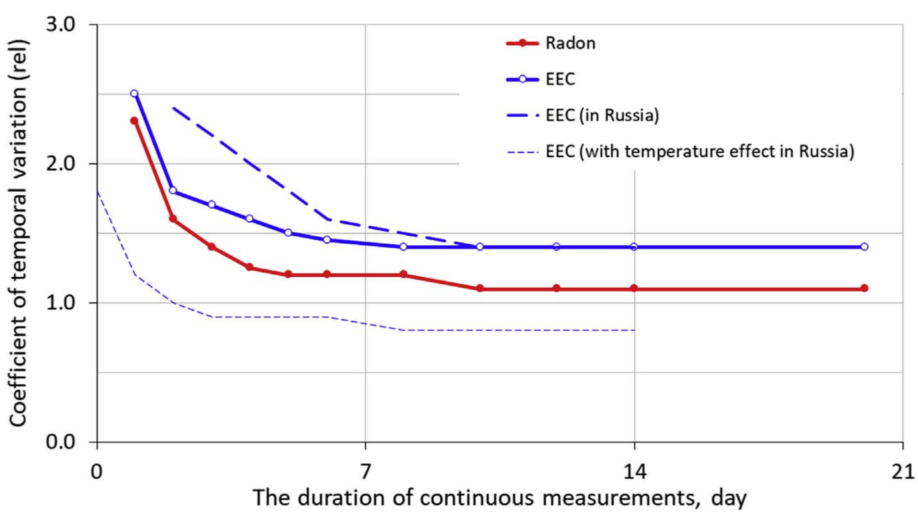


Fig. 6. A comparison of the values of the coefficients of temporal variation of radon and EEC depending on the duration of continuous measurements and the temperature effect; the data for Russia are taken from (MG 2.6.1.037, 2015).

At the same time, any detectable and visually recognizable factors affecting the behavior of indoor radon should be taken into account. The most important of these factors is the hydrogeological state of the soil at the base of the building. If the soil underlying the building is moisture-saturated, especially when water is found inside the basement, then the soil radon transport into the building is absent, as seen in Fig. 2(c) for the experimental room ER 5, which is located in the periodically flooded basement. In this room, after lowering the groundwater, the EEC gradually increases to exceed the reference level due to the increased exhalation of soil radon by intensive evaporation of moisture from the emanating soil. In such cases, conducting the indoor radon control does not make any sense, because part or even the entire building is in an emergency condition.

9. Representativity and verification of the results

Given the above considerations, we consider the data in Table 2, as well as the new principle and the criteria formulated in Section 2, as the first step to develop a universal strategy for indoor radon regulation and the most effective measurement protocol, based on a scientific approach. This means that both the most suitable (optimal) and reliable measurement protocol can be created by quantifying such important factors as: (a) the level of potential radon hazard of the territory, (b) the planned number of survey buildings in this area, (c) time and financial resources, (d) available radon devices, regardless of the measurement principle, etc. This is especially true in the case of large scale surveys conducted to identify buildings with a high radon concentration, exceeding the reference level. The development of the principles of such a universal strategy and the analysis of the effectiveness of a given measurement protocol is a separate scientific topic, the results of which will be published in our next articles.

An important problem for discussion is the representativity of the data showed in Table 2. The results of annual monitoring (Fig. 2) show a diverse behavior of indoor radon. These data were obtained in ten experimental rooms located in seven buildings, within the same geographical and climatic region. At the first sight, such number of experimental rooms in our study does not seem enough representative - in comparison, for example, with the large radon surveys, such as (Steck

Table 3
Comparison of coefficients of variation (COV) according to our results with the results of (Steck et al., 2004) depending on the duration of measurements.

Duration of continuous measurements	The average value of COV over 7 experimental rooms (ER 1,3,6–10), where annual radon monitoring was conducted (except for EP 6)[a]	According to Steck et al. (2004) (state of experimental rooms)
2 days	0.66 (0.73)	0.76 (closed)
4 days	0.59 (0.65)	0.70 (closed)
1 months	0.44 (0.48)	0.40 (normal)
3 months	0.31 (0.34)	0.30 (normal)
4 months	0.27 (0.29)	0.25 (normal)
6 months	0.18 (0.19)	0.17 (normal)

[a] The experimental room of EP 6 has a very low (not typical) air exchange (see Section 4).

et al., 2004) - conducted in 62 buildings, (Kozak et al., 2011) - in 132 buildings or (Ruano-Ravina et al., 2008) - in 391 buildings. However, Table 3 indicates a quite satisfactory convergence of our results with the results of (Steck et al., 2004), if the basic parameter for comparison is COV. In this case, the representativity of our data can be considered quite satisfactory. At the same time, this fact does not cancel a need to verify the results of the current study. It is important to clarify that Table 3 shows the values of COV only for analyzing the representativity of the results of our study, but not for the practical use (see the explanations in Sections 1 and 3).

Probably, separately the experimental rooms look not quite representative (5 basements of 10 rooms), but the set of ER as a whole is quite representative, since they have a wide coverage according to the following characteristics by Table 1: the range of AAIR is almost two orders of magnitude; source of radon (only soil, soil and building materials or only building materials due to simulation by an artificial source); the presence of windows and external walls (available in different numbers or absent); the function (technical, office and residential premises, manufacturing facilities and dwellings); ventilation conditions (closed or normal); floor (from basement to attic) and number of storeys (from 1 to 9); building materials (wood, brick or concrete); size of the rooms (from 3x3xh2.4 to 12x17xh3 m), the area of buildings (from 56 to 2000 m^2) and foundation type (slab or band-type); year built (from 1926 to 2003). Thus, the qualitative features, despite a relatively small number of experimental rooms, indicate the satisfactory representativeness of their set, but within a single region with almost homogeneous geology and climate.

The data of Table 2 have to be verified on the basis of annual continuous monitoring of radon activity concentrations in experimental rooms of other buildings located in the regions with a climatic and geological conditions that differ from the temperate climatic zone and, in particular, the Moscow region (Russia).

In our opinion, one of the most suitable countries for such verification may be Israel. The landscape and geology of Israel is mainly a mountain relief, fractured rocks and permeable soil, which differs significantly from the geology of the Moscow region, as well as the climatic conditions. Israel is located in a subtropical climate zone with the difference between the seasonal outdoor temperature averages about (12–15) °C. For comparison, in the Moscow region this difference is much higher - about (22–26) °C. In contrast to the seasonal variations, the differences in diurnal temperatures (5–6 °C in Moscow and 7–8 °C in Haifa) are quite small.

Indoor radon behavior strongly depends on the air exchange (mainly determined by the outdoor temperature and other weather parameters) and the soil conditions (ICRP, 2014). Because of the significantly lower seasonal variation of outdoor temperatures, the lower values of $K_V(t)$ for Israel, than those given in Table 2, are expected. However, given the less stable geological conditions in this country, the resulting difference between the coefficients $K_V(t)$ may not be significant. **If this assumption will be experimentally proven**, the data of Table 2 could be confidently recommended for indoor radon regulation in buildings throughout European and North American countries, where the climate is usually warmer than in Moscow region, but, at the same time, colder than in Israel.

In order to verify and clarify the data reported in Table 2, we would like to encourage the colleagues who conduct radon surveys in different countries of the world to send the results of annual monitoring of indoor radon (with a registration period of 1–6 h) to the authors in the form of electronic tables, taking into account the requirements for the selection of experimental rooms, according to Section 4, together with detailed description of the experimental rooms in accordance with Table 1. This information will help to classify the premises and to develop a universal and reliable strategy for indoor radon regulation, taking into account the influence of various factors on the behavior of indoor radon.

10. Conclusions

The annual continuous monitoring of indoor radon activity concentration and EEC radon progeny was conducted in 10 selected experimental rooms with a high content of radon and natural ventilation, located in 7 buildings, mainly in the Moscow region (Russia). The processing of the results of this monitoring using the original algorithm proposed in Section 3 allowed the following results.

(a) The tabulated values of the coefficients of temporal radon and EEC variation were determined, depending on the mode and duration of measurements, including 2 or 4 measurements in different seasons of the year (Table 2).
(b) Values of $K_V(t)$ can reliably estimate the uncertainty of the AAIR and allow a correct comparison with the reference level based on the new principle of indoor radon evaluation, which consists of three criteria presented in Section 2.
(c) The temporal variations of EEC radon progeny are usually higher than the variations of radon activity concentration - on average by (20–60)%. Therefore, EEC measurements, like measurements of the equilibrium factor (see Section 8), do not contribute much to improving the reliability of indoor radon regulation, with the exception of rooms with powerful mechanical ventilation.
(d) The maximum duration of short-term measurements (no more than 10 days after start) is justified, because exactly in this period, a rapid decrease of the coefficient $K_V(t)$ is observed.
(e) The new principle of indoor radon regulation and the experimentally determined tabulated values of the coefficient $K_V(t)$ make it possible to develop a universal strategy for indoor radon evaluation and selection of the most optimal measurement protocol. However, in some cases the reliability of the result of the indoor radon evaluation can be reduced by the influence of hidden geological processes and degradation of the insulation of the building foundation.
(f) Despite the limited number of experimental rooms, the experimental results are quite representative. Nevertheless, the obtained tabulated values of $K_V(t)$ should be subject to verification and, if necessary, be clarified on the basis of further accumulation of experimental data, as recommended in Section 9.

Acknowledgements

This study was supported by STC "Amplituda" (Russia) and the Center of Radiation and Chemical Safety at the Federal Medical and Biological Agency of Russia. The authors are especially grateful for the help of Prof. A. Marennyy, Dr. A. Gubin, Dr. A. Ermilov, Dr. O. Tutelyan, S. Kuvshinnikov, U. Samohvalov and A. Yankin.

The results of the study were presented in the Final Symposium of the European concerted action COST TU1301 “NORM4Building” in Rome, Istituto Superiore di Sanità (National Institute of Health), 6–8

June, 2017, and the authors are thankful to the members of COST TU1301 NORM4BUILDING for the fruitful discussions and support.

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