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SimTerm2022 PROCEEDINGS

20th International Conference on Thermal Science and Engineering of Serbia

Niš, Serbia, October 18-21

ENERGY EFFICIENCY ECONOMY ECOLOGY

SimTerm2022

PROCEEDINGS

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Quality Control of Soil and Water in the Vicinity of Coal Fired Power Plants – Radiological Aspect

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Abstract: The operation of coal fired power plants greatly influences the surrounding environment, especially water and soil, due to large amounts of combustion waste products released. Besides the quality of soil and water from the aspect of chemical and structural composition (release of heavy metals, ash and slag deposition on soil) it is of at most importance to maintain a strict control from the radiological safety point of view. All coal types contain certain amount of naturally occurring radionuclides, which depends on the composition of the rock and soil adjacent to the place of the coal excavation. These radionuclides are concentrated in the process of coal combustion, thus leading to potentially high levels of radioactivity in the ash and slag. These by – products are released into the environment and can increase the radiation burden on the environment, change the composition of the soil and bodies of water in the vicinity, thus potentially influencing the health of the general population. In this paper, the radiological analysis of soil and water samples taken from the vicinity of different coal fired power plants in Serbia in the period of 2019-2021. will be presented. Measurements are performed in the Radiation and Environmental Protection Department of Institute of Nuclear Sciences Vinča within the framework of regular environment monitoring of the power plants.

1. Introduction

Radionuclide and heavy metals contamination of the environment is one of more prominent problems of today's modern society. Due to the rapid development of the industry and large number of different technological processes applied, large quantities of waste and by – products of heavy industry, mining and energy industry find their way into the environment. It is noticeable that the degradation of the soil quality is caused not only by expanding of the urban areas, building of the industrial complexes and roads, but also by discarding of the waste, which leads to exposing large areas of soil to the pollution and erosion. The major part of the radionuclide content which can be found in the soil, originates from the basic substrate of the Earth's crust surface. Naturally occurring radioactive materials (NORM), containing natural radionuclides of the uranium and thorium series and potassium – 40 are considered as a natural level of radioactivity in the environment [1]. Many industrial processes as a result produce materials with technologically enhanced natural radioactivity, which means that the radionuclides present in the raw materials are concentrated in the final product or the waste. Also, the water basins which lie beneath those areas and other water bodies in the vicinity are also exposed to the same risk of pollution. All these situations can have, as a consequence, the elevated exposure of the public to the ionizing radiation [2]. This is the reason why potential pollutants have to be identified and the quality of the soil and water monitored, in order to take measures for preventing the contamination of the environment and public exposure to the ionizing radiation.

Coal fired power plants are identified as a potential source of contamination of the soil and water with naturally occurring radionuclides. Combustion of the coal in power plant results in a redistribution of various pollutants such as sulfur dioxide, nitrous oxides, carbon monoxide, toxic and heavy metals, organic particles and radionuclides of uranium and thorium series and ^{40}K. Every type of coal contains a certain amount of uranium and thorium as well as their daughters. During combustion, these radionuclides become concentrated, meaning that their concentration in combustion products can be many folds larger than that of the coal itself. The content and the concentration of radionuclides in the ash and slag depends on the series of factors such as type and characteristics of the coal, percentage of the ash in coal, caloric value of the coal, the temperature of the combustion, chemical and physical form of the present radionuclides etc [3]. The change of natural radioactivity that is a consequence of the operation of coal fired power plants can influence the food chain soil and water – plants – animals – human. Due to this, besides the quality control of soil and water from the aspect of chemical and structural composition (release of heavy metals, ash and slag deposition on soil) it is of at most importance to maintain a strict control from the radiological safety point of view. This implies the systematic control of radioactivity in the soil and water in the vicinity of the power plant with the aim to estimate the influence and the increase of the radiation burden on the environment and the population [4].

The control of the radioactivity in working and living environment in the vicinity of the coal fired power plants is regulated by the Rulebook on the radioactivity monitoring (Official Gazette 97/2011), which states the method and conditions of the systematic investigation of the radioactivity in the environment [5]. Based on this Rulebook and the contract between Public Enterprise "Elektroprivreda Srbije" and Institute of Nuclear Sciences Vinča, systematic long term measurements of different samples were conducted in order to assertain the exposure of public and working force in power plants to the ionizing radiation.

In this paper, the radiological analysis of soil and water samples taken from the vicinity of "Nikola Tesla"A i "Kolubara" coal fired power plants in Serbia in the period of 2019-2021. will be presented. Measurements are performed in the Radiation and Environmental Protection Department of Institute of Nuclear Sciences Vinča within the framework of regular environment monitoring of the power plants.

2. Measurement Method

2.1 Sampling

Sampling locations are defined in the Program of the radioactivity control in working and living environment for 2 power plants, TE "Nikola Tesla" A i TE "Kolubara", and are the same in the several years in which the control was conducted. Locations are chosen in such a way that they cover the immediate vicinity of the power plant, where the influence of the power plant operation would be most prominent. Also, the sampling was conducted in the remote locations, mainly in the populated areas, to ensure the knowledge of the local natural levels of radionuclide contents.

Each year, total of 14 soil samples were taken at different locations surrounding the TE "Nikola Tesla" A, 2 at the immediate vicinity and 12 at different distances from the power plant and 4 samples at different locations surrounding the TE "Kolubara", 1 at the immediate vicinity and 3 at remote locations. Before the soil samples were taken, the surface of the soil was cleared of vegetation, pebbles and other debris that were present in the upper layer of the soil. The sampling was performed using spade and shovel. First, using a spade, a square incision in the soil was made and then the upper layer of the soil, to the depth of 5 cm was collected using shovel. For each sampling location, an amount of about 1 kg of soil was taken. Samples were then placed in plastic bags and properly labeled.

Total of 5 samples of water from TE "Nikola Tesla" A and 5 samples from TE "Kolubara" per year were taken also. Overflow and drainage water samples were taken at every ash landfill. Also, the samples of drinking water, as well as water from river Sava and Kolubara were collected upstream and downstream from the each power plant. The sampling of overflow, drainage water and river water was performed by submerging the sampling vessel into the water. The sampling of the drinking water was performed at the faucet where it is normally used for drinking. The amount of each sample was 15 l, sufficient for performing all the required analysis.

2.2 Gamma spectrometry

The preparation of the soil samples consisted of cleaning of the bulk sample from remnants of plants, pebbles and other debris, drying at the temperature of 105 ºC, sifting and measuring the aliquot of the bulk sample into the measurement geometry. For soil samples, it was a Marinelli beaker of 0.5 l. This procedure was done in accordance with the method recommended in IAEA Technical Report Series No.295 – Measurement of Radionuclides in Food and the Environment - Section 5. Collection and Preparation of Samples-page 27 (5.2.3 Soil) [6]. The beaker containing the sample is then sealed with bee wax and left in the laboratory for at least 28 days in order to achieve the secular radioactive equilibrium between radon and its daughters.

Te preparation of the water samples begins with the evaporating of the bulk sample to dry under an infrared lamp. The dry residue is then ashed in the oven at the temperature of 450 ºC. This ensures that all the organic components still present in the residue are destroyed and all that is left is the mineral content of the liquid sample. The ashed residue is then transferred into the 100 ml plastic cylindrical container and can be measured immediately. This procedure is also in accordance with the [6].

The measurement is performed on three semiconductor High Purity Germanium (HPGe) detectors with relative efficiencies of 18%, 20% and 50%. The resolution of detectors is 1.8 keV at the energy of ^{60}Co (1332 keV). Efficiency calibration of the instruments for soil samples measurements was performed using a certified radioactive standard in Marinelli beaker, procured from Czech Metrology Institute Praha, Type CBSS 2, Cert. No.1035-SE-40845-17, total activity of 80.63 kBq at the date of 22.12.2017. It contained a mixture of radionuclides (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{51}Cr, ^{210}Pb), whose energies span from 46 keV to 1900 keV, thus covering the whole measurement range. For water samples, the efficiency calibration was performed using an inhouse made radioactive reference material, produced by spiking the ashed biota with the certified radioactive solution, procured from Czech Metrology Institute Praha, Type ER X, cert no. 1035-SE-40844-17, ER X, total activity of 79.89 kBq at the date of 22.12.2017. It also contained the same radionuclides as the standard in Marinelli beaker.

The measurement of all samples lasted for 60000 s and the spectra were analysed using GENIE2000 software by Canberra. Detected radionuclides were naturally occurring radionuclides of uranium and thorium series (^{238}U, ^{235}U, ^{226}Ra i ^{232}Th), naturally occurring ^{40}K and artificial radionuclide ^{137}Cs, present in the environment since the nuclear accident in Chernobyl.

The activity concentration of gamma emitting radionuclides, expressed in Bq/kg was calculated according to the following equation:

$$A = \frac{N}{t \cdot m \cdot \varepsilon \cdot P_\gamma} \qquad (1)$$

where N represents the number of detected photons at given energy with the background radiation subtracted, t is the measurement duration in seconds, m is the sample mass, ε is the detection efficiency at given energy and P_γ is the photon emission probability.

The measurement uncertainty of the results is expressed as an expanded measurement uncertainty with the coverage factor $k=2$, which, for the normal probability distribution, correspond to the 95% level of confidence. The expanded measurement uncertainty was determined using the following equation [7]:

$$u(A) = 2 \cdot \sqrt{(\delta N)^2 + (\delta \varepsilon)^2 + (\delta m)^2} \qquad (2)$$

where δN is a relative measurement uncertainty of the number of detected photons, $\delta\varepsilon$ is the relative measurement uncertainty of the measurement efficiency and δm is the relative measurement uncertainty of the sample mass. Contributions to the measurement uncertainty originating from the measurement duration and the emission probability are negligible. For all detected radionuclides, the measurement uncertainty was of the order of magnitude of 5%, except for ^{238}U and ^{235}U, where the uncertainty was significantly greater in some cases.

Based on the measured activity concentration in soil samples, the absorbed dose rate was calculated and based on this result, the annual effective dose originating from the radionuclides in soil was determined. The absorbed dose rate was calculated using the following equation [8]:

$$\dot{D}(nGyh^{-1}) = 0.462 \cdot A_{Ra} + 0.604 \cdot A_{Th} + 0.0417 \cdot A_K \qquad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of radium, thorium and potassium respectively.

Based on this result, the annual effective dose was calculated using the following equation [8]:

$$D_E(mSv) = 0.7SvGy^{-1} \times 0.2 \times 365 \times 24 \times \dot{D} \qquad (4)$$

Also, the $^{235}U/^{238}U$ ratio was determined. Since the operation of the coal fired power plant does not imply any process in which the uranium is enriched or depleted, it should not differ significantly from the natural ratio of 0.046 [8].

2.3 Gross alpha/beta measurement

For gross alpha/beta measurement, conducted for the water samples only, preparation of the samples begins with the evaporation of the 3 l of bulk sample to dry. After that, the dry residue is ashed at 450ºC. The mass of the remaining ash is then measured on the analytical scale and transferred to a stainless steel planchette, thus obtaining a thin sample adapted for the alpha and beta counting. The preparation and measurement of the samples is conducted according to the method defined in EPA [9].

For gross alpha and gross beta measurements, the samples were measured at gass proportional counter THERMO-EBERLINE FHT 770T. The efficiency for gross alpha counting is 26 %, and for gross beta measurement is 35 %. The calibration of the instrument was performed using certified point sources of ^{241}Am for alpha and ^{90}Sr for beta counting. Both point sources were produced by Czech Metrology Institute Praha. The ^{241}Am point source was Type EM 445, serial no. 160311-1099079 with the total activity of 224 Bq and surface emission rate of 106.5 s^{-1} at the reference date 01.08.2011, and ^{90}Sr, Type EM 145, serial no. 280211-1059179 had the activity of 189.4 Bq and surface emission rate of 249.5 s^{-1} at the reference date 01.08.2011. Measurement time was 14400 s and samples were measured in series of 3 measurement per sample.

3. Results and Discussion

The range of activity concentration of naturally occurring radionuclides in soil samples, as well as the artificial radionuclide ^{137}Cs for thr period of 2019-2021 are presented in Table 1. Also, in Table 1, maximal values of measured activity concentrations in soil samples taken at the vicinity of the power plant and on the remote locations, are presented.

As it can be seen from the Table 1, the range of measured activity concentrations is within the range of values measured in other parts of Serbia and values stated in the literature [8, 10]. In the case of naturally occurring radionuclides, maximal values obtained for the samples taken both in the vicinity of the power plant and on the remote locations are approximately equal. This means that the influence of the operation of the power plant can not be noted in the measured soil samples. In the case of ^{137}Cs, the maximal valuo of 48 Bq/kg was measured at the location of Veliki Crljeni, in the vicinity of the power plant "Kolubara" in 2020. The variations in the activity concentration of this radionuclide are expected in soil since its origin is a consequence of the nuclear accident in Chernobyl 1986 and is not linked with the processes in the regular operation of coal fired power plants. At the same location in previous years, as well as on the other locations in the investigated period, there was no high activity of ^{137}Cs detected.

Based on measured activity concentratios in soil samples, the ratio ^{235}U/^{238}U was calculated. This ratio ranged from 0.034 to 0.075. Taking into account measurement uncertaintites of the results used for obtainig this ratio, it can be concluded that the natural ratio of these two isotopes of uranium does not differ significantly from the natural ratio of 0.046, therefore leading to the conclusion that the natural ratio of uranium is not disturbed in the measured soil samples.

Table 1. Maximum and minimum values of the activity concentration for all radionuclides and maximal values detected in siol samples taken in the vicinity of the power plant and on the remote location. The values were obtained in the period of 2019. to 2021.

Element	^{226}Ra	^{232}Th	^{40}K	^{238}U	^{235}U	^{137}Cs
Minimal value for all samples [Bq/kg]	20	18.6	294	18.5	0.73	0.67
Maximal value for all samples [Bq/kg]	65	62	700	72	4.7	48
Maximal value obtained in the vicinity of power plant [Bq/kg]	65	57	480	72	4.7	48
Maximal value obtained at remote locations [Bq/kg]	53	62	700	61	3.6	28

Table 2 presents calculated annual effective dose on all locations for the period of 2019-2021.

As it can be seen from Table 2, there are no significant differences of the annual effective dose at different locations. Maximal annual effective dose at the locations in the vicinity of the power plant was 0.101 mSv, while the maximal value obtained for the remoted locations was 0.105 mSv. It is defined in the Rulebook on Limits of Exposure to Ionizing Radiation and Measurements for Assessment of the Exposure Levels (Official Gazette RS 86/11 and Official Gazette RS 50/18) that the maximal exposure of the population to the ionizing radiation should not exceed 1 mSv per year [11], we can see that the contribution to this valuo that originates from the operation of the investigated power plants ammounts to about 10%. This contribution is not considered as a high.

Table 2. Annual effective dose on all locations for the period of 2019. to 2021.

Location		Annual effective dose [mSv]	
		2019	2020/21
In the vicinity of TE "Nikola Tesla" A	Topolice 1	0.075	0.052
	Topolice 2	0.082	0.077
Remoted locations from TE "Nikola Tesla" A	Urovci 1	0.075	0.089
	Urovci 2	0.105	0.067
	Krtinska 1	0.078	0.086
	Krtinska 2	0.097	0.084
	Rvati 1	0.071	0.092
	Rvati 2	0.069	0.083
	Obrenovac 1	0.073	0.067
	Obrenovac 2	0.078	0.07
	Obrenovac 3	0.079	0.067
	Obrenovac 4	0.089	0.087
	Zabrežje 1	0.101	0.086
	Zabrežje 2	0.04	0.061
In the vicinity of TE "Kolubara"	Veliki Crljeni	0.092	0.101
Remoted locations from TE "Kolubara"	Sokolovo	0.102	0.086
	Stepojevac	0.085	0.091
	Junkovac	0.099	0.103
Min		0.04	
Max		0.105	

The water samples were analyzed both using gamma spectrometry and gross alpha/beta counting.

Radioactivity in the water can originate from uranium and thorium series and their daughters and ^{40}K, similar to the origin of the natural radioactivity in the surrounding soil. According tho the legislative in Serbia, drinking water is prescribed for for gross alpha/beta and gama spectrometry measurements [12]. In the drinking water, the limit for gross alpha activity in drinking water is set to 0.1 Bq/l and for gross beta activity, to 1 Bq/l. For gamma emitting radinuclides, the Rulebook prescribes derived activity concentrations which ensure that the limit of exposure is not exceeded. In case of monitoring of the environment in the vicinity of coal fired power plants, drainage and overflow water, as well as water from the near by rivers were analyzed in addition to the drinking water.

Gamma spectrometry measurements revealed the presence of ^{40}K in all samples in the range from 0.04 to 0.39 Bq/l. Artificial radionuclide ^{137}Cs was detected only once in the sample of river Sava downstream from the power plant (0.005 Bq/l) and in one sample of overflow water in the power plant "Kolubara" (0.0028 Bq/l). In all other samples in the whole period, the activity concentration of this radionuclide was below the minimal detectable activity (MDA) which was from 0.001 to 0.004 Bq/l. The activity concentration of ^{226}Ra ranged from 0.035 to 0.24 Bq/l which is below the limit of derived activity concentration defined in [12] for drinking water. In all samples, the activity concentration of ^{238}U and ^{235}U were below MDA, which was of the order of magnitude of 0.05 and 0.005 Bq/l respectively. Radionuclide ^{232}Th was detected in one sample of the overflow water in the power plant "Kolubara" (0.013 Bq/l) which was also below the derived activity concentrations defined in [12].

In all samples of water for the whole investigated period, the gross alpha activity was below MDA which was between 0.04 and 0.1 Bq/l. From these results we can conclude that in no sample was the defined limit of 0.1 Bq/l given in [12]. Gross beta activity was found to be between 0.07 and 0.4 Bq/l in all samples for the wole period. All the results were below the limit of 1 Bq/l defined in [12]. It should be noted that al these limits are defined for drinking water only, but we assumed the same strict criterion for the samples of river water as well as overflow and drainage water.

4. Conclusion

This paper presents the results of gamma spectrometric measurements of soil and water samples as well as gross alpha/beta measuremetns in water samples. All samples were taken at the preordained sampling sites arround the coal fired power plants “Nikola Tesla A” and “ Kolubara” as a part of the control of the radioactivity in working and living environment in the vicinity of the coal fired power plants, regulated by the Rulebook on the radioactivity monitoring (Official Gazette 97/2011). The results analyzed in this paper covered the period of 2019-2021, although this type of control is conducted regulary for many years. The activity concnetrations of gamma emitting radionuclides in soil samples were within the range of values measured in other parts of Serbia and values stated in the literature. Based on these results, the annual effective dose was calculated and it can be concluded that it does not exceed approximately 10% of the recommended limit of 1 mSv per year. Also, the difference between the results obtained for the samples taken at the immediate vicinity of the power plant and on remoted locations is not observed. Also, the gamma emitting radionuclides in the water samples were mainly below the MDA. Few results that were abowe the MDA were however below the derived activity concentration defined in [12], ensuring that these samples are in accordance with the regulation defined for drinking water. Gross alpha activity was below MDA in all investigated water samples and below the defined limits. Gross beta activity ranged between 0.07 and 0.4 Bq/l ensuring that the criterion for beta activity in drinking water was also satisfied.

Based on all presented results, we can conclude that the operation of these two coal fired power plants does not influence the quality of soil and water, from the radiation protection point of view.

It is important to maintain this kind of constant and planned environment monitoring because it provides a data base that can be used to observ any eventual change that would indicate the deterioration of the soil and water quality as a consequence of the power plant operation.

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