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Modified natural radiation exposure as a result of coal used to produce electrical energy

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MODIFIED NATURAL RADIATION EXPOSURE AS A RESULT OF COAL USED TO PRODUCE ELECTRICAL ENERGY

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ABSTRACT

During 1983-1993 we have assessed the activity concentration of natural radionuclides in air, soil, vegetation, snow and water around ten representative coal-fired power plants in Romania.

The average activities of Radon- 222 and of Radon-220 were 8-10 times higher than the repord\ted values in UNSCEAR 1998 Report.

The concentration of Uranium, Radium, Thorium, and Potassium were found to be higher in the upper 5 cm layer than in the 5-15 cm layer of soils in the surroundings of the coal-fired power stations, especially for those "old" and low filter efficiencies.

Snow, which can be expected to have a low background concentration of the natural radionuclides considered, has had Radium-226 concentration which ranged from 7 up to 70 mBq/kg.

The vegetation samples showed somewhat higher local activities for all four radionuclides with the highest Radium-226 content.

The found natural radioactivity levels due to Uranium-238, Radium-226, Thorium-232, Potassium-40 in surface water are generally comparable to those found in the other zones of Romania.

The mass of 384 x 10^6 kg fly-ash annually released to the athmosphere from these ten coal-fired thermal power plants is considered to be the main cause of disturbance of naturalradiation background, since the natural radionuclide concentrations in fly-asj are significantly higher than the corresponding concentrations in the earth's crust.

INTRODUCTION

It is well known that al types of coal, like most materials found in nature, contain trace quantities of the naturally-occurring primordial radionuclides. therefore, the combustion of coal results in the release to the environment of some natural activity which can modify ambient radiation fields and population radiation exposure.

In view of the importance of coal for generating electricity in Romania, this study dealt with the radiation exposures arising from its use to produce electric power.

With that end in view we selected ten representative modern and older coal-fired power plants with a totalinstalled capacity of 2.5 GW.

MATERIALS AND METHODS

Environmentalradiationand radioactivity monitoring have become since 1983 almost a routine programme on and around ten coal-fired power plants (CFPP's) from the country. Seven coal-fired power stations, Holboca (Iaşi II), Doiceşti II, Mintia, Oneşti II, Suceava, Timişoara, Vaslui, are relatively modern ones and are equiped with the electrofilters allowinf an ash retention of 96-99.5 %. The other

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three, Comăneşti, Doiceşti I, Ovidiu II are older ones and are equipped with multicyclones for which the efficiency of retention of the fly-ash particles is about 60-80 %. In 1993 Comăneşti plant, the poorest omong old plants, was modernized and equipped with electrofilters allowing an ash retention of 99 %.

Within the framework of the study measurements were performed of natural radioelements concentrations in coal wastes (ash and slag) and environmental samples, using gamma ray spectrometry techniques and radiochemical methods [1, 2].

Soil, vegetation, snow, surface water, atmospheric deposition and air samples have been collected at 20 locations from the environment of each thermal power plant within a 5 km radius from the power stations. The soil (top 5 cm and 5-10 cm) vegetation and snow samples were collected from 400 sq cm surface.

All the solid samples were dried at 105 °C and after grinding were passed through 12 mesh sieves. The sieved samples were placed inthe plastic Marinelli beaker and stored for 30 days to allow build up and reach radioactive equilibrium of Radon and its daughters.

cumulated wet and dry atmospheric deposition were sampled monthly and analysed seasonable weathers. These samples and vegetation samples were dried, calcinated and wet ashed. Arisingacid extracts were utilized for the assay of natural radioelements: Uranium-238, Thorium-232, Radium-226, Polonium-210 and Potassium-40.

Uranium-238 and Thorium-232 were calculated after determining the content of natural Uranium and Thorium by the method based on their separation and purification on a strong basic anion exchange resin and spectrophotometric measurements inthe form of their Arsenazo III complex. Radium-226 was determined through its decay descendant Radon-222, and by alpha ray measurement in a scintillating chamber. The assay of Polonium-210 was done by electrochemical deposition and alpha counting of Polonium-210 deposited on the nickel disc in a low background ZnS (Ag) scintillation counter. Potassium-40 was found by calculation after the photometric dosing in flame emission mixture of potassium natural isotopes.

Air was sampled every day at the same time for a week at 160 cm above the soil. the ^{222}Rn daughters concentrations in air have been measured and the equilibrium equivalent rRadon concentration (EEC, Bqm^{-3}) have been calculated. The method involves pumping a known volume of sample air through an open-faced, high-efficiency filter paper at a known flow rate for a certain collection time. The filter is removed and the deposited activity, assumed to be due to Radon daughters, is counted with a ZnS alpha scintillation counter during a certain period of time.

RESULTS AND DISCUSSION

The redistribution of natural activity by coal burning is presented in figure 1. There is approximately an order of magnitude enhancement of the activity concentrations from coalto ash, which can be characterized by the enrichment factor (EF). Among the radionuclides the most enriched was ^{238}U (EF: 0.9-3.4), followed by ^{226}Ra (EF: 1.1-2.64) and ^{232}Th (EF: 1-1.8).

The natural radionuclide concentrations in fly-ash are significantly higher than the corresponding concentrations in the coal. The enrichment of natural radioelements in fly-ash increases as the size of the fly-ash particles decreases [7, 8]. These smaller particles are less efficiently collected by the filtering systemand thus preferentially escape from the plant, falling out in the environment.

As a result of the activity deposited, we found the higher concentration of Uranium, Thorium, Radium-226 and Potassium-40 in the upper 5 cm layer than in the 5-15 cm layer of soils in the surroundings of the coal fired power stations (CFPPs), especially for the older ones. Therefore, the absorbed dose rate in air 1 m above the soil (from terrestrial gamma radiation) were increased by comparison with other counties zones without coal fired power plants. Table 1 gives a comparison of the natural radioactivity levels at these two soil layers.

It was obvious that the doses were highest at locations 1-2 km from the stack as well as on the grounds of the CFPP in the direction of winds.

The vegetation samples showed higher local activities for all the four radionuclides with the highest Thorium-232 content (Table 2). These values of the natural radionuclide concentrations are much greater than those found in the other areas of Romania where the radioactive background is moderate [5].

Snow, which can be expected to have a low background concentration of the natural radionuclides considered, has had great values for Radium-226 concentrations, as direct results of CFPPs emissions (Figure 2).

The foundnatural radioactivity levels due to Uranium-238, Radium-226, Thorium-232 and Potassium-40 in surface water (Table 3) were situated between the limits of variation of concentration values determined in Romanian surface water [3]. There were some what higher local activities for Potassium-40.

Measurements in surface air carried out during ten years ago, had clearly shown the presence of enhanced concentrationd of up to 44 Bq/m^3 of ^{222}Rn around the plants, compared with the "normal" concentrations of about 8 Bq/m^3 in Romania. However, recent measurements have failed to reveal a significant increase in the surface air Radon concentrations [4].

The quantity of monthly deposited powders exceeded in many places the admissible maximum quantity (AMQ) in Romania of 17 g/m^2 [6]. the concentrations of natural radionuclides were higher along th e most frequent wind direction. Differences in values of concentrations measured between the spring-summer and autumn-winter seasons are a consequence of a difference in the intensity of energy production of CFPPs and repair work usually carried out in the summer. The activities discharged in the spring-summer period are smaller than what they used to be in winter.

The athmospheric dischaarges of national radionuclides from CFPPs, and consequently the surrounding natural radioactivity levels depends heavily on the efficiency of the particulate control, the coal quality and ash content, and the tall of the stack. The taller stacks result in a much greater atmospheric dilution [8, 9]

CONCLUSIONS

1 The measurements in air, soil, snow and vegetation had clearly shown the presence of increased concentrations of the natural radionuclides in the surrounding of the coal-fired thermal power plants, particularly for the oldest and most poorly controlled ones. Therefore, there is an increase of the basic radiation rate in the neighbourhood area and higher exposure of the local population to radiation.

2 The mass of fly-ash annually released to the atmosphere from coal-fired power plants is considered to be the main cause of disturbance of natural radiation background.

3 It is necessary to devote much more attention to environmental pollution with radioactive material from coal-fired power plants in both research and scientific work as well as to lawful regulating of the technologically induced increase of natural radioactivity.

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Table 1 The activity cconcentrations (Bq/kg) of natural radionuclides in soiland annual gamma dose rate (mGy/y)

soil	CFPPs	^{238}U	^{226}Ra	^{232}Th	^{40}K	Gamma dose rate
The upper 5 cm layer	modern	9.6-74	8-142	11-85	412-1015	0.55
	old	22-86	23-156	17-96	496-1200	
The 5-15 cm layer	modern	1.4-54	11-94	0.7-36	329-840	0.43
	old	17-59	20-57	6.6-47	371-800	

Table 2 The natural radionuclides concentrations (Bq/kg) in the spontaneous vegetation

		^{238}U	^{226}Ra	^{210}Po	^{232}Th	^{40}K
CFPPs	Mean	25.5	23.4	27	37	451
	Range	0.06-61	0.7-40	0.07-74	1.5-83	220-551
Romania	Mean	-	2.5	-	2.2	505
	Range	-	1.8-3.7	-	1.6-3.5	350-640

Table 3 the concentration range of the natural radionuclides inthe surface water (mBq/l)

	^{238}U	^{226}Ra	^{210}Po	^{232}Th	^{40}K
CFPPs	1.9-11.2	2.4-10.3	0.8-8.5	1.6-8.9	76-655
Romania	0.8-18.5	2-20	-	1.5-18	25-670

Figure 1 Mean values of specific activity (Bq/kg) in the coal,slag and fly-ash

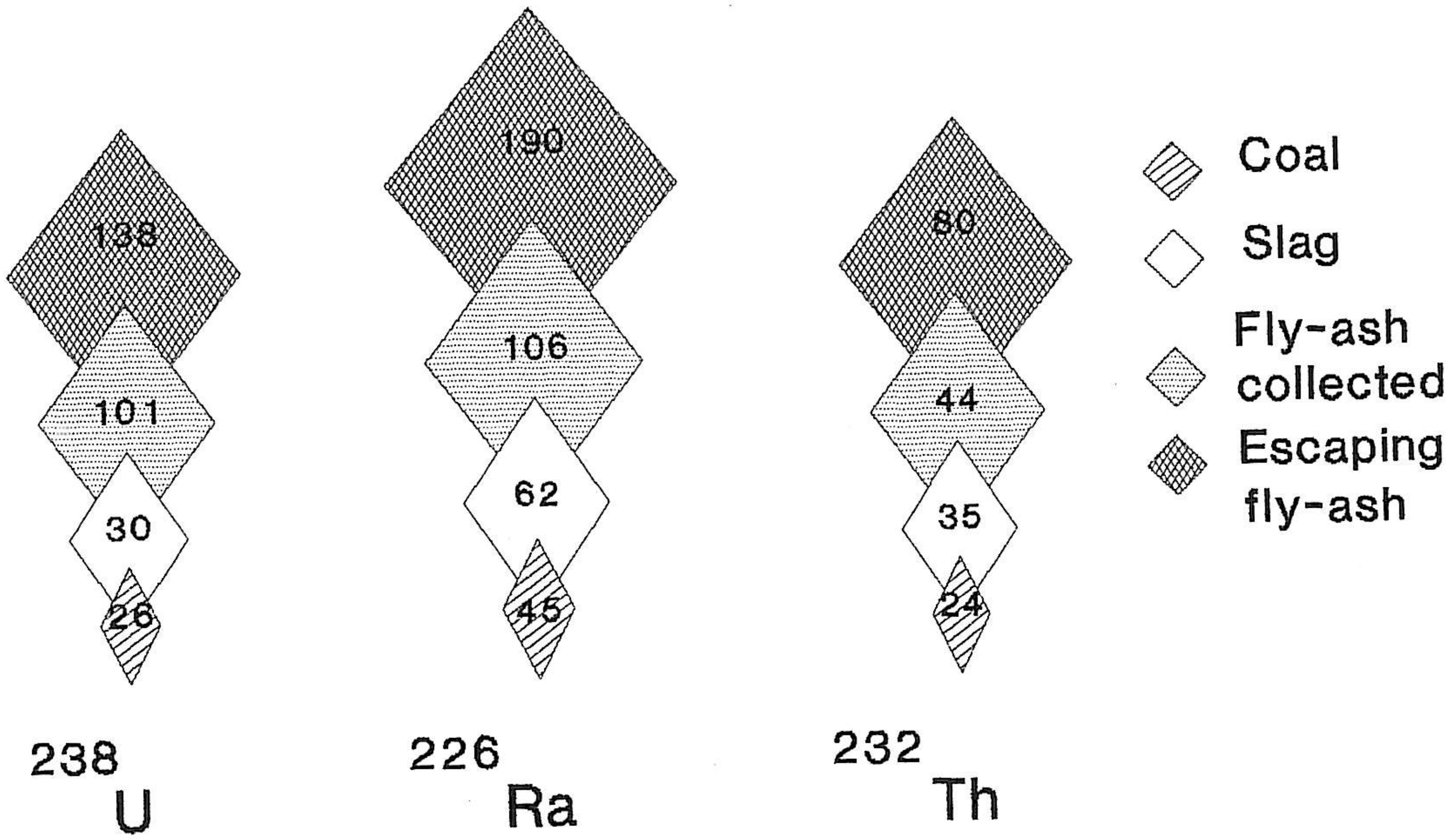

Figure 2 The concentrations of the natural radionuclides in snow around two coal fired power plants

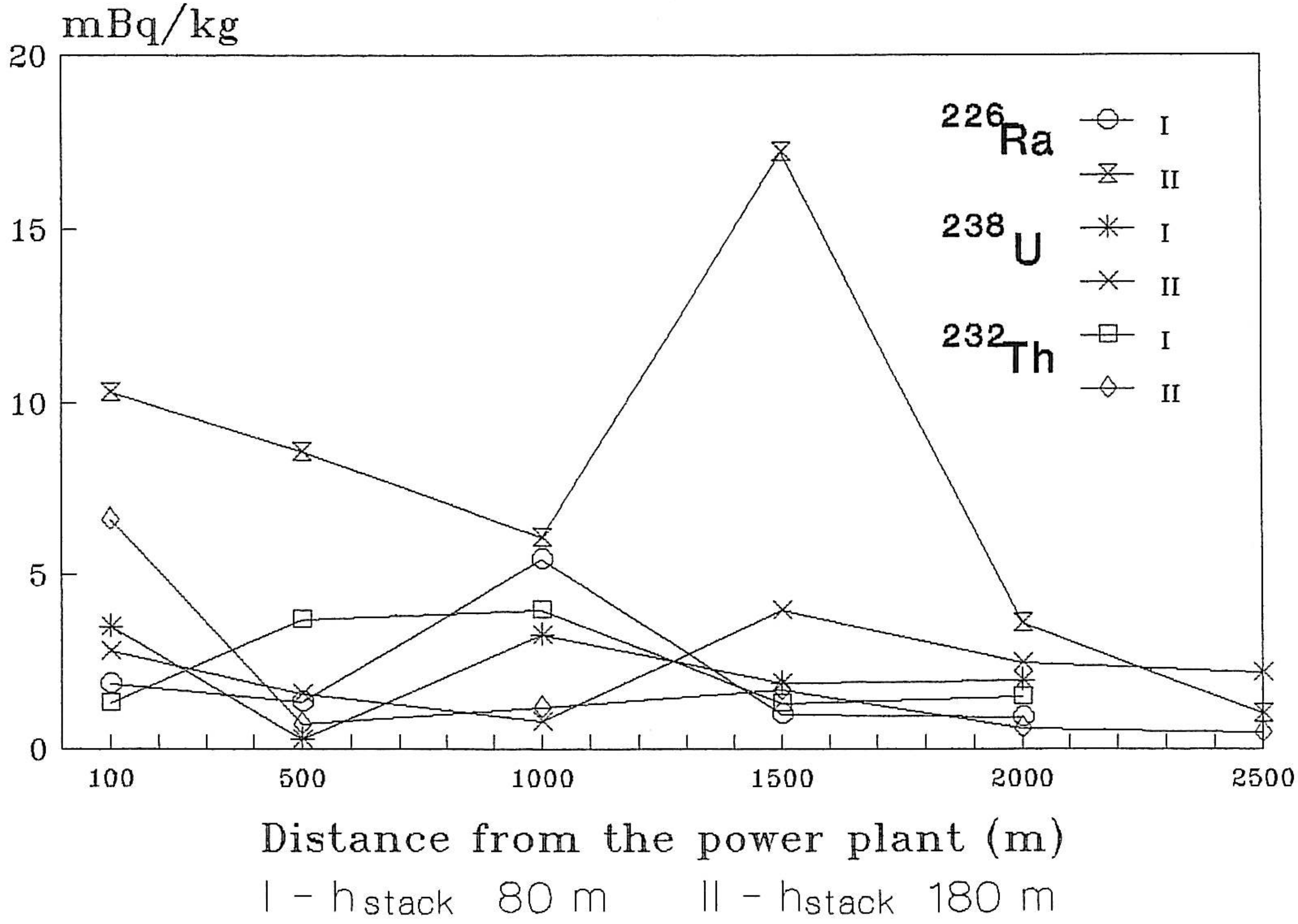

Figure 3. The natural radioactivity of the atmospheric deposition