URANIUM AND RADIUM IN WATER SAMPLES AROUND THE NIKOLA TESLA B LIGNITE-FIRED POWER PLANT – OBRENOVAC, SERBIA

by

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Scientific paper
UDC: 543.42:662.64:628.112
DOI: 10.2298/NTRP1101011Ž

This paper deals with the analysis of natural radionuclide content in 23 water samples collected in the vicinity of the Nikola Tesla B thermal power plant, Serbia. All samples were analyzed for ^{226}Ra and uranium isotopes (^{238}U, ^{234}U) activity using radiochemical methods and alpha spectrometry. Obtained results show that the activity concentrations for uranium and radium in the water around the thermal power plant are low when compared to those from areas across Serbia with their enhanced natural uranium and radium content. No important radiological hazard related to uranium and radium activity stored in heap was found.

Key words: alpha spectrometry, lignite-fired power plant, activity concentration, radium, uranium

INTRODUCTION

The assessment of the environmental impact of radioactivity in wastes from coal production takes into consideration the direct hazards resulting from wastes in the immediate surroundings and the dispersal of radionuclides in the environment through surface and groundwaters. Over a period of three years (2004-2007), the 6th Framework European Commission International Cooperation Project arose as an Assessment of Environmental Risks of Radioactively Contaminated Industrial Tailings (INTAILRISK) [1], aiming at investigating wastes containing naturally occurring radioactive material (NORM) from coal-burning power plants in Western Balkan countries. The test sites in eleven participating countries, Serbia being one of them, have been defined in detail in terms of the radionuclide type, distribution and dispersion. The test site in Serbia was the Nikola Tesla B (TENT B) thermal power plant, including its tailing pond, its immediate surroundings, *i. e.*, villages and settlements, the facility itself, and its raw materials.

The TENT B area is located on the bank of the river Sava, near the town of Obrenovac, about 40 km upstream from the city of Belgrade. The power plant produces about 4.5 10^9 kg of coal-ash per year. The ash is transported to the dump after being suspended in water taken from the Sava in an approximate ratio of 1:10, while all water-surplus is drained back to the river. The fly-ash deposit of TENT B covers an area of about 6 km^2 and is located 4.5 km from the thermal power plant itself. Thus, the primary environmental concern associated with radionuclides coming from a field disposal site, in this case, is potential groundwater contamination [1, 2] Hydrochemical data of waters from wells and springs collected during the field survey of TENT B revealed high mineralization (1200 mg/l), a high content of SO_4^{2-} (up to 450 mg/l), as well as a high content of Cl^- (up to 70 mg/l), supporting the hypothesis of potential pollution from the fly-ash deposit [3].

The TENT B area has been characterized by an investigation regarding gamma dose rates, radon concentration in soil gas, indoor and outdoor radon concentrations, and radionuclide activity concentration in soil and waste materials which were considered in previous papers [4, 5]. The present paper focuses on the

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results of the investigation of the influence of fly-ash deposits on groundwater radioactivity. Namely, the problem of ash repository is considered as being a problem of disposal of low-level radioactive waste material [6, 7], because natural radionuclide concentrations in the ash and slag are higher than the corresponding concentrations in the Earth's crust, *i. e*., fly-ash deposits may be considered as a source of natural radionuclides which enable the migration of radionuclides of the uranium and thorium series through soil and groundwater [8].

Field work on water sampling in the vicinity of TENT B has been carried out in October 2004 and June 2006. All field surveys were carried out by a joint international team from the Electrochemical Etching Laboratory (ECE Lab) of the Vinča Institute of Nuclear Sciences, Belgrade, Serbia, and the Earth Science Department of the Sapienza University of Rome, Rome, Italy, while the radiochemical analysis of the water samples was done by the Laboratory of Radioactivity Analysis of the Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Sciences (IFJPAN), Krakow, Poland. This paper presents the results of the alpha spectrometric measurements on ^{226}Ra, ^{234}U, and ^{238}U in groundwater and their correlations.

GEOLOGICAL AND HYDROLOGICAL SETTING OF THE TENT B AREA

This chapter considers the ability of succeeding equilibriums of water levels in aquifers in the vicinity of TENT B, determining mainly the status of natural radionuclides.

TENT B surroundings are composed of quaternary river sediments consisting of loam and argillaceous sandy sediment. These lithological components usually lie across the river terrace sediments and the clay marl, carbonic clay, diatomaceous earth, and sands of the Lower Pliocene (Pontian). The sediments are concordant, usually without any tectonic disturbances. The only significant tectonic structure (south from TENT B) is the fault that stretches in the direction of E-NE, close to the villages of Dren and Grabovac. The presence of this fault was detected thanks to photogeological and satellite photos. A deep borehole was drilled in the village of Vukićevica for geological and stratigraphic purposes [9]. Moreover, shallow wells reaching a depth of about 25 m are present over a wide area, enabling us to have a geological insight of the region. The geological and hydrogeological profile of the Obrenovac area shows the presence of a main sandy gravel aquifer between two impermeable formations of shallow alluvial clay sediments and the Miocenic marly clay (fig. 1). The groundwater flows in the NW direction toward river Sava. According to the results obtained from the deep borehole drilled for stratigrahpic purposes in Vukićevica, at this location the first water horizon is situated in the fine grain quartz sand at depths ranging from 23 m to 33.5 m or at the absolute altitude of 97 m to 107 m. Below this series of rocks, clay sediments are dominant up to 273 m where limestones occur. Near the surface, a thick series of dark diluvial loam is present, further below, fine grain yellow and grey dusty quartz sands with quartz gravel occur. Surface flows that are formed here have a very small dip causing swamp terrain. Clays, which are near the surface at the depth of 4-5 m, make the water sustainable at ground level. As a result, the terrain is covered with numerous swamps and ponds. Some channels were dug for drainage, but basically the terrain has retained its pond and peat characteristics. One of the channels is the Vukićevica channel which gathers waters from Grabovac and takes them north, to the Sava.

In this region, the aquifers are formed of sands and gravel and are of the freatic type. Since impermeable clay lays a top of them, they are not fed with rain water. Due to this, changes in the water level are small, about 1m up or down. In the period of intensive rain, the terrain becomes wet and the water stays and gathers in natural depressions, creating ponds and swamps. Such occurrences are frequent on the Grabovac-Vukićevica terrace. Shallow surface flows gather water and take it away. In dry periods, the clay dries out and fractures appear taking in a certain amount of water in the initial rainy period, so some water manages to flow into the aquifers.

The freatic aquifers on the higher terrace are of small thickness, but have a large surface. Depth levels at various points of the terrain differ, but on the terrace

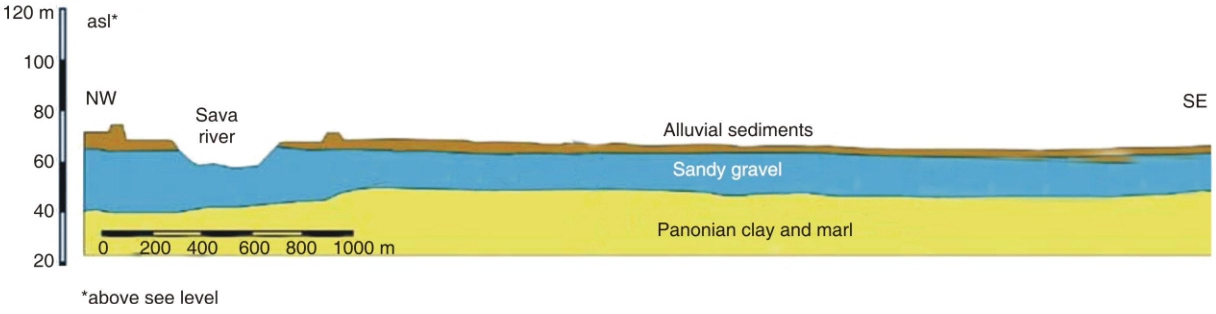

Figure 1. Geological-hydrogeological profile of the TENT B area

itself, the depth usually varies between 18 to 22 m, sometimes up to 25 m. This is the case with the wells in the villages of Vukićevica, Trstenica, Orašac, Dren, and Grabovac. Water comes from coarse grained sands and gravels by force of gravity or is carried upward by gas bubbles. At the same altitude, in the alluvial plane of river Vukićevica in the village of Orašac, a lot of springs are located.

It is obvious that this is the place where the aquifer, formed in Vukićevica, discharges. In Grabovac, at the altitude of 95 m to 100 m, there are numerous springs emptying the Grabovac aquifer. They are on the same altitude and usually not very strong. The terrain in their immediate vicinity is wet and water diffuses from the ground. The water from this freatic aquifer flowing out through the springs is directed to the main channel in the village of Grabovac located on the edge of the terrace and then directed to the NW and Sava river. With this channel, surface waters are taken away from this part of the terrain. The level of the aquifer is at the depth of 22-24 m in the area of Vukićevica and at a depth of 18 m to 21 m in the Grabovac area. Water temperature in the aquifers at these locations is 13.5 °C to 14.5 °C. Considering the chemical composition of underground waters in the area of Ušće, Orašac, Vukićevica, Ljubinić, Trstenica, and Dren, most of them have mineralization that ranges between 0.5 to 1 g/l. Less mineralized waters are in the area around the villages of Vukićevica and Trstenica. The levels of underground water on the lower terrace in the vicinity of villages Ušće, Skela, Grabovac, and Ratari are much closer to the surface than those on the higher terraces. Their water levels are at a depth of 0.00 to 4-6 m below the surface. Geological conditions have enabled the creation of an aquifer of a confined type, with complicated transboundary characteristics of the aquifer level (freatic-artesian type). In some periods of the year, the clay layer that is 3-4 m thick in some places limits the level of underground water. The aquifer is formed in sands which are below alluvial and diluvial sediments. The thickness of these sands and gravels differs. In some places, their thickness is 5-6 m, at others, less. Amplitudes of water level changes in the aquifer are 2-3 m. Water temperature is 14.5 ºC. The chemical composition of waters in the villages of Ušće, Skela, Grabovac, and Ratari is similar to those previously mentioned (ones on the higher terraces), but with a slight difference, because these waters are more mineralized. In the period these studies were carried out, the level of underground water in the village of Skela was at a depth of 7.4 m to 8 m.

According to local testimonies, the amplitude of level changes is much higher in this than in regions in the south of Serbia and directly dependant on changes in the level of the Sava. According to the studies on aquifer levels at various distances from the Sava riverbank, we can conclude that these aquifers are very permeable and that their levels are approximately the same. This, obviously, points to a quick ability of succeeding equilibrium of water levels in aquifers (normal dynamic curve). A comparison between the chemical composition of waters in the riverbank zone of Sava with those from the river flow itself, shows a tight link between the two. All these waters have low mineralization values of 0.72-0.83 g/l.

ALPHA SPECTROMETRIC MEASUREMENTS

Although the organization of the experimental work initially encompassed 38 water samples collected during the first and second field survey, only 23 were analyzed for ^{226}Ra and uranium isotopes (^{238}U, ^{234}U) activity concentration, using radiochemical methods followed by alpha spectrometry, due to the fact that several samples exhibited a much too thick alpha source for further spectrometric measurements. All water samples (WS) were collected in villages near the TENT B site (fig. 2).

After sampling, acidified water was transferred to the IFJPAN laboratory in Krakow, in polyethylene bottles of 1 dm^3. A sub-sample of 90-250 ml was taken for radium analyses. Using hydrochloric acid, the pH was set below 2. The samples were filtered and evaporated to around 50 ml of the original volume. A spike of ^{133}Ba tracer was added. The exact activity of the spike was not determined. This was not necessary anyway, since the chemical yield was established in a relative measurement comparison with the Ba count rate (for the 356 keV line) in the final Ba(Ra)SO$_4$ co-precipitated alpha source and in the same alpha source as spike ^{133}Ba activity evaporated on the stainless steel plate. Besides a spike at the beginning of the radiochemical analysis, 0.2 mg of the Ba^{+2} carrier (in the form of a barium chloride solution) was added. A simplified method was used in which we do not care too much about the content of calcium ions in water samples. This was checked in the initial batch of samples and then accepted as a rule for the whole set (which would later prove to be a rather bad choice). In this case, a Na$_2$SO$_4$ solution was added in excess and samples were heated to 50-60 °C for about one hour. In such conditions, radium micro-co-precipitated with BaSO$_4$. An additional hour later, the formed tinny crystals were filtrated using a 100 nm membrane filter. The filter was rinsed with ethanol and subsequently glued to a stainless steel disk (2.5 cm in diameter). The chemical yield was determined by means of low background gamma spectrometry in the relative measurement method described above. The sample was then measured for a further few days using the Silena Alpha-Quattro alpha spectrometer. Unfortunately, for some samples, the calcium content was not negligible. In almost a third of all cases, this simplified radiochemical procedure failed – the calcium content

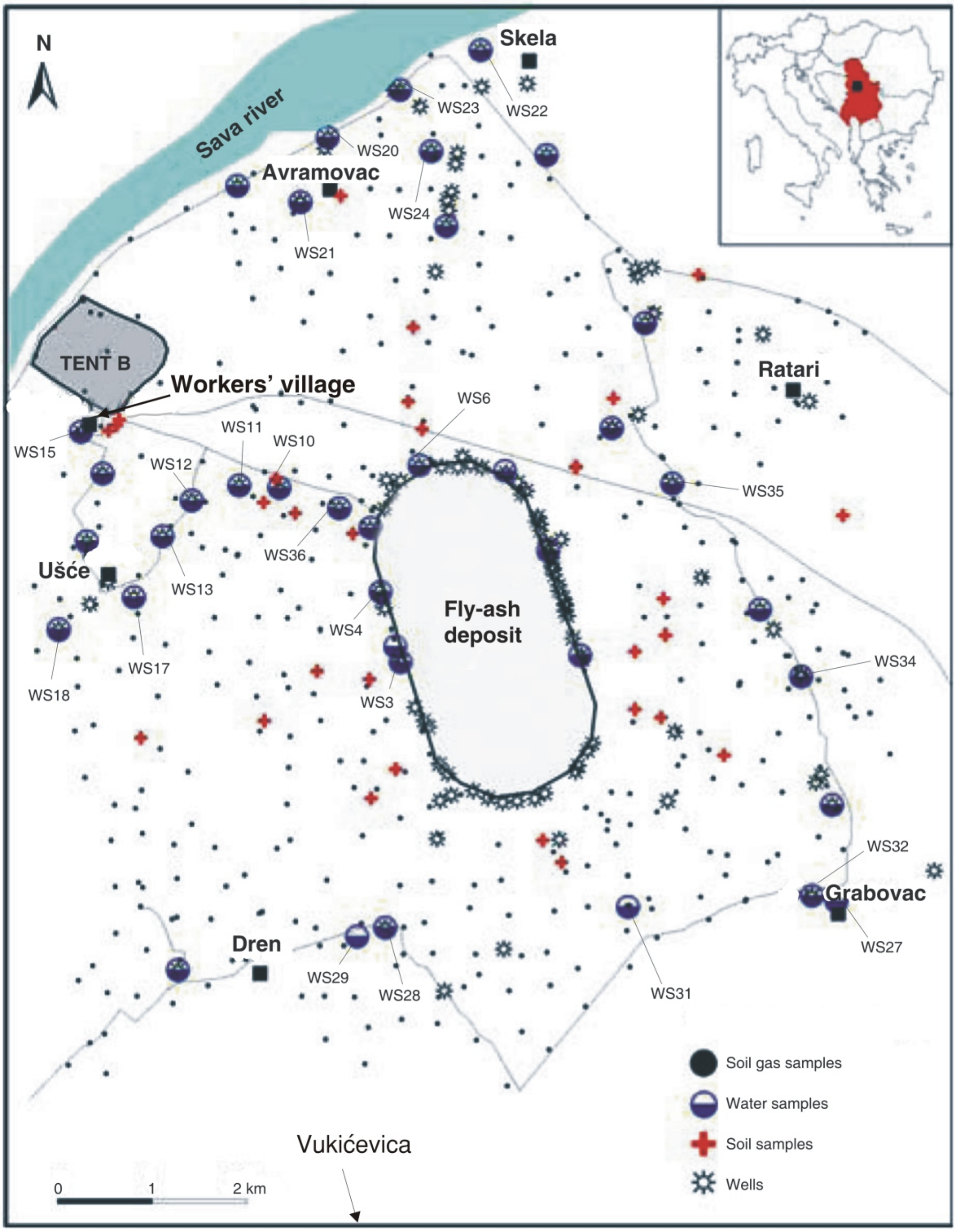

Figure 2.Water sampling locations in the TENT B area

was too high and the obtained source too thick to get a good quality source, meaning that the results obtained were unreliable.

For the purpose of uranium analyses, another sub-sample of 250-400 ml of each water sample was weighed, acidified, filtered and then evaporated to <50 ml. A spike of ^{232}U was added (209 ± 9 mBq). The samples were then evaporated to dryness and dissolved in 50 ml of hot 9 M HCl. After cooling, the solution was filtrated and passed through an anion-exchange column preconditioned with 9 M HCl Dowex-1. U and Fe ions were attached to the resin. Iron was removed by 25 ml of 8 M HNO_3 and then uranium was eluted using 50 ml of 0.5 M HCl with 0.8 g hydroxylamine added. To this solution, 1 g of Mohr salt, 50 μg of Nd^{3+} ions (in the form of a $NdCl_3$ solution) and 5 ml of concentrated HF were added to produce NdF_3 co-precipitated sources [10]. The sample was then measured for a few days using the Silena Alpha-Quattro alpha spectrometer.

The quality of the analyses was monitored by means of the reference material for the uranium in the water [11]. During the time of the samples' treatment, a Polish National Intercomparison Run for ^{226}Ra in water samples was also conducted (with other nuclides and other kinds of samples) [12-14]. Data obtained on applied quality assurance for uranium and radium in water samples examined is presented in tab. 1.

Table 1. Data on applied quality assurance for uraium and radium activity concentration in water samples

Radionuclide	Reference value [Bqkg^{-1}]	Our result [Bqkg^{-1}]
^{226}Ra	1.114 0.033*	1.096 0.035
^{238}U	0.0360 0.0010**	0.0391 0.0025
^{234}U	0.0450 0.0013**	0.0482 0.0027

*Ref. [14]
**Ref. [11]

RESULTS

The results of alpha spectrometric measurements for ^{226}Ra activity concentration (mean value and its standard deviation) are presented in tab. 2, those for ^{234}U and ^{238}U in tab. 3. Activity concentration ratios are presented in tab. 4. The ratios between

Table 2. Results of alpha spectrometric measurements of ^{226}Ra activity concentration in water samples

Sample code	m*[g]	Chemical yield [%]	^{226}Ra [Bqkg^{-1}]
WS3	121	22.0 3.4	0.0097 0.0036
WS4	120	5.3 1.3	0.0180 0.0063
WS6	121	25.8 3.8	0.0063 0.0014
WS10	90	60.2 6.9	0.0038 0.0007
WS11	90	46.7 4.8	0.0271 0.0063
WS12	90	47.6 4.9	0.0053 0.0014
WS13	90	56.1 5.1	0.0040 0.0009
WS15	122	43.8 5.9	0.0052 0.0010
WS17	90	78.9 14.5	0.0037 0.0009
WS18	90	60.5 6.8	0.0028 0.0007
WS20	119	42.1 5.3	0.0026 0.001
WS21	90	69.4 7.9	0.0021 0.001
WS22	120	29.7 2.5	0.0040 0.001
WS23	90	71.6 6.0	0.0023 0.001
WS24	90	69.4 7.2	0.0029 0.001
WS27	121	91.9 11.0	0.0031 0.001
WS28	253	43.3 5.6	0.0029 0.0005
WS29	90	65.8 7.1	0.0049 0.001
WS31	90	46.2 5.3	0.0061 0.001
WS32	122	18.5 5.9	0.0075 0.003
WS34	118	26.1 3.6	0.0037 0.001
WS35	120	64.1 7.5	0.0002 0.0001
WS36	90	54.3 6.2	0.0020 0.0005

*m means mass of sample

Table 3. Results of alpha spectrometric measurements of ^{234}U and ^{238}U activity concentration in water samples

Sample code	m* [g]	Chemical yield [%]	^{234}U [Bqkg^{-1}]	^{238}U [Bqkg^{-1}]
WS3	297	74 4	0.0074	0.0034 0.0006
WS4	337	91 4	0.0108 0.0009	0.0057 0.0007
WS6	347	95 5	0.0232 0.0014	0.0157 0.0011
WS10	393	76 4	0.2303 0.0058	0.1884
WS11	413	67 3	0.0831 0.0039	0.0800 0.0039
WS12	388	78	0.1199 0.0041	0.1013 0.0039
WS13	411	83 4	0.0641 0.0031	0.0455 0.0027
WS15	221	77 4	1.040 0.013	0.854 0.011
WS17	427	85 4	0.0876 0.0032	0.0604 0.0026
WS18	399	87	0.0334 0.0020	0.0202 0.0016
WS20	335	66 3	0.2850 0.0061	0.2434 0.0056
WS21	363	81 4	0.3802 0.0085	0.3305 0.0077
WS22	369	81	0.0714 0.0036	0.0553 0.0031
WS23	388	64 3	0.1180 0.0037	0.0732 0.0029
WS24	382	79 4	0.2040 0.0058	0.1575 0.0050
WS27	378	89 5	0.0959 0.0038	0.0598 0.0029
WS28	239	90 4	0.0708 0.0020	0.0522
WS29	394	88 5	0.0849 0.0034	0.0475 0.0025
WS31	386	95 5	0.0553 0.0028	0.0389 0.0024
WS32	363	86 4	0.0637 0.0033	0.0367 0.0026
WS34	292	73 4	0.0857 0.0029	0.0646 0.0025
WS35	283	66 3	0.1008 0.0034	0.0801 0.0030
WS36	378	85 4	0.0572 0.0027	0.0418 0.0023

*m means mass of sample

Table 4. Ratios of activity concentration of analyzed water samples

Sample code	^{234}U/^{238}U	^{226}Ra/^{238}U
WS3	2.17 0.44	2.9 1.2
WS4	1.91 0.28	3.2 1.2
WS6	1.47 0.14	0.40 0.09
WS10	1.22 0.05	0.020 0.004
WS11	1.04 0.07	0.339 0.081
WS12	1.18 0.06	0.052 0.014
WS13	1.41 0.11	0.089 0.020
WS15	1.22 0.02	0.006 0.001
WS17	1.45 0.08	0.062 0.015
WS18	1.65 0.17	0.140 0.037
WS20	1.17 0.04	0.011 0.003
WS21	1.15 0.04	0.006 0.002
WS22	1.29 0.10	0.072 0.021
WS23	1.61 0.08	0.031 0.010
WS24	1.30 0.06	0.019 0.005
WS27	1.60 0.10	0.052 0.009
WS28	1.36 0.06	0.055 0.010
WS29	1.79 0.12	0.102 0.018
WS31	1.42 0.11	0.156 0.033
WS32	1.74 0.15	0.205 0.079
WS34	1.33 0.07	0.057 0.016
WS35	1.26 0.06	0.002 0.001
WS36	1.37 0.10	0.048 0.012

Table 5. Comparison of statistical data for uranium and radium activity concentration in examined wells and springs at TENT B area

Radionuclide	Wells at the edge of fly-ash deposit of TENT B	Wells or springs in villages near TENT B
^{226}Ra	0.0113 0.0060	0.0048 0.0055
^{238}U	0.0083 0.0054	0.115 0.170
^{234}U	0.0145 0.0069	0.147 0.230

Table 6. Comparison of statistical data of radium activity concentration in examined wells and springs in the villages near TENT B and other areas across Serbia

Field site	^{226}Ra [Bq l^{-1}]
TENT B	0.0048 0.0055
Niška Banja	0.62 0.10
Vranjska Banja	0.08 0.02
Slatina	0.11 0.03

^{234}U and ^{238}U are typical of water samples slightly enhanced by lighter isotopes. The highest values of this ratio did not exceed 3, which is a quite moderate value. The mean value and its standard deviation for radium and uranium activity concentration (tab. 5) show a difference between samples from the fly-ash deposit area (where more ^{226}Ra than ^{238}U was detected) compared to the remaining samples from the villages. This seems to be the only difference between the water collected in the TENT B heap and the rest of the samples. However, the radium activity concentration in examined water samples from the villages near the thermal power plant (tab. 6), shows low level in comparison with the high radon area (Niška Banja) and other regions (Vranjska Banja, Slatina) across Serbia [15] . No correlation was found between the activity concentration of ^{238}U and ^{226}Ra in the examined water samples (fig. 3).

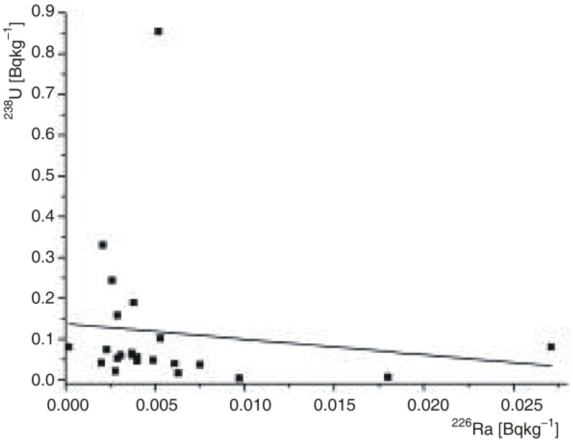

Figure 3. Correlation between uranium and radium in water samples

CONCLUSION

Uranium and radium activity concentration of the water around TENT B is low and occurs in a small area around the fly-ash deposit. The groundwater downstream of the waste facilities is not, or is merely weakly affected by uranium and radium isotopes present in the TENT B heap. Thus, our conclusion is that uranium and radium leakage from the TENT B heap does not present an acute problem. Results obtained show that there is no correlation between the activity concentration of ^{238}U and ^{226}Ra in the water samples. Furthermore, uranium and radium contents in water around the lignite thermal power plant are low when compared to those from areas with enhanced natural uranium and radium content and other surveyed regions in Serbia. Therefore, no significant radiological hazard related to heap-stored uranium and radium may be concluded.

ACKNOWLEDGEMENT

This work was supported by The Ministry of Education and Science of the Republic of Serbia under contract 41028.

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Received on December 20, 2010
Accepted on March 8, 2011

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УРАНИЈУМ И РАДИЈУМ У УЗОРЦИМА ВОДЕ У ОКОЛИНИ ТЕРМОЕЛЕКТРАНЕ "НИКОЛА ТЕСЛА" Б – ОБРЕНОВАЦ

Рад се бави анализом садржаја природних радионуклида у 23 узорака воде сакупљених у околини термоелектране "Никола Тесла" Б – Обреновац, Србија. У свим узорцима је анализирана радиоактивност ^{226}Ra и уранијумових изотопа (^{238}U, ^{234}U) користећи радиохемијске методе и алфа спектрометрију. Добијени резултати показују да је радиоактивност уранијума и радијума у води у околини термоелектране ниска у поређењу са мерењима у областима Србије са повећаним садржајем природног уранијум и радијума. Активност уранијума и радијума наталожених на пепелишту не представља значајну радиолошку опасност за животну средину у околини.

Кључне речи: алфа спектрометрија, термоелектрана, радиоактивност, радијум, уранијум