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Research Article

Assessment of Lignite-Fired Power Plants Impact on Radon Activity Concentrations

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Abstract:

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radon lignite mine activity vicinity Radon activity concentrations has been surveyed in dwellings in the vicinity of an opencast lignite mine and two lignite-fired power plants in the Pristina region, Kosovo. For this purpose solid-state nuclear track detectors CR-39 were exposed in the winter period for three months. Radon activity concentration ranged from 41 Bq m⁻³ to 327 Bq m⁻³, and the resulted annual effective doses was, from 0.74 mSv y⁻¹ to 5.83 mSv y⁻¹. The relationships of radon levels with the age of buildings, building material and local geology has been sought and is discussed. The impact of lignite-fired power plants on indoor radon concentration has not been observed.

1. Introduction

Radioactive noble gas radon and its radioactive short-lived products occur in the three naturally occurring decay chains headed by ²³⁸U, ²³⁵U and ²³²Th. Uranium and thorium are found in trace amounts in most rocks and soils; the most abundant isotope of uranium (over 99%) is ²³⁸U whose chain [1] includes ²²²Rn. The extensive studies on indoor radon concentration have shown considerable variations in different countries throughout the world [2]. In Kosovo, the first radon measurements were carried out in the course of prospecting uranium between 1983 and 1989 [3, 4]. It was soon followed by a survey in 89 dwellings, showing concentrations in the range from 20 to 450 Bq m⁻³, and exceeding 1000 Bq m⁻³ only in a few houses built of local stone in the uranium deposit area [4]. In the dwellings of Sharri community, the concentrations were <200 Bq m⁻³, except in one building [5]. Radon concentration in the dwellings of Planej and Gorozhup villages exceeded the value of 400 Bq m⁻³ only in two houses [6]. Also, the measurements in schools in Malisheva and Suhareka [7], Prizren [8] and

Sharri municipalities [9] found out low radon levels, with only a few concentrations in Prizren exceeding 400 Bq m⁻³, the level adopted from the International Commission on Radiological Protection [10] as our national radon limit for old buildings. On the other hand, as expected [11-14], in the Gadime Cave [15] higher radon concentrations were found, ranging from 400 to 1780 Bq m⁻³.

Coal generally contains trace amount of radionuclides with a typical range of activity concentrations [16] of 30–100 Bq kg⁻¹, 10–600 Bq kg⁻¹ and 10–200 Bq kg⁻¹, respectively, for ⁴⁰K, ²³⁸U and ²³²Th. However, some types of coal contain considerably higher amounts of ²²⁶Ra, which for lignite is not necessary in equilibrium [17]. Moreover, when coal is burned in coal-fired power plants, the remains, such as coal slag and fly ash, become more enriched in naturally occurring radionuclides than the unburned coal. The concentrations of $^{226}\text{Ra},~^{232}\text{Th}$ and ^{40}K in onsite bottom-ash were found to be 139 Bq kg⁻¹,108 Bq kg⁻¹ and 291 Bq kg⁻¹, respectively. From some studies in Kosovo's coal-fired power plants activity of radionuclides in coal, ash and slag were found to be lower compared with several available studies. The mean activity concentrations of 40 K, 226 Ra, and 232 Th in lignite were low, i. e., 36 ± 8 Bq kg⁻¹, 9 ± 1Bq kg⁻¹ and 9 ± 3 Bq kg⁻¹, respectively. They were markedly higher in fly ash and bottom ash, being 133 ± 16 Bq kg⁻¹, 30 ± 3 Bq kg⁻¹ and 30 ± 3 Bq kg⁻¹, respectively, in the former and 195 ± 13 Bq kg⁻¹, 28 ± 3 Bq kg⁻¹ and 34 ± 2 Bq kg⁻¹, respectively, in the latter [18, 19].

The national radon survey in Kosovo has not been finished yet and is continuing in steps, depending on the financial support, to eventually comprise representative number of schools, dwellings and workplaces in all municipalities. The study area of this work has not been investigated before and results will upgrade the situation of indoor radon in Kosovo. Thus, the aim of this paper is to complement the data of radon survey in Kosovo and eventually to contribute to the European Radon Map. In addition, we are going to assess the impact of lignite-fired power plants on indoor radon concentrations and investigate the dependence of indoor radon levels on the age of building, building material and geology.

2. Methods

The study was performed in the Prishtina region. The industrial complex, with a lignite mine and two thermal power plants, is located about 3 km northwest from the capital Prishtina, near settlement Obiliq. The climate of this region is characterised by an annual precipitation amount of 595 mm and an annual mean temperature of 10°C, summer maximum temperature of 39°C and winter minimum of -27° C. The local climate is classified as humid and mild continental. From the geological point of view, the Kosovo area is characterised by a variety of geological formations. Among them, there are rocks ranging from old crystalline Proterozoic to Quaternary age, comprising sedimentary and magmatic types together with rather less frequent metamorphic rocks. The geology of the study area which is characterized by metal enriched minerals with higher contents of heavy metals Cr, Mn, Co, Zn (concentrations in ash ranged for Mo, Pb, Th, U, Zn and Br between 0.1 and 10 mg kg⁻¹, for As, Co, Cr, Cu, Ni, Pb, V and Zn between 10 and 100 mg kg⁻¹, for Ba, Cr, Ni, Sr and Cl between 100 and 1000 mg kg^{-1} and for F as well as Mn between 1000 and 10000 mg kg⁻¹), is likely in contact zones between harzburgite (ultra-mafic igneous rock), serpentinite (metamorphic rock) and limestones [20, 21]. Kosovo has the largest coal reserves in South-East Europe; coal (in the form of lignite) is therefore expected to power the bulk of domestic electricity production in the future. The energy sector in Kosovo remains one of the most important sectors of the economy. It is also one of the most polluting, since lignite, oil and fuel wood make up 96 % of Kosovo's energy consumption [22].The exploited lignite, principally used for electricity production, generates a large amount of residues, as fly ash and bottom ash, that are derived from coal combustion. The total amount of ash is estimated to be approximately 55 million tons, according to the Kosovo Environmental Protection Agency. Fly ash from the combustion units A1 and A2 of the Kosova A power plant was disposed of as pulp on settling ponds (wet disposal technique). Bottom ash, slag and fly ash from the remaining combustion units have been transported by open conveyor belt systems to disposal sites (dry disposal technique). There is no protection against wind erosion of ash from ash deposal [20], thus allowing ash to be re-deposited in the region, depending on wind conditions.

The measurements of indoor radon concentration were performed by exposing solid-state nuclear track detectors CR-39 (Radosys), in 51 selected dwellings in the Pristina region.

The CR-39 detectors were provided and evaluated by the Institute of Radiochemistry and Radioecology at the University of Pannonia, Hungary. The exposure time of detectors in most dwellings was about three months, in winter season, from March to June. In all houses detectors were placed in the ground floor, preferably in the living room, where the residents spent most of their time indoors. They were fixed at 1-1.5 m above the floor and more than 0.5 m away from any wall and also any other object. In general surveyed buildings can be classified into two groups: older ones constructed between 1960 and 1980, and newer ones constructed after 2000 (Figure 1).

Most houses were built of adobe bricks, concrete and red (clay) bricks. Clay bricks are the most common building material in Kosovo [23]. For radiological safety reasons, estimating the concentrations of naturally occurring radioactive materials (NORMs) in the clay deposits of potential mining sites is essential to evaluate the occupational exposure during mining and processing, especially due to fine dust inhalation [24,25]. From some studies in the world, the activity concentrations (average+standard deviation) of the naturally occurring radioactive materials (NORMs), ²³⁸U, ²²⁶Ra, ²³²Th, ²²⁸Ra and ⁴⁰K, in clays samples were 49 \pm 20, 47 \pm 23, 34 \pm 11, 40 \pm 20 and 751 Bq kg⁻¹, respectively [26]. In the majority of cases, ventilation was natural, i.e., by opening windows.



Years of construction

Figure 1: Distribution of houses with respect to year of construction

To calculate annual effective doses for the residents, $E_{\rm eff}$ (mSv y⁻¹), caused by the exposure to the indoor radon, the following general formula was used [5, 27, 28]:

$$E_{\rm eff} = C_{\rm Rn} \times \frac{F}{3700} \times \frac{t}{170} \times f_{\rm DC}$$

 C_{Rn} is the radon concentration, *F* is the equilibrium factor between radon and its shortlived products for which the value of 0.40 was taken [10], and f_{DC} is the dose conversion factor with a value of 5 mSv WLM⁻¹ for dwellings [10]; denominators 3700 and 170 are necessary to express exposure to radon in WLM (old, though practical and still widely used unit), and *t* = 7000 h y⁻¹ was taken as the time spent indoors [10]. Because no statistical data were available on the living habits of residents, an occupancy factor of 0.80 was used, as proposed by the National Radiological Protection Board [29],

although other values [30-32], might better represent the actual situation.

3. Results and Discussion

Table 1 shows the radon concentrations in all surveyed dwellings. Radon concentrations ranged from 41 to 327 Bq m^{-3} with an average value of 111 Bq m^{-3} (Table 2). The majority of

values (82 %) were lower than 200 Bq m⁻³ (Figure 2), the value accepted as our national limit for new buildings. Our average is higher than that in Bulgaria [33, 34], of 90 Bq m⁻³ but lower than annual average of 249 Bq m⁻³ in 400 dwellings in Slovenia [35].

Our results of indoor radon concentration compared with neighbouring countries, are higher than in 126 dwellings at the Montenegrin coast [36], with the highest value of 175 Bq m^{-3} , and in 167 dwellings in Belgrade, Serbia's capital [37], with the highest value of 218 Bq m^{-3} . They are also higher than the values found in 94 dwellings in the Montenegrin capital, Podgorica, where 200 Bq m⁻³ was exceeded only in winter time [38]. On the other hand, they are lower than those reported for 437 dwellings in Macedonia where in winter time, 400 Bq m⁻³ was exceeded in 29 buildings [39, 40]. But, our results may not be compared, for example, with those obtained in dwellings in Niška Banja [41, 42], southern Serbia, where at some places radon concentration reached orders of kBq m⁻³. Based on experience gained in dwellings and schools in Kosovo [6, 9] the radon concentrations in Table 1, obtained in the spring period, were assumed to represent an approximate annual average, because autumn values would be similar, while summer and winter values would be 15-25% lower and higher, respectively.

No.	<i>C</i> _{Rn} (Bq m ⁻³)	<i>E</i> _{eff} (mSv y ⁻¹)	No.	C _{Rn} (Bq m ⁻³)	$\frac{E_{\rm eff}}{({\rm mSv}\ {\rm y}^{-1})}$
1	241 ± 20	4.29	27	58 ± 02	1.04
2	222 ± 39	3.95	28	48 ± 12	0.86
3	287 ± 26	5.11	29	42 ± 18	0.76
4	96 ± 14	1.70	30	54 ± 06	0.96

Table 1: Radon activity concentrations (C_{Rn}) and annual effective doses (E_{eff}) .

5	96 ± 23	1.72	31	109 ± 10	1.94
6	87 ± 32	1.55	32	95 ± 24	1.69
7	110 ± 09	1.96	33	229 ± 32	4.08
8	135 ± 16	2.40	34	49 ± 11	0.87
9	92 ± 27	1.64	35	114 ± 05	2.03
10	71 ± 11	1.26	36	48 ± 12	0.86
11	60 ± 01	1.06	37	52 ± 08	0.92
12	81 ± 21	1.44	38	112 ± 07	2.00
13	48 ± 12	0.86	39	119 ± 01	2.12
14	46 ± 14	0.81	40	41 ± 19	0.74
15	70 ± 10	1.24	41	144 ± 25	2.57
16	71 ± 11	1.26	42	74 ± 14	1.32
17	115 ± 04	2.05	43	88 ± 28	1.57
18	80 ± 20	1.43	44	50 ± 10	0.89
19	159 ± 40	2.84	45	83 ± 23	1.47
20	50 ± 10	0.89	46	68 ± 08	1.21
21	253 ± 08	4.51	47	59 ± 01	1.06
22	58 ± 02	1.03	48	176 ± 57	3.13
23	247 ± 14	4.40	49	52 ± 08	0.93
24	177 ± 58	3.15	50	92 ± 27	1.64
25	282 ± 21	5.03	51	54 ± 06	0.96
26	327 ± 66	5.83			

Table 2. Statystical parameters analysis.

Statistical parameter	C_{Rn} (Bqm ⁻³)	$E_{\rm eff} (mSv y^{-1})$	^a arithmetic mean, ^b arithmetic standard deviation,	
AM ^a	111	1.98		
ASD ^b	74.3	1.32	^c minimum value,	
Min. ^c	41	0.74	value,	
Max. ^d	327	5.83	^e geometric mean	
GM ^e	92.8	1.65	fgeometric	
$\mathbf{GSD}^{\mathrm{f}}$	1.8	1.8	standard deviation.	



Figure 2: Distribution of ^{222}Rn activity concentrations $(C_{Rn}/Bq m^{-3})$ in dwellings



Figure 3: Distribution of the annual effective doses (E_{eff}) in dwellings.

Thus, values from Table 1 were considered as annual averages and used in $E_{\rm eff}$ calculation. The annual effective doses for each house included in the survey are presented in Table 1. The annual effective doses ranged from 0.74 to 5.83 mSv y⁻¹ with an average value of 1.98 mSv y⁻¹ (Table 2). From the distribution of annual effective doses in Figure 3, it can be shown that in most houses the value fall in the range between 0.5 and 2 mSv y⁻¹. Table 2 gives for radon concentrations and effective doses the followings: minimum and maximum values, their

arithmetic (AM) and geometric (GM) means, and arithmetic (ASD) and geometric (GSD) standard deviations. AM and GM values for annual effective doses were 1.98 and 1.65 mSv y⁻¹. These values were found to be higher than the world mean value $(1.15 \text{ mSv y}^{-1})$ [2].

The highest values of ²²²Rn activity concentration were observed in the old houses (constructed before 1980) and the lowest, in the new houses (constructed after 2001). The highest value of the ²²²Rn activity concentration was observed in the buildings, constructed of red (clay) bricks.

This can be explained by the fact that red bricks are made of the clay, possibly rich in natural radionuclides (²³⁸U, ²³²Th, ²²⁶Ra and ⁴⁰K). The highest mean of the ²²²Rn activity concentration was observed in the houses, constructed of adobe bricks. The lowest values of the ²²²Rn activity concentrations were found in the houses constructed of concrete bricks.

A relationship has been observed between radon activity concentrations and the year of the house construction. Higher activity radon concentrations were found in houses constructed before 1980 than in those constructed between 1981 and 2000. Also the buildings constructed between 2001 and 2014 have lower activity concentrations. The reason of lower concentrations in newer houses maybe is in better isolation of basement and good ventilation of these houses. No impact of lignite-fired power plants on indoor radon concentrations has been found.

4. Conclusion

The radon activity concentration was measured by exposing solid state nuclear track detectors (CR-39) in the period of winter season, from March to June, in dwellings in the Pristina region. Radon concentrations ranged from 41 Bq m⁻³ to 327 Bq m⁻³, with an AM of 111 ± 74.3 Bq m⁻³ and GM of 92.8 ×/:1.8 Bq m⁻³. Annual effective doses ranged from 0.74 to 5.83 mSv y⁻¹, with an AM of 1.98 \pm 1.32 mSv y⁻¹ and GM of 1.65 ×/: 1.8 mSv y⁻¹. Maximum radon concentration value was found to be 327 Bq m⁻³. The national limit of 200 Bq m⁻³ for new buildings was exceeded in 11 houses, and the national limit of 400 Bq m⁻³ for old buildings was not exceeded. Radon activity concentrations and the resulting annual effective doses were higher than in some other locations in Kosovo, and also higher than at several places in the neighbouring and other countries. No impact of lignite-fired power plants on indoor radon concentrations was found. The investigations should be extended to other locations,

with emphasis to those with potentially higher radon levels, as for instance the Stublla region.

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