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Fly Ash Radioactivity Measurements in Electric Power Industry of Serbia Thermal Power Plants

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Abstract

Serbian thermal power plants (TPPs) produce siliceous fly ash from lignite in the quantity of approximately 6 million tons per year. The potential market for the use of fly ash is operational, but for the time being, only used by cement producers. Fly ash radioactivity could be one of the major points of concern when larger use of fly ash is planned, particularly in the Serbian construction industry.

Radioactivity measurements have been conducted regularly for decades. This paper presents the results of a ten-year fly ash radioactivity measurements at the Nikola Tesla B TPP located in Obrenovac.

In addition, the paper compares the natural radionuclides coal content data coming from the Kolubara Basin and ash created as the coal combustion by-product in the Nikola Tesla B TPP boilers. Following the obtained results indicating the ^{26}Ra, ^{232}Th, ^{40}K content and joint concentration of all artificial radionuclides, it may be concluded that TPP ash and slag are environmentally friendly. Moreover, they may be used in the construction industry since they meet the legal criteria defining the radionuclides content.

Following the obtained natural radionuclides content results it may be concluded that the Nikola Tesla B TPP ash may be disposed into the environment. Ash may be used also in the construction industry (civil engineering). In building construction applications, ash share as the additive to other building materials depends from its physical and chemical characteristics, as well as from the radionuclides activity: ^{266}Ra, ^{232}Th and ^{40}K.

Keywords: Serbian power plants, fly ash, radioactivity measurements, natural radionuclides, ^{238}U, ^{232}Th, construction industry.

1 Introduction

Fly ash is an inorganic part of the soft to hard brown coal (lignite) combustion products in thermal power plants. Ash is collected in hoppers below electrostatic precipitator sections, subsequently transported and mixed with water in ejectors and discharged into the slurry reservoirs. Bottom ash falling to the boiler bottom (grate) is also mixed with water and transported to the slurry reservoirs and afterwards hydraulically transported (1:10 ash – water ratio – old technology) and disposed on open ash disposal sites. The new technology enables fly ash collection in silos and dry fly ash delivery for industrial purposes. Systems of this kind have been recently introduced at the Nikola Tesla B TPP, Kolubara A TPP and Kostolac B TPP – thermal power plants with the largest electricity generation in Serbia.

At the Nikola Tesla A TPP, fly and bottom ash are transported and disposed hydraulically to the disposal site (old technology, solids – water ratio – 1:10), while at the Nikola Tesla B TPP from 2009/2010 fly and bottom ash are transported and disposed by applying the new technology, i.e. in the form of thick slurry, where the solids and water ratio amounts to 1:1 [1].

Electric Power Industry of Serbia thermal power plants annually combust some 32,000,000 t of lignite from the Kolubara and Kostolac Basins. Depending on the combusted coal amounts, annual ash amount separated in electrostatic precipitators at the Nikola Tesla A TPP (TENT A) and the Nikola Tesla B TPP (TENT B) is some 3,700,000 t, while it equals to some 375,000 t at the Kolubara A TPP and 100,000 t at the Morava TPP [2]. Combustion of coal coming from the Kostolac Basin, at the Kostolac A and B TPPs produces some 1,700,000t of ash annually. After it is separated by electrostatic precipitators, fly ash is collected, mixed with water and transported by pipelines to open fly and bottom ash disposal sites.

Ash disposal sites occupy large land areas: TENT A – 400ha, TENT B 400 ha, Kolubara A TPP – 77ha, Morava TPP – 35ha and Kostolac A and B TPPs – 246ha. They are surrounded by settlements and arable land and represent diffusion air, water and soil pollution sources [3]. Mitigation of adverse environmental impacts of ash disposal sites call for constant application of adequate protection measures which have to be regularly improved. This requires large-scale investments. In addition to the above costs, far greater costs are incurred throughout the collection, transport and disposal of ash. The above costs altogether considerably increase the electricity price [2].

Mass ash application in the construction industry (cement, concrete and brick production), civil engineering (road construction) and for other purposes (soil pH value adjustment) would considerably mitigate problems, since the disposed ash amounts would be reduced. Moreover, ash usage would reduce the usage of natural materials possessing similar chemical properties, such as sand, clay, marl and limestone [2].

2 Natural Radionuclides Content in Coal

Coal combusted by thermal power plants is a main energy source in Serbia. Coal as such may contain significant amounts of natural radionuclides – Uranium 238 (^{238}U), Radium 226 (^{226}Ra), Thorium 232 (^{232}Th) and Potassium 40 (^{40}K), i.e. Normally Occurring Radioactive Material (NORM). Statistical analysis of data obtained by testing Uranium (^{238}U) and Thorium (^{232}Th) content on 95 samples coming from the Serbian and Montenegrin lignite basins is shown in Table 1 [4].

Compared to the coals from the above Serbian basins, Kolubara Basin coal has a low uranium 238 (^{238}U) content, while its thorium 232 (^{232}Th) content is higher, which is also goes for the Krepoljin Basin coal.

Krepoljin Basin coal is characterised by higher uranium content and the highest thorium content compared to all the investigated coals. Uranium and thorium content in the Sjenica and Soko Banja Basins is low. Senje – Resavica Basin coal is characterised by low uranium and thorium content. Pljevlja Basin coal (Montenegro) has higher uranium and thorium concentrations [4].

Table 1 Natural radionuclides content: ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K in Serbian coals in comparison to other coals [4]

Coal basin	^{238}U (mg/kg)			^{232}Th (mg/kg)		
	min	max	MD	min	max	MD
Kostolac	0.60	70.10	0.95	0.20	2.60	1.08
Kolubara	0.65	3.20	1.84	0.84	6.57	3.18
Krepoljin	0.95	6.59	2.99	1.48	6.48	3.65
Sjenica	1.20	6.05	3.11	0.12	2.71	1.18
Soko Banja	0.80	6.66	3.17	0.13	4.95	0.80
Bogovina – East Field	0.18	89.90	13.55	0.14	3.48	0.33
Senje - Resavica	0.19	4.14	1.35	0.29	3.56	0.90
Pljevlja	0.28	3.52	1.30	0.17	1.89	0.78
MD- median values						

3 Environmental Control of the Nikola Tesla B TPP Natural Radioactivity

Throughout the coal combustion process, natural radionuclides are mainly concentrated in the solid coal combustion products (by-products), i.e. fly and bottom ash, representing the thermal power plant industrial by-products. As a result, fly and bottom ash may contain increased natural radionuclides amounts, i.e. they are the so-called Technologically Enhanced Occurring Radioactive Material (TENORM). This material type may be disposed to the open fly and bottom ash disposal sites or reused as a building material additive.

Targeted investigations of the natural radionuclides content, uranium (^{238}U, ^{226}Ra), thorium (^{232}Th) and potassium (^{40}K) series, was executed in 1996 to identify potential radioactive contamination of soil and groundwater originating from the TPP industrial by-products – Nikola Tesla B TPP ash and slag disposal site. The above investigations indicated noteworthy but not very large content of these radionuclides [5]. In 2004, natural radionuclides activity was measured during the short-term active investigations (based on the Alpha Guard method), indicating low radon and thoron concentrations. This proved that the Nikola Tesla B zone is an area with the low basic natural radiation levels. Preliminary investigations, conducted between 2005 and 2007, confirmed low radionuclides level around the Nikola Tesla B TPP [6]. Measured radon and thoron radionuclides specific activity around the Nikola Tesla B TPP disposal site, on 29 measuring points in the ground at the depth of 80cm, identified radioactive gases specific activity below 10 000 $Bq \cdot m^{-3}$, with the log-normal radon specific activity distribution. Obtained results indicate that the disposal site area contains low basic natural radiation levels. Results comply with the previous investigations carried out between 1996 and 2007, when low natural specific activity was also identified in soil and groundwater (uranium, radium, thorium and potassium) [6].

Furthermore, the authorised institutions have regularly controlled living and working environment radioactivity levels from 1990 for the Nikola Tesla B TPP needs. Control activities are aimed at evaluating the natural radioactivity above the natural levels in the Nikola Tesla B TPP surroundings, assessing the technologically altered natural radioactivity health effects and identifying the potential local artificial radioactivity origin [7].

Table 2 Gama spectrometric analyses between 1990 and 2011, Nikola Tesla B TPP [8] [9].

Measuring points / Parameters		^{232}Th [Bq/kg]			^{226}Ra [Bq/kg]			^{238}U [Bq/kg]			^{40}K [Bq/kg]		
		min	max	AM	min	max	AM	min	max	AM	min	max	AM
Coal from the feeder		10.6	66.8	25.2	14.1	86.8	36.3	11.0	68.9	38.3	36.2	376.9	112.2
Bottom ash prior to boiler grate		19.8	91.6	59.0	28.9	276.2	88.6	18.9	160.3	81.2	118.0	472.4	294.4
Fly ash		71.0	104.0	84.2	91.0	152	127.1	114.0	174.0	142.60	357	473.0	415.6
Disposal site ash	Active cassette	11.3	143.0	75.3	46.0	318.8	120.9	19.1	284.5	110.7	81.3	693.4	343.1
	Passive cassette	12.4	109.0	73.9	27.0	279.9	117.4	15.5	241.0	119.5	241.0	669.0	398.2
Plants	Passive cassette	0.5	89.0	10.3	0.3	138.0	14.0	1.3	24.0	8.4	38.0	641.0	192.7
	Active cassette	0.5	7.9	3.3	0.7	29.0	6.0	1.8	14.8	6.6	58.0	597.0	231.8
	Soil around the disposal site	0.2	10.8	2.7	0.5	35.3	6.8	2.0	26.0	7.6	46.8	879.0	344.3
	Soil from the settlement in the disposal site vicinity (some 10km away)	0.1	10.1	2.0	0.3	13.5	2.8	1.3	25.0	5.0	55.7	945.0	318.4
Soil	Disposal site surroundings	3.9	72.0	52.3	4.5.0	154.8	51.4	6.5	102.0	48.8	390.0	877.0	648.6
	Soil from the settlement in the disposal site vicinity (some 10km away)	0.2	74.0	49.5	26.0	83.0	50.5	13.9	106.0	53.5	555.0	659.0	609.0
Water	Sava River upstream	0.017	0.20	0.06	0.01	1.10	0.31	0.100	54.00	7.83	0.025	0.91	0.31
	Disposal site overflow	0.010	0.90	0.14	0.010	0.80	0.34	0.100	37.00	6.56	0.120	0.63	0.29
	Disposal site drainage	0.010	0.10	0.04	0.010	0.80	0.27	0.100	37.00	6.82	0.140	0.69	0.32
	Sava River downstream	0.010	0.16	0.05	0.010	0.85	0.27	0.090	44.00	7.22	0.100	0.87	0.24

Data were analysed on the basis of the measurements carried out by the following authorised institutions:

Dr Dragomir Karajovic Industrial Medicine and Radiologic Protection Institute – Belgrade (1990-2002, 2005, 2006 and 2011),

Vinca Nuclear Sciences Institute - Belgrade (2003, 2004, 2007-2010),
AM -arithmetic mean values

Gamma-spectrometric analyses of the following samples were conducted:

- Coal;
- Bottom ash;
- Fly ash from the generation process;
- Fly and bottom ash disposed at the disposal sites;
- Surrounding disposal site soil;
- Agricultural land in 10km radius around the power plant (arable land);
- Disposal site plants and plants from adequate land;
- Rivers (Sava River before and after wastewater discharge)
- Disposal site wastewater.

Radionuclides specific activity measurements included the uranium series (^{238}U, ^{226}Ra), thorium series (^{232}Th) and ^{40}K, activation radionuclides (^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{65}Zn), fallout radionuclides (^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce, ^{134}Cs and ^{137}Cs) and cosmogenic radionuclide (^{7}Be). Together with the sampling, ambient gamma radiation air levels were measured, one metre above the ground surface. Measurements also included the total alpha and beta activity of wastewaters and the Sava River waters [8] and [9].

Table 2 provides the gamma emitters spectrometry measurement results for the Nikola Tesla B between 1990 and 2011. It shows minimum, maximum and arithmetic mean concentrations of the natural radionuclides – uranium series (^{238}U, ^{226}Ra), thorium series (^{232}Th) and potassium 40 (^{40}K) [8] and [9]. Natural radionuclides activity measurements in coal, bottom ash, fly ash, disposal site ash and soil (Table 2) indicate the following:

- Fly ash contain the following natural radionuclides activity:

  ^{232}Th in coal and fly ash amounts to 25.2 [Bq/kg] and 84.2 [Bq/kg] respectively

  ^{238}U in coal and fly ash amounts to 38.3 [Bq/kg] and 142.60 [Bq/kg] respectively

  ^{40}K in coal and fly ash amounts to 112.2 [Bq/kg] and 415.60 [Bq/kg] respectively

- Disposal sites radionuclides content is higher than their content in soil (except ^{40}K), with the following average radionuclides activity:

  ^{232}Th in the settlement soil and passive ash cassette amounts to 49.5[Bq/kg] and 73.9 [Bq/kg] respectively

  ^{238}U in the settlement soil and passive ash cassette amounts to 53.5 [Bq/kg] and 119.5 [Bq/kg] respectively

  ^{40}K in the settlement soil and passive ash cassette amounts to 609.0 [Bq/kg] and 398.2[Bq/kg respectively

- There is no large difference in the radionuclides content on active (disposal site area where ash is temporarily disposed) and passive cassettes within the observed period. This is justified by the fact that active and passive cassettes role alternates over time, i.e. active become passive and vice versa.

Long-term radionuclides measurements results in the living and working environment of the Nikola Tesla B TPP have also indicated the following:

Coal, bottom ash, fly ash, ash disposal site and soil samples contain natural radionuclides, uranium 238 and 235 (^{238}U, ^{235}U), radium 226 (^{226}Ra), thorium 232 (^{232}Th), lead 210 (^{210}Pb) and produced radionuclide, caesium 137 (^{137}Cs). The effect of the Chernobyl nuclear hazard was detected only in soil samples [9].

Analysed waters (overflow and drainage wastewaters from the ash disposal site and the Sava River upstream and downstream from the Nikola Tesla B TPP) exhibit low total alpha and gamma activity values. Values are lower than the maximum permitted ones for potable water (Limit value of the total alpha unstable radionuclides activity – 0.5 Bq/l; Limit value of the total beta unstable radionuclides activity - 1 Bq/l) [10].

Absorbed gamma radiation dosage intensity measurement results fall within the basic radiation level ranges, i.e. the obtained values do not indicate any basic radiation level increase caused by the Nikola Tesla B TPP [8] and [9].

When gamma emitters spectrometry results for fly ash are compared with the literature data obtained in other countries (USA, ex-USSR, Poland, India, Turkey), it may be noticed that the values are the same, Table 3 [11 - 16].

Table 3 Specific radionuclid activity for the fly ash in some world countries [Bq/kg]

Country	Radionuclid Specific Activity							
	^{232}Th[Bq/kg]		^{226}Ra[Bq/kg]		^{238}U[Bq/kg]		^{40}K[Bq/kg]	
	min	max	min	max	min	max	min	max
USA	78	122	41.8	140.6	111	204	259	370
Ex-USSR	185		185		-		1850	
Poland	47.5	91.5	54.2	119.3	-	-	448.5	758
India	96.2	177.7	40.7	151.7	-	-	148.0	840.1
Turkey	57.97		149.43		-		94.15	
China	118.7	195.6	76.1	165.7	-	-	261.5	520.8
Germany	88		118.4		133.2		514.3	

4 Ash Use in the Construction Industry

According to the data for Serbia, ash use in the construction industry, chemical industry, metallurgy and agriculture is quite small. To expand industrial use of ash, new technologies need to be applied in individual industrial sectors. Ash use in road construction is of special importance since it has large ash use potentials [2]. Given that the natural radionuclides content in ash may be a limiting factor for road construction applications, natural radionuclides content needs to be identified. In addition to the fly and bottom ash radioactivity investigations aimed at identifying its disposal suitability, their construction industry applications are also investigated.

Long-term investigations confirm industrial applications of ash [2]. Due to the increased natural radioactivity levels in fly and bottom ash samples, industrial by-products need to be constantly controlled enabling their construction industry application. According the Serbian regulations, different criteria were established for the radionuclides content in the building material. Such criteria are provided in Table 4 (*Articles 13 – 15), relating to the building material used in the building construction, both exterior and interior applications, and civil engineering construction as road, playground, base, etc. [10].

Table 4 Radionuclides concentration limits in the construction material [10]

*Article	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
Article 13 (interior applications)	300	200	3 000
Article14 (exterior applications)	400	300	5 000
Article 15 (road and sports grounds bases)	700	500	8 000
Formula label	Max (Ra)	Max (Th)	Max (K)

*Radionuclides Content Limits for Potable Water, Food, Fodder, Medicines, General Use Objects, Construction Material and other Goods subject to Sale Rules, Official Gazette of the Republic of Serbia, № 86/2011.

For a certain material to be used in the construction industry, its ^{226}Ra, ^{232}Th, ^{40}K specific activity need to meet the following relation:

$$\frac{C_{Ra}}{Max(Ra)} + \frac{C_{Th}}{Max(Th)} + \frac{C_{K}}{Max(K)} < 1$$

where C_x denotes adequate radionuclides content, while *Max(x)* denotes radionuclides content limits (Table 4) for adequate material application category, both in the building construction (exterior and interior applications) and civil engineering construction (roads, sports grounds, etc.).

Following the obtained natural radionuclides content results it may be concluded that the Nikola Tesla B TPP ash may be disposed into the environment. Ash may be used in the construction industry (civil engineering), when its gamma index is lower than 1 [10]. In building construction applications, ash share as the additive to other building materials depends from its physical and chemical characteristics, as well as from the radionuclides activity: ^{266}Ra, ^{232}Th and ^{40}K. Therefore, natural radionuclides content in all the input components and the final product should be regularly controlled.

Fly ash use originating from the Nikola Tesla TPPs Company is economically and environmentally justified and of interest for the following reasons [2]:

- environmental fee exemption or reduction, since the annual Nikola Tesla A and B ash production is in question, come 3,700,000 t and large funds paid for this fee (ca. EUR 6,500,000 annualy)
- ash sale profits
- transport and disposal costs reduction (reduced ash amount disposed at the disposal site)
- disposal site operating life extension
- natural resources use reduction (sand, clay, limestone, marl having similar properties)
- mitigation of environmental problems.

The new fly and bottom ash collection, transport and disposal technology introduced at the Nikola Tesla B TPP and other thermal power plants provided separate dry fly and bottom ash collection in silos and dry ash delivery to the construction industry. During 2011, the Nikola Tesla B delivered (on the basis of contracts) some 40,336 t of dry ash to the Lafarge Cement Plan – Beocin and 3,045 t of dry ash to the Titan Cement Plant – Kosjeric. Moreover, some 5,267 t of bottom ash were also delivered to the Obrenovac Construction Company and used as the uncategorised road base within the Obrenovac Municipality.

5 Conclusion

1. Kolubara Basin coal combusted by the Nikola Tesla B TPP boilers contain natural radioactive materials (Normally Occurring Radioactive Material – NORM). Compared to coals from other Serbian basins, its uranium (^{238}U) content is low, while its thorium (^{232}Th) content is increased.
2. Fly and bottom ash have an increased natural radionuclides content compared to coal, the so-called Technologically Enhanced Occurring Radioactive Material – TENORM of industrial by-product, with the following average radionuclides activity:

   ^{232}Th in coal and fly ash amounts to 25.2 [Bq/kg] and 84.2 [Bq/kg] respectively

   ^{232}U in coal and fly ash amounts to 38.3 [Bq/kg] and 142.60 [Bq/kg] respectively

   ^{40}K in coal and fly ash amounts to 112.2 [Bq/kg] and 415.60 [Bq/kg] respectively

3. Ash may be used in civil engineering, since its gamma index is lower than 1. In building construction applications, ash share as the additive to other building materials depends from its physical and chemical characteristics, as well as from the radionuclides activity: ^{266}Ra, ^{232}Th and ^{40}K.
4. New fly and bottom ash collection transport and disposal technology at the Nikola Tesla B TPP and other thermal power plants covers dry ash needs of the construction industry.

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