Dose in Buildings Caused by Built-in Coal-Sags with Elevated ^{226}Ra Concentration, and the Possibilities of Intervention

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INTRODUCTION

From the radiological point of view, the use of coal-slags as building material, which may affect indoor doses from external radiation and the inhalation of radon decay products, may be significant.

In the Transdanubian region in Hungary in many cases the ^{226}Ra concentrations of coals exceed significantly the world average. (1,2) Slags originated from stoves, boilers and power plants by burning these coals, were applied in buildings as backfill or insulation material underneath the floors and in the ceilings.(3,4,5)

In this region there are three towns (Ajka, Tatabánya, Várpalota) with coal-fired power plant in their neighbourhood. Radionuclide concentrations of slag samples comes from these three towns have been investigated. Dose rates were measured in schools, kindengartens and dwellings and, in some cases, the radon concentration was determined. The intervention possibilities were also investigated. The results of the measurements do not represent the average levels of the cites because only the dwellings containing slag were examined.

EXPERIMENTAL

Activity measurements of coal-slag

Sampling and sample preparation – In some homes and schools, we were able to take samples of the built-in slags either from under the floor or from the ceiling. Coal-slag samples of about 2 kg were gently broken and dried in air, later at 105 ^{o}C. The dried samples were stored for 30 days in airtight aluminium Marinelli beakers with a volume of 600 cm^{3}, to reach the radioactive equilibrium of ^{226}Ra with its progeny. The concentrations of natural radionuclides were determined by high-resolution gamma ray spectrometry, using an n-type Oxford / Tennelec HPGe detector with an efficiency of 14% and energy resolution of 1.8 keV at 1332.5 keV. The gamma spectra were recorded by the Oxford / Tennelec PCA-ME 8192-channel analyser. The data collection time was 60,000 seconds and the system was calibrated using an etalon containing natural uranium ore certified by the Hungarian National Authority of Measures. The ^{226}Ra concentration of the etalon was $28.3 \cdot 10^3 \pm 1.4 \cdot 10^3$ Bq kg^{-1}.

The ^{226}Ra concentrations were determined by measuring the activities of its decay products, ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 and 1120 keV), that were in secular equilibrium with ^{226}Ra following the 30-day storage. The activity of ^{40}K was measured by the 1461 keV gamma ray, the ^{232}Th by the 911 keV gamma ray of ^{228}Ac and the 2614 keV gamma ray of ^{208}Tl.

Determination of the external gamma dose rate

Depending on the dimensions of the rooms, the absorbed gamma dose rate was measured at 8 – 20 spots at 1 m height above the floor and calculated the average values. Additional measurements were provided at the floor at the height of beds and chairs (0.5 m above the floor) and at the ceiling.

Monitoring of radiation level was carried out with a portable dosimeter (type of Berthold AUTOMESS 6150ADB) calibrated by the Automation und Messtechnik GmbH, Germany. The energy dependence is in the range of $\pm$ 20%. 30 keV to 7 MeV

Radon concentration measurements

Measuring the radon concentration two methods, the short period and the integral ones, were used.

Short period measurements – In this case some weeks long continuous measurement was carried out. The used instruments were a PYLON AB-5 with PCRD detector and a Radim 2P with semiconductor detector. These radon monitors store the hourly averages in their memory, so the variation of the radon concentration also can be detected.

Integral method – For these, some months long, measurements solid state CR-39 type TASTARK nuclear track-etch detectors were used. Prior to exposures, the detectors were washed in 5% ethyl alcohol, then pre-etched in 6 M NaOH solution at 70 ± 0.5 ^{o}C for 2 h. Finally, background level of etch-density was determined. The prepared detectors were placed in holders – made by the National Protection Board, UK – and distributed to homes. After 3-6 month exposure the detectors were etched for 4 h, under similar conditionsin pre-etching. In the evaluation a Virginia software was used. The nuclear track detectors were calibrated in a chamber (EV 03209 Genitron Instruments GmbH) with a PYLON RN 2000A calibrating standard.

Dose calculation

Dose rates measured at 1 m height were used to assess the individual external dose, although this could have an underestimation in some cases, depending on the situation of the built in slag. 7000 h occupancy were used for homes and the conversion factor was 0.8 Sv Gy^{-1}.

The dose from radon was calculated using the recommendations of ICRP 65 (6), namely, supposing 7000 h y^{-1} occupancy and 0.4 equlibration factor, and 100 Bq m^{-3} average ^{222}Rn concentration cause an annual 1.72 mSv effective dose contribution.

Options of interventions

Decrease of the gamma dose rate – The decrease options of gamma dose rate were studied firstly by computer simulations, namely the MicroShield 5.0 software was used for assessment of the radiation field from sources in the building material. The slag usually was built in under the floor and/or above the ceiling. The slag layers were usually 15-25 cm thick, so these two values were used for the calculations. The averages taken from the measurements were used in the model as the concentrations ^{232}Th and ^{40}K. The ^{226}Ra concentrations were used between 500-2500 Bq kg^{-1} because they were very different in the real cases as well. The room dimensions were 3x4x2.8 m. Dose rates at 1 m heigt were calculated considering different thick (0-10 cm) concrete and barite-concrete layers. The results of the model was compared with the measured values of two real situation, where the important parameters (radionuclide concentratios of slags, thickness of the concrete layer, etc.) were known.

Decrease of the radon concentrations - The change of the average radon concentration were examined during a 20 days long periods caused by the frequent airing (4-5 times per day).

An active intervention was carried out in a dwelling. Perforated pipes were put into the slag under the floor which were led to the roof where a ventilator (10 W power) was joined to the pipes.

The variations of the radon concentration of different time periods were examined in both cases of working and not working ventilator.

RESULTS AND DISCUSSION

Radionuclides in slag samples

Table 1. presents the activity concentrations of the ^{226}Ra, ^{232}Th, and ^{40}K in slag samples used as construction materials. The highest value was determined for ^{226}Ra including the highest variability.

Table 1. Radinuclide concentration of slag samples originated from the three towns.

Place of origin	Average radionuclide concentrations (min.-max.) (Bq kg^{-1})		
	^{40}K	^{232}Th	^{226}Ra
Ajka	194 (45-386)	36 (16-81)	1714 (578-2893)
Tatabánya	347 (257-468)	80 (50-119)	1928 (843-2407)
Várpalota	274 (238-333)	42 (37-48)	315 (160-523)

It can be seen that the ^{226}Ra concentrations exceed significantly the 50 Bq kg^{-1} world average of building materials. The materials with the high levels of radioactivity are banned for use as building ones in the most of the countries.

Gamma dose rates

The distribution of gamma dose rates measured at 1 m heights can be seen in the Table 2. In the most of the cases the dose rates exceed the 80 nGy h^{-1} world average (7) and the 116 nGy h^{-1} Hungarian average as well.

Table 2. Distribution of absorbed dose rates in schools, kindergartens and flats determined at 1 m heights

Dose rate (nGy h^{-1})	Number of rooms					
	Schools, kindergartens			Flats		
	Ajka	Tatabánya	Várpalota	Ajka	Tatabánya	Várpalota
- 150	6	4	271	0	66	65
150 – 250	13	56	37	13	47	9
250 – 350	8	40	40	18	41	0
350 – 450	2	22	0	9	34	0
450 – 550	3	10	0	3	8	0
550 – 650	8	2	0	4	2	0

At the height of beds and chairs the gamma dose rates usually were 10% higher then the values measured at 1 m. But the floor level the rates were significantly higher, and in some cases they exceed the 1000 nGy h^{-1} value.

Results of radon concentrations

The results of measurements made by nuclear track-etch detectors are given in Table 3.

Table 3. The radon concentrations in the three towns measured by nuclear track-etch detectors.

Town	Number of rooms monitored	Radon concentrations ($Bq\ m^{-3}$)		
		Average	Medián	Range
Ajka	8	887	862	547 - 1310
Tatabánya	35	268	177	26 - 678
Várpalota	28	128	124	29 – 338

The higher levels in town Ajka was confirmed by investigation of the emanation of slags originated from the use of local mined coal.

In schools and kindergartens 1-2 weeks long continuous measurements were carried out as well. The radon concentrations in the classrooms were under 200 Bq m^{-3} during the occupation by the children in all the cases, due to the frequent opening of doors and windows, even it sometimes the average radon concentration was high according to the integral radon measurements. The Figure 1. represents a variation of radon concentration in a kindergarten room. It can be seen that the radon concentration increased significantly during the nights and the weekend when the kindergarten was closed.

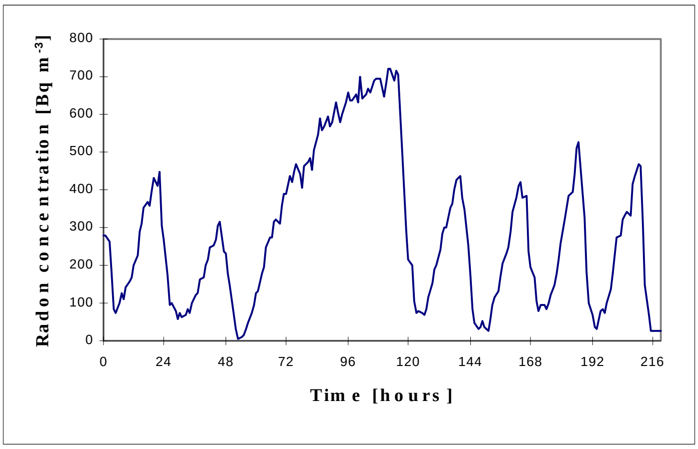

Figure 1. The variation of radon concentration in a kindergarten.

Effective doses in flats

Doses in some flats from the towns Ajka and Tatabánya are in Table 4. The yearly effective dose was the sum of gamma and radon dose contribution assessed as mentioned in the experimental part. It can be seen that the yearly effective dose in these flats much more higher then the world average one and in some cases it can exceed the annual occupation dose limits (20 mSv).

Table 4. The dose contributions in some flats in Ajka and Tatabanya

Flat	Yearly dose contributions		
	gamma	radon	sum
A_1	1.13	9.41	10.5
A_2	1.58	9.59	11.2
A_3	1.4	10.3	11.7
A_4	1.6	19.6	21.2
A_5	2.67	20.5	23.2
A_6	2.95	22.5	25.5
T_1	2.34	5.95	8.29
T_2	1.75	6.09	7.84
T_3	2.24	7.38	9.62
T_4	2.67	8.31	11.0
T_5	2.26	8.37	10.6
T_6	2.82	8.49	11.3
T_7	1.73	8.48	10.6
T_8	1.89	9.73	11.6
T_9	3.25	11.3	14.5
T_{10}	2.22	11.6	13.8

Interventions

Shielding of the gamma radiation - The shielding effects of concrete and barite-concrete layers plotted against the thickens of shielding in the cases of 15 and 25 cm thick slag layers with different ^{226}Ra concentrations can be seen in Fig. 2. and Fig. 3. In the most places the thickness of the applicable shielding layers are limited, so the shielding material and method should be planned by taking into consideration the calculated absorption and dose rates. For example the dose rate can be decreased to one fifth applying 10 cm barite-concrete coverage. The effects of the 10 cm thick shielding are presented in Table 5 by comparison of the calculated and measured ones. It can be seen that there are no significant differences between them. The difference could come from the inhomogenety of radium concentration and thickness of slags.

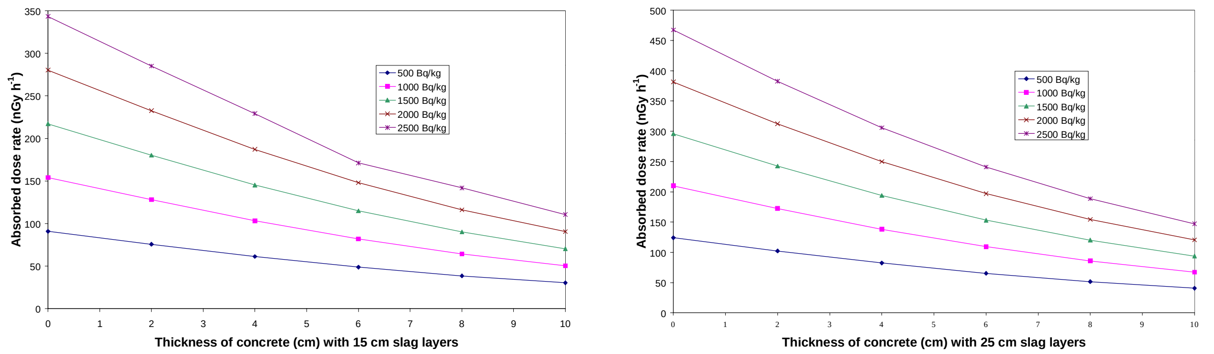

Figure 2. Plots of dose rates versus thickness of concrete layers in the cases of 15 cm and 25 cm slag layers with different ^{226}Ra concentrations.

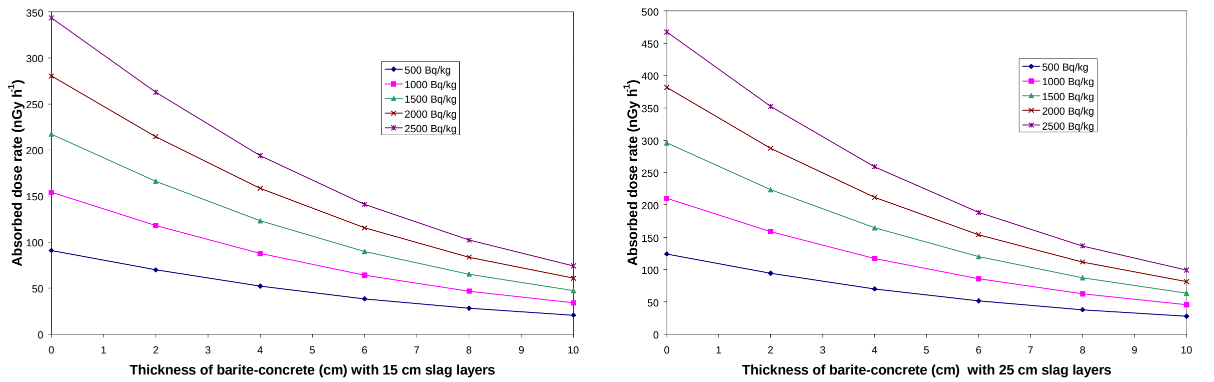

Figure 3. Plots of dose rates versus thickness of barite-concrete layers in the cases of 15 cm and 25 cm slag layers with different ^{226}Ra concentrations

Table 5. Measured and calculated dose rates in two rooms

Room	Measured/modelled absorbed dose rates (nGy h^{-1})		
	floor	1 m	ceiling
1.	400/391	350/360	330/323
2.	350/390	310/334	306/322

Airing and ventilation for decrease of the radon concentration - The frequent airing results a significant decrease in the radon concentrations during the 20 days measuring periods, namely the 951 Bq m^{-3} average decreased to 511 Bq m^{-3}. 46% decrease might be significant, although the method is less rentable considering the heating expenses.

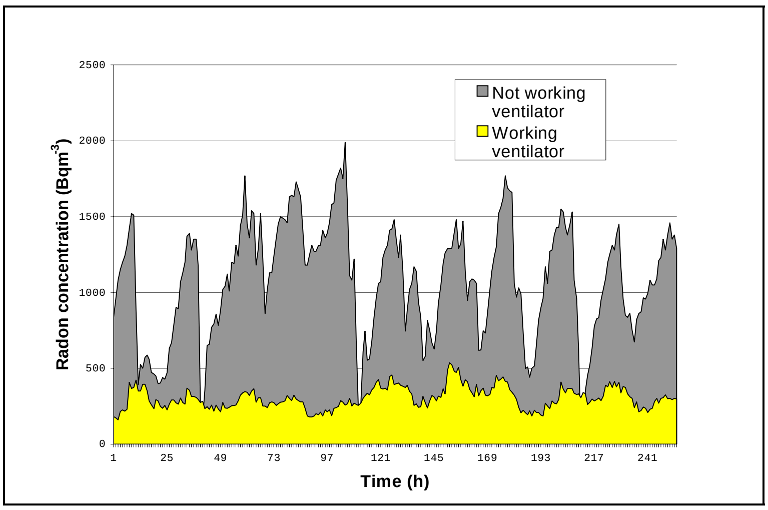

Figure 4. Variation of the radon concentrations without and with an active the ventilation.

The Fig. 4. show the results of the ventilation method in a 10 days period. The upper curve show the radon concentrations when the ventilator did not work and the lower one when the ventilator was working. The average 966 Bq m^{-3} radon concentration decreased to 247 Bq m^{-3}. This means 70% decrease. (This results can be increased with increase the power of the ventilator.)

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