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RADIOACTIVE CHARACTERISTICS OF THE LIASSIC COAL OF PÉCSBÁNYA AND EFFECTS OF ITS MINING ON THE ENVIRONMENT (MECSEK MTS. – SOUTH HUNGARY)

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ABSTRACT

It is a widely known fact that the Liassic coal mined for more than 200 years in the environs of Pécs-Komló (Mecsek-Mts., South-Hungary) represents a higher radioactivity than that of the formation average. Hence, in order to carry out a successful recultivation it is necessary to determine precisely the radioactive state of the involved areas and measure the amount of excessive radioactivity affecting citizens. This is important especially because measurements of this kind haven't been made in the area before. By performing in situ and laboratory measurements on waste heaps, accumulated over several hundred years, and areas under different stage of mine works the values of total gamma-ray activity, the specific activity of rocks building up the area and the concentration of the three most significant natural radionuclides (U, Th, K) were determined.

Based on in situ measurements, both the winter and the summer-time (representing totally different meteorological conditions) gamma-ray dose rate distribution map of the Karolina opencast mine, and its vicinity was drawn. The background value of total gamma-ray dose rate, based on values measured in the farther environs of the mine was 85-90 nGy/h in the dry hot summer period and 75-80 nGy/h during the humid and cold wintertime. On areas still under active mine works the same measurements gave 220% higher results in average, and in terms of uncovered waste heaps these values were just slightly lower (150-160 nGy/h). On the other hand, in case of covered waste heaps the applied 40-60 cm thick soil cover almost completely absorbs excess activity, thus values received on covered heaps were only 10 % higher than the environmental background.

Gamma-ray spectroscopic measurements have shown that not only U bound to organic material is responsible for increased radioactivity, but K and Th as well. The concentration of these later elements proved to be the highest in rocks abundant in argillic minerals. Hence, the activity is the highest especially in those rock types which are characterised not only by a high organic material content but argillic mineral content too.

Key words: Liassic coal, gamma ray activity, gamma-ray spectrometry, Mecsek-Mts.

INTRODUCTION

Rocks of high organic material content (oil-shales, coals) are usually rich in radioactive elements as well. U and Th contents often exceed by several times or even by an order of magnitude the characteristic world average referred to coals. The world average is claimed to be 1-5 ppm and 1-7 ppm for U and Th, respectively (Valkovic, 1983; Hoffman, 1988; Swaine, 1990, 1997; Eisenbud and Gesell, 1997).

The radioactive element content of Hungarian coals was first examined by Szalay and Földvári parallel to fissile material explorations in the country (Szalay, 1948; Földvari, 1951; Szalay and Földvari, 1951). According to them, the following Hungarian coals proved to represent higher radioactivity than the usual value: the Cretaceous coal of Ajka; the deposits of Kisgyón, certain deposits in the Tatabánya coal basin and the Liassic coal of the Mecsek Mts. (Szalay, 1952, Szalay and Almássy, 1956; Bodrogi et al., 1959; Upor et al., 1960). In his summarising work Szalay states that only those deposits are enriched in radioactive elements – primarily uranium – that are located relatively close to the denudation zones of still traceable granitoid bodies. Based on laboratory research, he explained the increased radioactivity – significantly higher than the formation average – with the uranium accumulating character of organic material (primarily humic acids) (Szalay, 1952).

As a continuation of the above studies, the aim of the present research is to measure the total gamma-ray activity appearing in the Karolina opencast coal mine and in its surroundings, and besides, to aid an environmentally successful recultivation by determining the amount of excess exposure dose affecting not just the environment but local people as well.

Preliminary laboratory gamma-ray spectroscopic analyses had shown that the sometimes astonishingly high values of total gamma-ray activity (250-300 nGy/h) should not be caused by the average 1-6 ppm uranium concentration alone. Thus, in situ total gamma-ray activity measurements were extended to three elements: U, Th and K^{40}. Field results were always supplemented by laboratory gamma spectroscopy, and the concentration of radioactive elements was controlled by the ICP-MS method.

STUDIED AREA

Detailed in situ total gamma-ray activity analyses were performed in the Mecsek Mountains (South Hungary),

7 km ENE of the region's centre, Pécs, on samples collected in the Karolina opencast coal mine, located west of village Pécsbánya. Further samples were taken from recultivated and unrecultivated waste heaps north and south of the mine, the neighbouring settlement, and areas not involved in mining activity. The mineworking conditions of the Karolina opencast mine are presented on Fig. 1.

The mine explores a Lower Liassic sedimentary complex, which contains 18-22 exploitable coal deposits, 50-120 cm thick each. The waste material of the coal deposits is composed of aleurolites of low organic material content and arkosaic sandstone.

Methods

Factors determining the radiological conditions of an area (total gamma-ray activity, radon concentration and exhalation, falling dust or aerosol activity) are highly dependant on meteorological parameters. During the cold and windy wintertime the dust and aerosol content of the air is significantly lower, and during long lasting humid periods the radon exhalation of soils and the base rock is also lower. Meanwhile, the dry and warm summertime is adequate for high dust and aerosol content, and dry weather results increased radon exhalation, too. Hence, when examining the quality and quantity of total gamma-ray activity it is necessary to repeat the measurements in different seasons.

During total gamma dose rate analyses regardless of its actual components the intensity of gamma-ray was determined, as it is the most penetrating radiation and thus occurs everywhere. In situ total gamma-ray activity measurements were carried out both during summer- and wintertime at 375 locations in the Karolina opencast area and its environs maintaining a 50 x 50 m sampling grid. The measurements were made with a portable NC-483 nuclear analyser equipped with a NaI(Th) scintillation detector (MEV, ND-482), which enabled the required energy-independent determination. Measurements were made following the recommendations of the International Atomic Energy Agency, 1 m above ground level with a 10 s sampling time. The method provides a precise result even in terms of close to background values, still, the measurements were repeated three times in each sampling points and the final result was the average of these. Nevertheless, on the area of the actual mine the very intensive relief inhibited the use of a grid when sampling, thus average values detected above boundaries of different rock types were applied in order to characterise the area of the mine works.

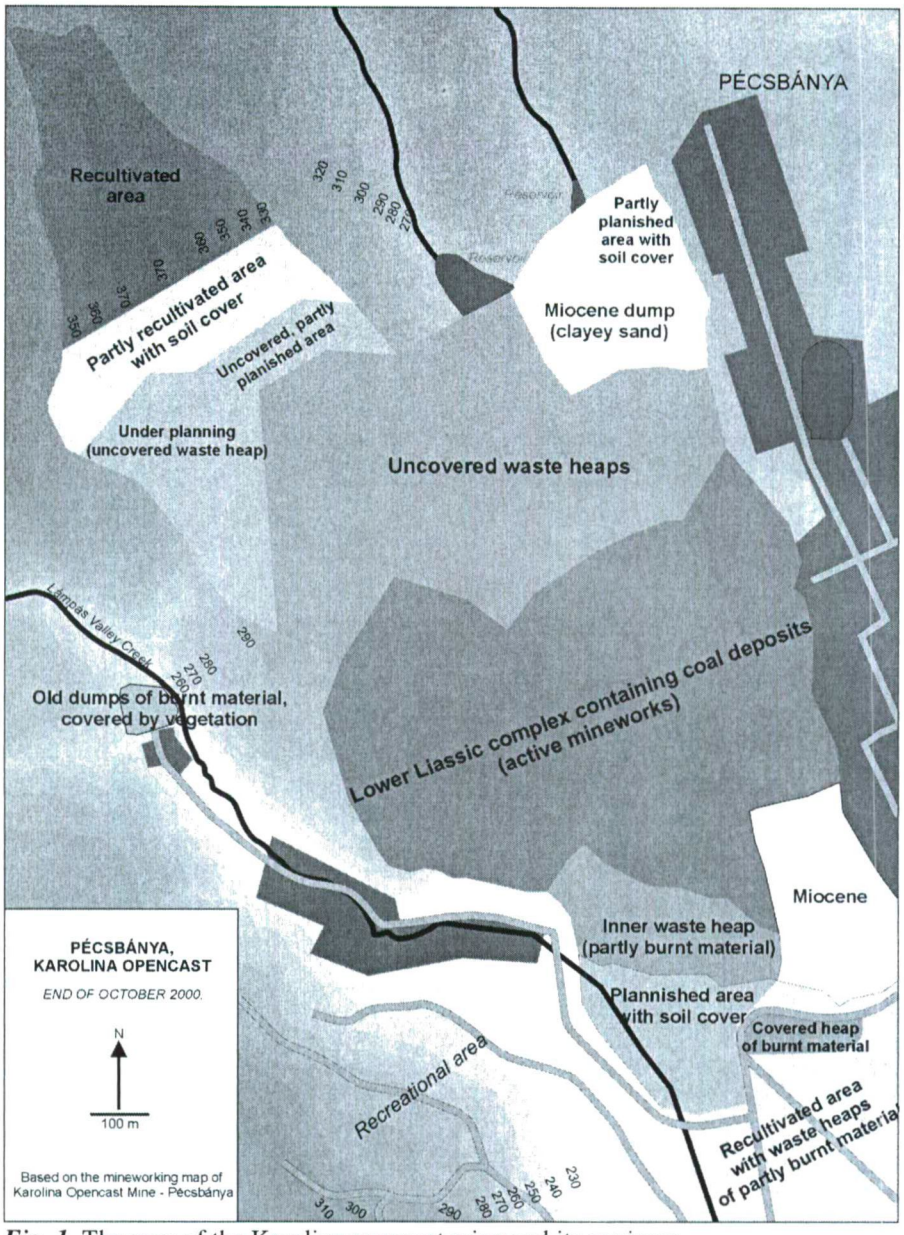

Fig. 1. The map of the Karolina opencast mine and its environs

In situ gamma-ray spectroscopic analyses aimed at the measurement of the main radioactive elements – Th, U(Ra), K – occurring in the soil and the base rock. Actually, in case of uranium the radium content of the samples was determined, and the uranium content was calculated assuming a radioactive equilibrium. It is possible during gamma-ray spectrometric measurements to determine the concentration of radioelements and radiaoactive families, too, by selectively detecting the photons of different energy level that make up the total gamma-ray radiation. Measurements were carried out with a ND-482 scintillation detector attached to a MEV NC-483 analyser following IAEA recommendations, i.e. with a probe placed on the ground and due to low concentrations with a long measuring time (10 min). For increasing the preciseness of the results two measurements were made in each point.

The collected samples' laboratory analyses was carried out with a four banded nuclear analyser (NP484-P), while the control samples were processed following the ICP-MS (HP 7500) method after partial nitric acid extraction (using microwave, 40 minutes, 250 °C)

Discussion and conclusions

In situ total gamma-ray activity and gamma-spectroscopy

The measurement of total gamma-ray dose rate and gamma-ray spectroscopy performed on samples originating from the Karolina opencast mine, surrounding waste heaps and village Pécsbánya enabled the detection of excess radioactive exposure and the determination of the environmental background or zero radioactivity both of which is crucial for launching successful recultivation works in the area. Applying the data of summer and winter measurements two maps were drawn representing the distribution of total gamma-ray dose rate in the two different meteorological periods (Fig. 2, 3).

The average values of environmental background were determined on the basis of 30 measuring points, that were located farther of the mine and represented areas that have never been involved in mining activity and have a different rock base but always with a soil cover. Thus values during the dry, hot summer period were 85-90 nGy/h, while at wintertime, when the weather is cold and humid the measurements gave 75-80 nGy/h. Concerning the settlement of Pécsbánya, due to traffic, coal transportation and heating the average value of total gamma-ray dose rate was 95-103 Gy/h. Values measured on the territory of the mine works and on uncovered waste heaps

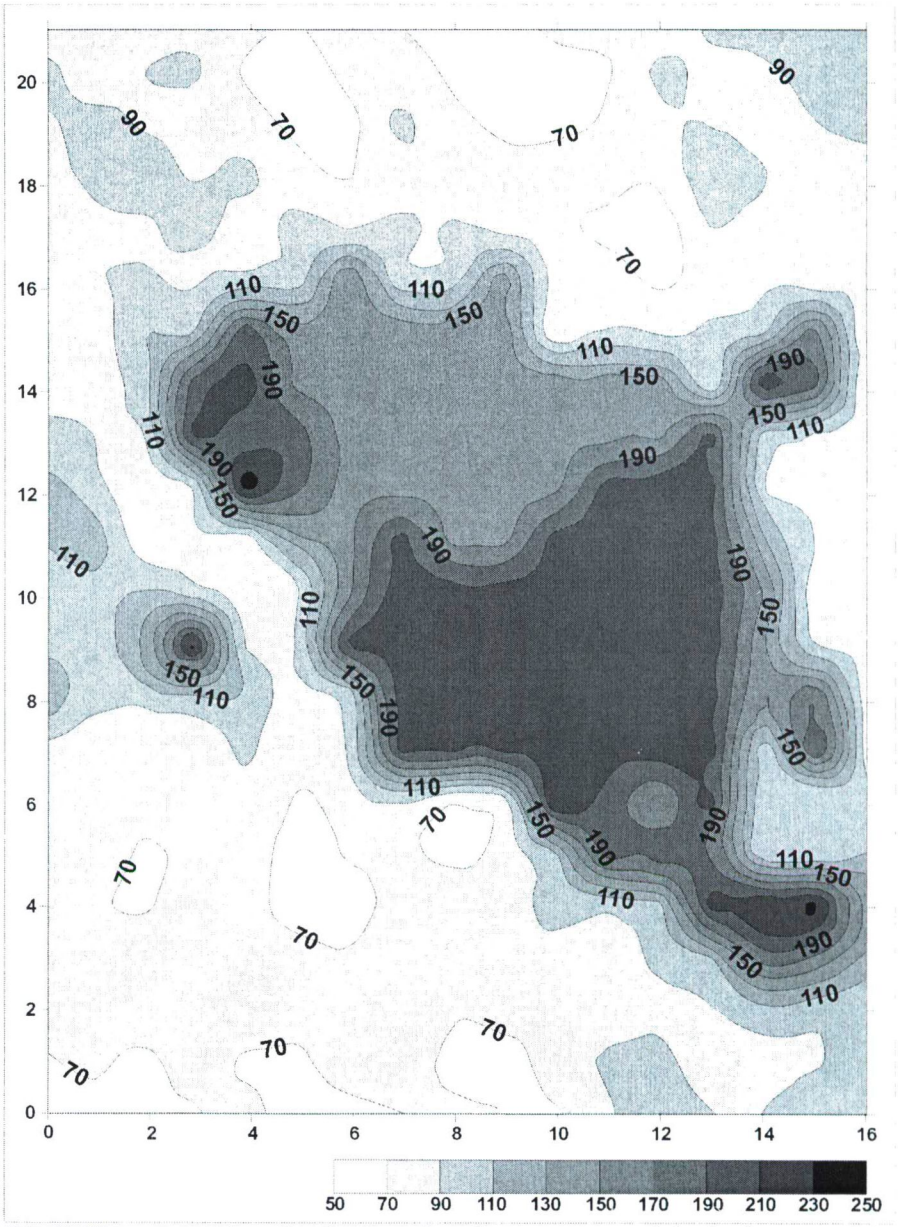

Fig. 2. The summertime distribution of total gamma-ray activity in the area of the Karolina opencast mine

are 220% higher in average than the zero dose rate measured in the undisturbed environs of the mine (Table 1). However, it is notable that high total gamma activity concluded in terms of the opencast mine is partly resulted by the fact that more samples were taken from above productive coal layers, thus measuring points of higher radioactivity are slightly over-represented in the measurement series. The mean total gamma-ray activity of

Table 1. Values of total gamma ray activity in the summer- and winter period at locations of different mining activity

Types of mining activity	Total gamma-ray activity (nGy/h)							
	Summer period:				Winter period:			
	mean	max.	min.	sampled	mean	max.	min.	sampled
Opencast	219	320	121	57	207	334	120	38
Uncovered, „active" waste heaps	169	247	85	31	154	223	81	31
Uncovered, planished waste heaps	176	154	205	6	177	205	141	6
Covered, planished waste heaps	90	112	67	13	85	110	65	13
Recultivated waste heaps	97	181	76	31	89	131	71	31
Partly covered waste heaps of burnt	212	247	161	12	171	201	101	12
Settlement (Pécsbánya)	103	231	56	33	95	185	61	33
The vicinity of the opencast mine (max.	82	141	52	194	76	131	50	194

the opencast therefore is closer to that of the uncovered waste heaps but it is still higher due to vast coal deposits of high radionuclide concentrations and aerial dust deposition. Data on gamma-ray dose rates determined on the territory of the open cast, and the calculable concentrations of elements are presented in Table 2.

In case of uncovered and/or planished waste heaps the value of total gamma-ray activity is twice as high as the environmental background, i.e. 154-177 nGy/h. Nevertheless, on permanently recultivated waste heaps the 40-60 cm soil cover almost completely absorbs the excess radiation, and values measured here are higher than the environmental zero only by 10% (Table 1).

The precise determination of excess exposure affecting nearby living people would require the measurement of falling dust and aerosol activity, too. Total gamma-ray activity measurements however suggest that the rate of excess exposure on the dwellers of Pécsbánya is approximately 15-25%. Based on international data, the ratio of the time spent inside the dwelling and outside is 20/80 %. Thus, counting with an annual 1800-2200 hours of exposure, the dose equivalent of the excess load on citizens of Pécsbánya, resulted by the additional gamma activity due to coal mining, is 20-30 μSv/year. However, the calculated dosimetric quantities even in case of unreal length of exposure are well below medical thresholds, still they draw the attention to old waste heaps of burnt material and the accumulated clinker of the power station of Pécs, the recultivation of which has not been solved yet. Total gamma-ray activity values were far the highest (170-210 nGy/h) at these locations.

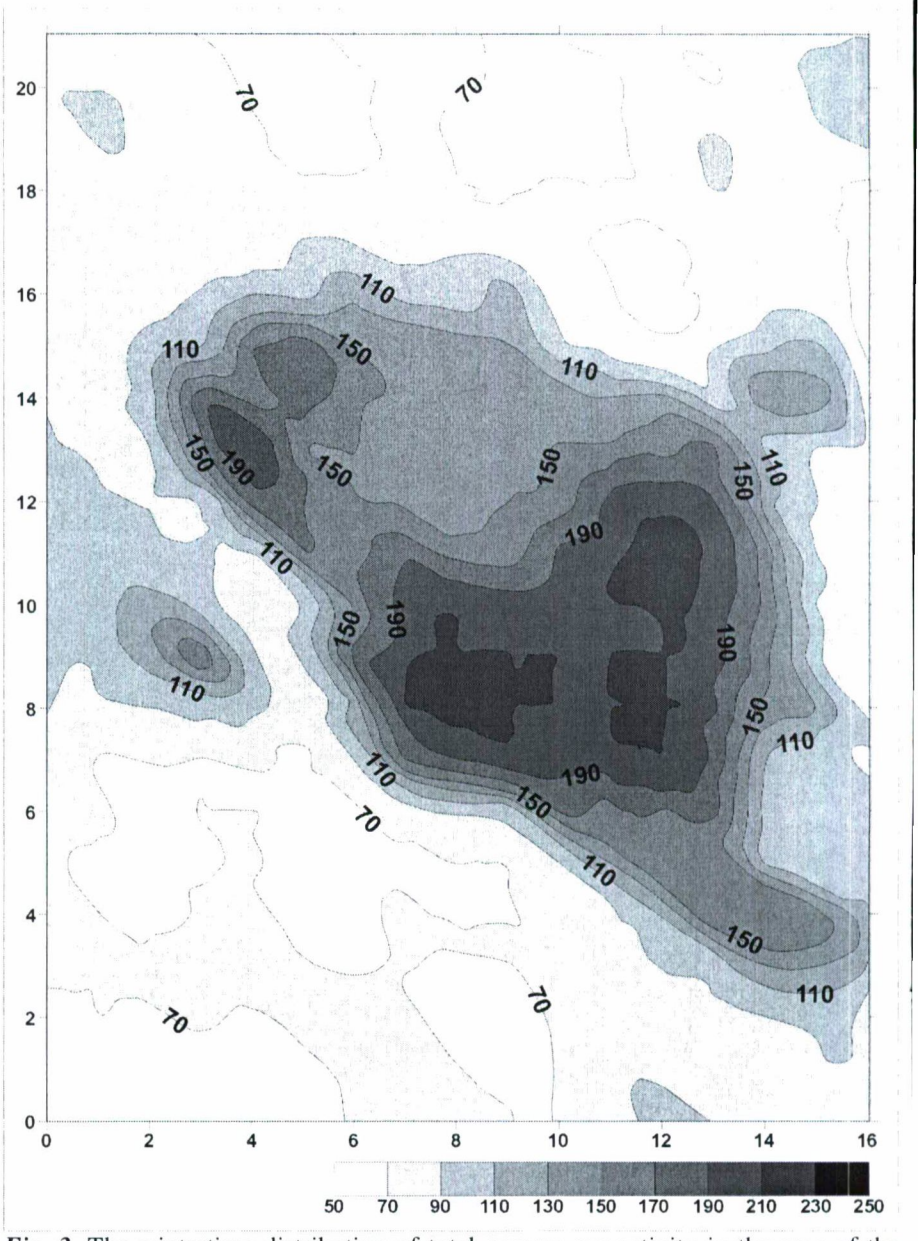

Fig. 3. The wintertime distribution of total gamma-ray activity in the area of the Karolina opencast mine

Gamma-ray spectroscopy

Productive layers occurring on the area of the opencast mine and adjacent waste sandstones and aleurites were all sampled for gamma-ray spectroscopy.

The measurements have proved that not exclusively the concentration of U, which shows an average value, is responsible for increased total gamma-ray activity but also the unusually high

Table 2. Calculated concentrations of radioactive elements in different rock types of the Karolina open cast

Rock type	No. of samples	Mean TOC (%)	Specific activity (Bq/kg)			Average radioactive element concentrations (gamma-spectroscopy)			Average radioactive element concentrations (ICP-MS)			
			mean	min.	max.	U (ppm)	Th (ppm)	K %	No. of sampl.	U (ppm)	Th (ppm)	K %
Coal	12	77	276	141	396	6	13	1,7	6	4,0	13	1,9
Argillaceous coal	14	61	298	198	387	5	21	2,2	5	4,9	15	2,3
Carbonaceous claystone	12	46	312	205	497	6	22	2,3	5	5,3	16	2,2
Aleurolite	6	17	216	149	326	3	15	2,0	4	3,8	12	2,1
Sandy aleurolite	7	16	178	123	213	4,5	14	2,2	5	4,2	9	2,2
Sandstone	8	6	148	111	245	3,5	12	2,0	5	2,7	10	2,1

Table 2. continued

Measuring points	Gamma-ray dose rate (nGy/h)	Calculated concentrations of elements		
		K (%)	Th(pp	U(pp
Soil cover	69	1,8	3	1,5
Sandstone (deposit 7)	101	2,2	8	4
Sandstone (deposit 21)	123	2,3	10	2,5
Aleurolite (deposit	201	2,4	18	7
Aleurolite (deposit 7)	156	2,2	16	5,5
Coal (deposit 11)	181	1,8	29	8
Coal (deposit 23)	169	2	18	7
Coal (deposit 25)	212	2,1	27	12

– compared to the world average – Th concentration of coals and K-content of argillites, aleurites and sandstones (Table 2.). Uranium and thorium content can mainly be related to organic material rich coals and carboniferous claystones. However, higher Th concentrations can also be detected in rocks of relatively low organic material content, but these samples proved always to be rich in clayey minerals and their grain-size was also small. Thus, the concentration of radionuclides (U, Th) is highly determined by clayey minerals, i.e. those samples were representing the highest specific activity that contained both organic material and clayey minerals in a significant quantity (carboniferous claystones, clayey coals). In case of certain samples the Th/U ratio can even be 7-9 (Table 2).

Further analyses would be necessary to determine the radiological characteristics of the clinker produced in the power station of Pécs, which is fuelled with the coal of Pécsbánya. Here, even higher values can be expected, since radioelements may further concentrate in the ashes of the burnt coal.

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