ESTIMATION OF RADIOLOGICAL IMPACT ON RESIDENTS DUE TO HOUSEHOLD STORAGE OF COAL USED FOR HEATING IN SERBIA

by

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This paper aims to estimate a potential radiological risk from different kinds of coals used for domestic heating in Serbia, by measuring the activity concentration of radionuclides and radon exhalation rate. The obtained radon mass exhalation rate ranges from (5.3 3.1) $Bqkg^{-1}s^{-1}$ to (70.3 9.4) $Bqkg^{-1}s^{-1}$ and was highest for lignite type of coal. It is estimated that coal stored in the basement could contribute up to 50 Bqm^{-3} of indoor radon concentration at the ground level. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb in analysed coal samples agree with previously reported concentrations of coal used in Serbia. The values of radium equivalent concentration and external hazard index indicate that the used coal does not represent a significant radiation hazard.

Key words: radon exhalation rate, gamma spectrometry, radiation hazard index, coal

INTRODUCTION

One of the important steps to reduce or prevent dangerous climate change is to reduce the emission of greenhouse gases (GHG), especially CO_2. In order to follow the line of the Paris Climate Agreement, in December 2020 the European Council committed to reduce the emission of greenhouse gasses by 2030, to at least 55 % of the level that was in 1990 [1]. According to the energy balance for the 2020 year, the largest consumption of coal in Serbia, namely 96 % is for the production of electrical and thermal energy, while 43 % of the remaining coal consumption goes for households *i. e.*, for domestic heating [2]. There are still regions, especially in less developed, rural communities where usage of coal for home heating is considerable.

Besides the effect of GHG emission, the burning of coal can have a significant radiological impact as well. It is known that different types of coal can have different concentrations of primordial radioactive elements. A considerable variation of up to several orders of magnitudes for activity concentration of radionuclides from uranium and thorium series were found [3]. The concentration of these elements in fly ash can be an order of magnitude higher than their concentration in coal. Therefore, activity concentration of these isotopes is monitored on regular basis in coal and different types of coal combustion wastes [4].

A high concentration of ^{238}U and subsequently ^{226}Ra in coal could lead to a high exhalation rate of radon (^{222}Rn), which is known as the second cause of cancer after smoking [5]. The main contributors to indoor radon concentrations are soil beneath the building and building materials. However, it is not uncommon, especially in rural communities, to store coal necessary for the whole heating season, in the cellar of the dwelling. It is assumed that a high amount of coal in a cellar could produce a non-negligible radon concentration that can migrate to a residential area.

Therefore, this paper aims at providing insight into the radiological impact both due to the radon exhalation rate and external gamma dose rate of different types of coal commonly used in Serbia for domestic heating.

MATERIALS AND METHODS

The activity concentration of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb and radon exhalation rate of different types of coal were measured. According to the

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Table 1. Radon mass exhalation rates, for different coal samples and their contribution to indoor radon concentration

Coal sample ID	Type of coal and its origin	m [kg]	T [h]	E_m [Bqkg^{-1}s^{-1}]	C [Bqm^{-3}]	
					λ_v = 0.63 [h^{-1}]	λ_v = 0.2 [h^{-1}]
1	Hard coal, Russian mine	1.04	161	5.3 3.1	2.5	8.0
2	Brown coal, Russian mine	1.15	190	5.7 4.4	2.7	8.5
3	Brown-lignite, Pljevlja mine	1.27	204	41.4 6.2	20	62
4	Brown coal, Miljevina mine	1.25	188	47.5 3.6	23	71
5	Lignite, Kovin mine	0.91	278	70.3 9.4	33	105
6	Brown coal; Breza mine	1.23	254	36.4 5.3	17	55
7	Lignite, Kolubara mine	1.43	428	65.0 8.4	31	98
8	Hard coal, Banovići mine	1.48	282	33.6 3.9	16	50

classification of coal by its carbon content [6], the following types were selected: lignite with the least carbon content, followed by, brown lignite, brown coal, and hard coal with the most carbon content (excluding anthracite not commonly used is residential heating). In total, eight samples of coal available in markets in Serbia for domestic heating were analysed. Coal samples differ either by their type or by the location of mines they were excavated from, as indicated in tab. 1.

Radon exhalation rate measurements

Radon exhalation rate was measured using the close-chamber method with the RTM1688-2 device of SARAD GmbH [7, 8]. Samples were deployed in a 30 L accumulation chamber for a period of between one and two weeks, tab. 1. The radon concentration in the chamber was then either continuously measured for the whole deployment period, measuring a radon build-up in the chamber, or at the end of the deployment period measuring, therefore a maximal radon concentration. In both cases, the sampling time was one hour. Radon exhalation rate was extracted from a build-up curve by using the following equation [8]

$$C(t) \quad \frac{E_{\mathrm{m}} m}{V\lambda_{\mathrm{eff}}}(1 \quad \mathrm{e}^{\;\lambda_{\mathrm{eff}} t}) \quad C_0 \mathrm{e}^{\;\lambda_{\mathrm{eff}} t} \tag{1}$$

where $C(t)$ [Bqm^{-3}] is the radon concentration in the chamber during time t [s], C_0 [Bqm^{-3}] – the initial radon concentration present in the chamber immediately after its sealing, E [Bqkg^{-1}s^{-1}] – the radon mass exhalation rate, m [kg] – the mass of the sample, V [m^3] – the total volume of the measuring system consisting of the volume of the chamber, the volume of the measuring device and connecting pipes, and λ_{eff} [s^{-1}] – the effective decay constant.

Effective decay constant of radon represents a radon removal probability from the chamber and consists of the radon decay (defined by the decay constant –λ), leakage probability (λ_{L} – the probability that radon will leave the chamber) and the back diffusion (λ_{b} – the probability that radon will diffuse back to the sample). In the case of a much smaller sample volume compared to the chamber volume, the back diffusion effect can be neglected [9].

Gamma spectrometric measurements

All samples for gamma spectrometric measurements were crushed, selected using a sieve with a 2 mm opening, and dried for 24 hours at 110 °C. Samples were packed in air-tight cylindrical PVC containers of 250 ml volume and stored for 40 days to achieve secular equilibrium before measurements.

Gamma spectrometric measurements were performed on coaxial HPGe detector, model GX5019, from Canberra®, with the relative efficiency of 55.1 % and the resolution of 1.75 keV for 1332.5 keV energy of ^{60}Co.

The efficiency and energy calibration were performed using a standard, prepared in the same geometry as samples using a soil matrix spiked with the common mixture of gamma ray emitters (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, and ^{88}Y) purchased from the Czech Metrology Institute [10]. The measurement time was between 80000 seconds and 255000 seconds.

RESULTS

An accumulation of radon concentration in the chamber as a function of time for brown coal from Miljevina mine (ID sample – 4) is shown in fig. 1.

The measured radon concentration was fitted using eq. (1) with a known sample mass and a volume of the measuring system, while radon exhalation rate, effective decay constant, and initial radon concentration were free parameters of the fit. From the fit, following values were obtained: the radon mass exhalation rate of E_{m} = (47.5 3.6) Bqkg^{-1}s^{-1}, the effective decay constant of λ_{eff} = (71.9 6.4) 10^{-7}s^{-1}, while the initial radon concentration of C_0 = (35.1 7.5) Bqm^{-3}.

In cases when the radon concentration in the chamber was small with large uncertainty, or the radon measurement was performed at the end of the deployment period, radon mass exhalation rate was extracted from the measured radon concentration at the end of the measuring period with already determined λ_{eff} and C_0 from the Sample 4.

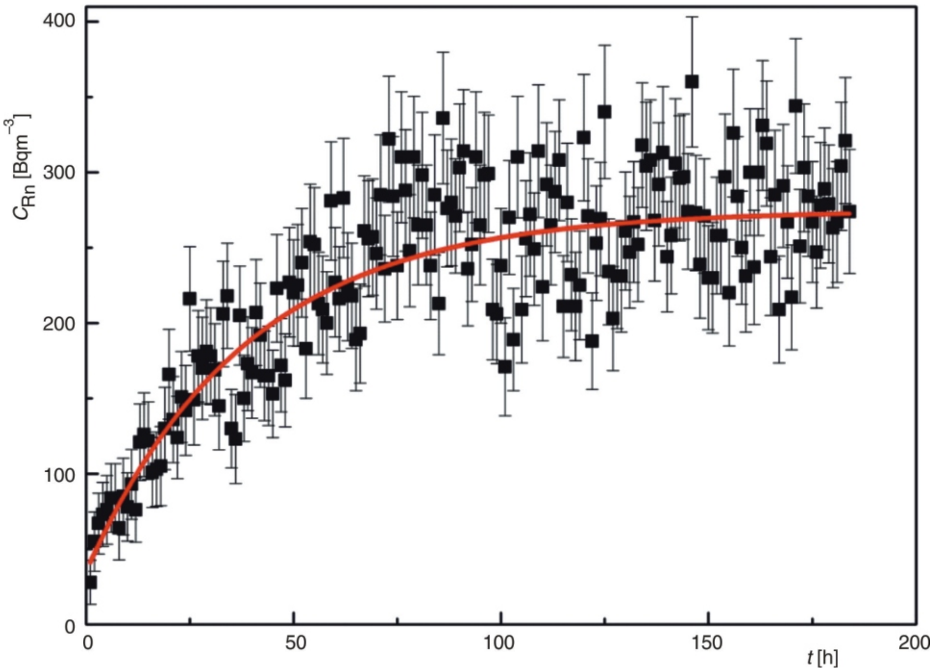

Figure 1. A build-up curve of radon exhaling from brown coal from Miljevina mine (sample ID: 4) in 30 L volume chamber

The obtained results of the radon mass exhalation rate, with an indicated coal sample and deployment period, are presented in tab. 1.

The measured radon mass exhalation rate ranges from (5.3 3.1) Bqkg^{-1}s^{-1} to (70.3 9.4) Bqkg^{-1}s^{-1}. These data are similar to the exhalation rates of typical building materials used in Serbia [7, 11].

If averaged by the type of coal, the obtained radon mass exhalations are: (67.7 2.7) μBqkg^{-1}s^{-1} for lignite; (41.4 6.2) μBqkg^{-1}s^{-1} for brown-lignite, (30 13) Bqkg^{-1}s^{-1} for brown and (19 14) Bqkg^{-1}s^{-1}for hard coal. Although there are small statistics and large uncertainty of mean values, there is a vague tendency of decrease in radon exhalation rate from lignite toward hard coal. The results could be understood in terms of the formation of different types of coal. Namely, lignite is formed with the least ground heat and pressure, making it more porous than other types of coal that are formed with much more heat and pressure, being, therefore more dense and solid.

The contribution of radon exhaling from coal to the indoor radon concentration can be expressed using the following expression

$$C_{\text{indoor}} \quad \frac{E_{\text{m}} m}{V\lambda_v} \tag{2}$$

where C_{indoor} [Bqm^{-3}] is the indoor radon concentration, E_{m} [Bqkg^{-1}h^{-1}] – the radon mass exhalation rate, m – the mass of the coal (assumed 5000 kg, for the whole season), V – the volume of the room with standard dimensions (5 4 3) m^3, and λ_{v} [h^{-1}] – the ventilation rate.

Typical values of ventilation rate are between 0.2-2 h^{-1}, with a geometric mean of 0.63 h^{-1} [12]. In the estimation of indoor radon concentration, two values of exhalation rate are used: the lowest value of the ventilation rate 0.2 h^{-1}, and 0.63 h^{-1} as the geometric mean of the ventilation rate. The obtained results are presented in tab. 1. The highest radon concentration of 105 Bqm^{-3} is obtained for lignite from Kovin mine, assuming that 5000 kg of coal was stored in a cellar with a ventilation rate of 0.2 h^{-1}. Although the obtained value does not exceed the recommended value of 300 Bqm^{-3}, the contribution of coal to indoor radon concentration from coal should be considered significant and kept as low as possible. Assuming that the transfer factor from the basement to the ground floor is equal to 0.5, the contribution to indoor radon concentration at the ground floor, from the coal placed in the basement, would be around 50 Bqm^{-3}. It can be understood as another factor influencing variability of indoor radon concentration, among many that were investigated in a typical family house in Serbia [13].

Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb in analysed coal samples are presented in tab. 2. Measured activity concentrations of ^{226}Ra are between 10.9-74.3 Bqkg^{-1}, of ^{232}Th are between 2-15 Bqkg^{-1}, of ^{40}K are between 2.2-107.3 Bqkg^{-1}, of ^{238}U are between 10.5-47.2 Bqkg^{-1}, for ^{235}U are between 0.48-4.11 Bqkg^{-1}, and of ^{210}Pb are between 9.47-78.0 Bqkg^{-1}.

The measured results agree with previously reported results of radionuclide activity concentration in coal used in Serbia [4, 14].

Since radon exhalation rate from any material depends on many parameters such as grain size, density (porosity) of the material, surface texture, and humidity, it is not surprising that there is a lack of correla-

Table 2. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb for different types of coal

Coal sample ID	^{226}Ra [Bqkg^{-1}]	^{232}Th [Bqkg^{-1}s^{-1}]	^{40}K [Bqkg^{-1}s^{-1}]	^{238}U [Bqkg^{-1}s^{-1}]	^{235}U [Bqkg^{-1}s^{-1}]	^{210}Pb [Bqkg^{-1}s^{-1}]
1	24.1 0.1	10.6 0.4	23.0 1.3	25.9 2.4	1.14 0.21	26.2 2.1
2	10.9 0.5	2.0 0.10	2.2 0.1	10.5 1.5	0.56 0.09	9.5 1.2
3	24.9 1.0	8.9 0.4	101.5 5.8	26.2 1.2	2.15 0.43	22.0 2.8
4	24.0 0.9	11.9 0.4	96.6 5.2	27.4 6.1	1.60 0.34	19.8 2.0
5	26.3 1.0	7.4 0.4	29.1 1.8	26.3 5.9	1.82 0.34	22.0 2.7
6	44.0 1.4	13.7 0.4	88.4 4.6	47.2 5.3	2.40 0.30	38.0 2.5
7	19.2 0.8	13.0 0.6	23.2 1.5	19.2 3.1	0.48 0.11	22.9 3.2
8	74.3 2.6	15.0 0.8	107.3 6.0	103.1 7.3	4.11 0.81	78.0 6.0

tion between activity concentration of ^{226}Ra and radon mass exhalation rate (Pearson correlation coefficient is 0.033). A similar observation was confirmed by investigating building materials in Serbia [7, 11, 15].

To assess gamma ray radiation hazard, various indices are used. One of such parameters is radium equivalent activity *Ra* that is used to compare different activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K of various materials. Radium equivalent activity is expressed by [16]

$$Ra_{\mathrm{eq}} \quad C_{\mathrm{Ra}} \quad 1.43C_{\mathrm{Th}} \quad 0.077C_{\mathrm{K}} \tag{3}$$

where C_{Ra}, C_{Th}, and C_{K} in [Bqkg^{-1}] are the activity concentrations of radium, thorium, and potassium, respectively.

External hazard index is defined as [16, 17]

$$H_{\mathrm{ex}} \quad \frac{C_{\mathrm{Ra}}}{370} \quad \frac{C_{\mathrm{Th}}}{259} \quad \frac{C_{\mathrm{K}}}{4810} \tag{4}$$

Results of Radium equivalent and external hazard index are presented in tab. 3.

The Ra_{eq} and H_{ex} of all samples are below recommended values of 370 Bqkg^{-1} and 1, respectively. Therefore, the analysed coal does not represent a significant radiation hazard.

Table 3. Radium equivalent activity and external hazard index for different coal samples

Coal sample ID	Ra_{eq} [Bqkg^{-1}]	H_{ex}
1	41.1	0.11
2	13.9	0.04
3	45.4	0.12
4	48.5	0.13
5	39.1	0.11
6	70.3	0.19
7	39.6	0.11
8	104.0	0.28

CONCLUSIONS

In this paper, the radiological impact of different types of coal used for domestic heating in Serbia is investigated. In total, eight different coal samples were analysed to determine activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb and radon mass exhalation rate.

The obtained radon mass exhalation rate ranges from (5.3 3.1) Bqkg^{-1}s^{-1} to (70.3 9.4) Bqkg^{-1}s^{-1} and is the highest for lignite type of coal due to its formation. It is the most porous and least dense coal compared to the other types of coal. Storing such coal for a heating season in the basement of the dwelling would cause an increase of indoor radon concentration at the ground floor by 50 Bqm^{-3}. Although the obtained value does not exceed the recommended one of 300 Bqm^{-3}, storing coal in such a way should be avoided.

Measured specific activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, and ^{210}Pb are similar to previously reported results of coal used in Serbia. The Ra_{eq} and H_{ex} indices, used to assess gamma ray radiation hazard, indicate that analysed coal does not represent a significant radiation hazard.

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AUTHORS' CONTRIBUTIONS

The idea of the research came from the discussion between I. Čeliković and G. Pantelić. Collection and preparation of samples were performed by M. Drašler and B. Lončar. Radon exhalation rate measurements were performed by M. Drašler, T. Milanović, and I. Čeliković. Gamma spectrometric measurements were performed by A. Kandić, A. Samolov and I. Čeliković. Data analysis performed by M. Drašler, A. Kandić, and I. Čeliković. B Lončar has cross-checked all the results. All authors contributed to writing the manuscript.

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ПРОЦЕНА РАДИОЛОШКОГ РИЗИКА ЗА СТАНОВНИШТВО УСЛЕД ИЗЛОЖЕНОСТИ РАЗЛИЧИТИМ ТИПОВИМА УГЉА КОРИШЋЕНИМ ЗА ГРЕЈАЊЕ У СРБИЈИ

Циљ овог истраживања је да се на основу мерења специфичне активности радионуклида и масене јачине ексхалације радона, процени потенцијални ризик који потиче од коришћења различитих врсти угља за грејање домаћинства у Србији. Измерене масене ексхалације радона се креће у опсегу од 5.3   3.1 μBqkg^{-1}s^{-1} до 70.3   9.4 μBqkg^{-1}s^{-1} и највећа је за лигнит. Процењено је да би угаљ ускладиштен у подруму објекта, могао да повећа до 50 Bqm^{-3} унутрашњу концентрацију радона у приземљу. Специфичне активности ^{226}Ra, ^{232}Th, ^{40}K, ^{238}U, ^{235}U, и ^{210}Pb у анализираном угљу су у сагласности са претходно измереним специфичним активностима радионуклида у угљевима коришћеним у Србији. Вредности радијум еквивалентне концентрације и екстерног хазард индекса су у границама препоручене и указују да коришћени угаљ не представља значајан радиолошки ризик.

Кључне речи: брзина ексхалације радона, гама спектрометрија, индекс радијационог ризика, угаљ