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NATURALLY OCCURRING RADIONUCLIDES AND BASIC CHARACTERISTICS OF SOIL AND ASH SAMPLES NEARBY COAL–FIRED POWER PLANTS

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Abstract

Deposition of (fly and bottom) ash generated after coal combustion in the coal fired power plants (CFPP) in Serbia is carried out in active and passive lagoons. Ash waste mixed with water is directly transported to the lagoon currently active and the other one is passive in the stage of temporary inactivity for technical consolidation of ash and drainage and subjected to revegetation process using grass–legume mixtures with the purpose of creating plant cover. In order to obtain similarity, samples studied in this work were all taken from the area covered with grass which included: (1) soil close to CFPP (<2 km), (2) soil further from CFPP (>2 km) and (3) ash from the flat area of associated passive lagoon. Investigated sites were four power plants: TE "Kolubara" (TEK), TE "Morava" (TEM), TE "Nikola Tesla" A (Tent A) and B (Tent B). In order to analyse environmental implications of ash deposition in the surrounding area, basic characteristics such as texture, particle size distribution, pH value, organic matter and carbonate content were determined in the soil and ash samples. Simultaneously, ^{238}U, ^{226}Ra, ^{210}Pb and ^{232}Th activity concentrations were measured as it is known that after elimination of the organic component of the coal in the process of combustion naturally occurring radionuclides activity concentrations in the coal ash could be enhanced up to 10 times. Analyses of differences between soil and ash samples collected in this study showed that for one group of soils some changes of physical and chemical characteristics occurred compared to the rest of the soils. These changes were found to be related to the soil texture, percentages of clay size particles and ^{232}Th/^{226}Ra activity concentration ratios.

Keywords: soil; coal ash; CFPP; naturally occurring radionuclides

INTRODUCTION

Coal as naturally found material contains traces of naturally occurring primordial radionuclides that include radioactive decay series precursors (^{238}U, ^{235}U and ^{232}Th) and their decay products (such as ^{226}Ra, ^{210}Pb, ^{228}Ra) as well as potassium isotope ^{40}K, in different quantities depending on geological origin of the coal. After burning of coal in the

coal fired power plants, naturally occurring radionuclides are released from the original coal matrix and distributed between the gaseous phase and solid combustion by-products. Gaseous phase contains volatile nuclides (such as Rn, Pb, Po) while less or non-volatile nuclides (U, Ra, Th, K) are trapped and concentrated in the bottom-ash and slag or in the smallest and lightest particles of fly-ash which are expelled together with the hot gases (Papastefanou, 2008; Karangelos et al., 2004). As a result, after elimination of organic component of the original coal, natural radionuclides activity concentration could be up to 10 times higher in solid combustion wastes (Papastefanou, 2008). For example, if lignite is used as feed coal it could be expected that secular radioactive equilibrium would be maintained in the ^{238}U radioactive series in coal and disturbed in the ashes due to variations in radionuclide enrichment processes in different fractions of ash. Then, fly-ash would be more enriched in ^{238}U and ^{226}Ra than bottom-ash while the smallest fly-ash particles would be highly enriched in ^{210}Pb and depleted in it within bottom-ash (Karangelos et al., 2004). Subsequently, as a result of CFPPs activities, radionuclides are being dispersed into the environment through the leaching of solid combustion by-products during disposal of ash waste to the disposal sites and after deposition on the nearby soil or through atmospheric emissions in the gaseous or particulate form. Over 80% of the disposed ash waste consists of fly-ash particles ranging in size between 0.5 μm to 300 μm. Nevertheless, coal fly-ash detection in the soil environment is found to be challenging given its small particle size and in the study of Wang et al. (2021) low percentage of fly-ash in the soil (<10%) did not yielded to appreciable differences relative to the reference soil and only the increased fractions of fly-ash leaded to a more distinguishable soil-ash mixtures. The study also explored if trace elements and radioactive isotopes of radium could be used as a detection tool for fly-ash presence in the surface soils and discussed distinctions in radium abundance and ^{228}Ra/^{226}Ra ratios (reflecting the Th/U activity ratios) between coal, fly-ash and common soil.

Deposition of fly-ash and bottom-ash generated after coal combustion in the CFPPs in Serbia is carried out in active and passive lagoons. Ash waste mixed with water is directly transported to the lagoon currently active and the other one is passive in the stage of temporary inactivity for technical consolidation of ash and drainage. At the site Tent A, results of revegetation process were investigated 3 and 11 after the sowing of a grass–legume mixture directly onto the ash (without the application of topsoil), using agronomic measures (Kostić et al., 2018) and concluded that passive lagoons should be subjected to the revegetation creating plant cover in order to improve physical and chemical characteristics and ensure stabilisation of ash waste deposits.

In order to analyse environmental implications of ash redistribution in the surrounding area of four CFPPs from Serbia, basic characteristics such as texture, particle size distribution, pH value, organic matter and carbonate content were determined in the soil and ash samples and simultaneously activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb and ^{232}Th were measured. The aim of the present study was to analyse whether there were: i) any differences between ash and soil according to the measured physical and chemical parameters and radionuclides activity concentrations and ii) any relations between investigated radionuclides activity concentrations and basic characteristics regardless of sample type.

MATERIALS AND METHODS

Study area

Area in the vicinity of four coal fired power plants from Serbia was under study in this work: "Nikola Tesla A" and "Nikola Tesla B" situated on the Sava River bank, TE "Kolubara" on the Kolubara River and TE "Morava" on the right bank of the Velika Morava River. In the area near Tent A different soil types such as calcaric fluvisols, vertisols, gleyosols, phaeozems, stanic gleyosols, cutanic cambisols, humic gleyosols are found to be developed, but as a consequence of CFPP activities, technosols and spolic regosols could be present (Tanić et al., 2016). In summary, fluvisols may be recognized as the most common soil type distributed along the river valleys where CFPPs are situated.

Soil and ash sampling

The sampling points were chosen to be located within the radius of 6 km around each CFPP. Studied samples were all taken from the area covered with grass. It was assumed that ash and soil samples would be more appropriate to compare according to their properties if conditions for vegetation development are met since revegetation of ash deposits improves physical and chemical characteristics of ash waste. Sampling locations included: (1) soil close to the CFPP (<2 km), (2) soil further from the CFPP (>2 km) and (3) ash from the flat area of associated passive lagoon. In the period from 2012 to 2014, a total of 20 soil and 8 ash samples were collected from the surface horizon of 0-10 cm depth. At each location grass was cut off and removed from the surface layer prior to sampling and about 1–1.5 kg of soil or ash was collected.

Soil and ash analysis

In the laboratory, debris and grass residues were removed from the collected material. Samples left to be air-dried at room temperature and afterwards to be oven dried at 105°C to a constant mass. Prepared like that, soil and ash samples were sieved through 2 mm mesh sieve. The pH-reaction, content of carbonates and organic matter content were analysed using standard procedures. Particle size distribution analysis was conducted by combined pipette and sieve techniques. Determined fractions were sand (particle sizes of 2000–200 μm and 200–50 μm), silt (50–10 μm and 10–2 μm) and clay (<2 μm).

For the determination of radionuclides activity concentration, all prepared samples of soil and ash were packed in 500 ml Marinelli beakers, sealed, covered with a film of beeswax and left for 4 weeks in order to ^{226}Ra and ^{232}Th attain secular equilibrium with their decay products. Applying the gamma spectrometry method, measurements were performed with the HPGe detectors (Canberra Industries, Inc., Meriden, CT, USA) with 18%, 20% and 50% relative efficiency and energy resolution of all the detectors of 1.8 keV at the 1332 keV gamma ray energy of ^{60}Co. The detectors were calibrated using secondary reference material with soil matrix which was produced using certified radioactive mixture solution (9031–OL–427/12 type ERX by Czech Metrology Institute, Inspectorate for Ionizing Radiation, Praha) that contained radionuclides: ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{88}Y and ^{210}Pb with total activity 7.4 kBq at reference date 31.08.2012.

Calibration was performed in the same geometry of V=500 ml Marinelli beaker as the geometry of measured samples.

The activity of ^{238}U was determined through its daughter products in equilibrium in soil ^{234}Th (63 keV) or ^{234m}Pa (1001 keV). The activities of ^{226}Ra were determined by its decay products: ^{214}Bi (609.3; 1120.3 and 1764.5 keV) and ^{214}Pb (295.2 and 351.9 keV) and ^{232}Th activities by its decay product ^{228}Ac (338 and 911 keV). Using 46.5 keV γ-energy photons, ^{210}Pb activity concentrations were determined. The spectra were recorded and analysed using Canberra's Genie 2000 software. Counting time was about 60000 s. Activity concentrations are expressed along with their combine measurement uncertainty at the 95% confidence level.

Statistical analysis

Normality of the data was assessed by the Shapiro-Wilk's test. For data groups with a tendency toward normal distribution summarized results are presented by the arithmetic mean and for one with a tendency towards log-normal distribution by the geometric mean. Statistical analysis was conducted by one-way analysis of variance (ANOVA) to specify the main differences in measured physical and chemical properties and radionuclides activity concentrations between investigated soil and ash samples. To find relationships between radionuclides activity concentration ratios and the measured basic properties, simple linear regression analysis was performed. Significant differences and regression analysis results were considered at the 95% confidence level and the results which were not significant are not presented.

RESULTS AND DISCUSSION

Measurement results from 2012., 2013. and 2014. of basic properties of soil close to each CFPP, soil further from the CFPP and ash from the flat area of associated passive lagoon including their ^{238}U, ^{226}Ra, ^{210}Pb and ^{232}Th activity concentrations are presented in Appendix 1 as supplementary material. Texture of investigated samples was also determined and denoted as: LS–Loamy Sand, SL–Sandy Loam, L–Loam, SiL–Silt Loam, SiCL–Silty Clay Loam and CL–Clay Loam.

Differences between samples of ash and soil

The tests of normality showed that data groups consisting of measured pH values, humus content, fractions of silt (coarse, fine and total), clay and physical clay followed normal distribution. The values of $CaCO_3$ content and sand fractions (coarse, fine and total) followed normal distribution after logarithmic transformation of the data. One-way analysis of variance was applied to identify differences between the properties of ash and soil and the results are shown in Table 1. The mean value of $CaCO_3$ content was not significantly different between ash (1.5%) and soil (1.9%), nor did the humus content, which was about 3% on both substrates. The accumulated humus in the ash samples from the passive lagoon could come from vegetation residues, but also from unburned parts of the coal. Ash was weakly alkaline with pH (in H_2O) in the range of 7.3 to 8.4 and the mean

pH of ash (7.9) differed significantly ($p<0.01$) from that in soil (7.3) which was in the range 6.1 to 7.9.

Table 1. The one-way analysis of variance of investigated basic properties and natural radionuclides activity concentrations of ash and soil samples. The geometric mean (bold letters) is presented for log–normal distribution and the arithmetic mean for normal distribution of data groups.

	Soil	Ash	*F* –ratio	*p* –value
2000–50 μm (%)	**12.0** (1.6–42.3)	**56.0** (13.3–79.6)	28.22	<0.0001
2000–200 μm (%)	**6.7** (0.1–24.8)	**20.3** (2.7–43.6)	10.73	<0.01
200–50 μm (%)	**10.4** (0.1–33.5)	**35.7** (10.6–57.1)	29.63	<0.0001
50–2 μm (%)	54.7 (30.6–77.4)	36.8 (16.9–75.8)	7.7	<0.05
50–10 μm (%)	28.2 (14.1–62.6)	19.0 (8.8–39.7)		
10–2 μm (%)	26.4 (5.0–41.9)	17.8 (6.6–36.1)		
<2μm (%)	28.3 (12.3–39.8)	3.6 (2.3–5.4)	87.39	<0.0001
<10μm (%)	54.7 (27.1–74.6)	25.0 (10.4–63.7)	21.45	<0.001
OM (%)	3.3 (1.2–5.3)	3.5 (0.6–7.3)		
$CaCO_3$ (%)	**1.9** (0–15.0)	**1.5** (0–4.4)		
pH (H_2O)	7.3 (6.1–7.9)	7.9 (7.3–8.4)	9.66	<0.01
pH (KCl)	6.5 (5.4–7.2)	7.5 (6.2–8.2)	15.95	<0.001
^{238}U ($Bqkg^{-1}$)	**38.2** (24–56)	**106.3** (46–190)	66.10	<0.0001
^{226}Ra ($Bqkg^{-1}$)	**38.7** (25–56)	**102.9** (42–167)	66.62	<0.0001
^{210}Pb ($Bqkg^{-1}$)	**46.5** (30–92)	**83.5** (32–258)	9.43	<0.01
^{232}Th ($Bqkg^{-1}$)	**45.9** (24–71)	**73.7** (52–130)	20.3	<0.001

Sand particles of sizes of 2000–50 μm were most abounded in the samples of ash, with the mean content in ash (56%) significantly higher ($p<0.0001$) than in soil (12%). The mean content of silt particles of sizes of 50–2 μm was significantly higher ($p<0.05$) in soil (54.7%) than in ash (36.8%), however individual fractions of silt, coarse (50–10 μm) and fine (10–2 μm), were not statistically different between the two substrates (Table 1). The main difference ($F=87.39$; $p<0.0001$) was observed for the mechanical fraction of clay of

particle sizes of <2 μm whose mean content was significantly higher in soil (28.3%) than in ash (3.6%). The study of the particle size distribution of the ash waste generated at the Tent A also showed a quite small representation of particles of the smallest sizes (≤2 μm) (Kostić et al., 2018). Although, distribution like that of mechanical fractions of samples of ash collected from the surface could also be influenced by the leaching of finer particles towards the deeper layers of passive lagoon during the period of rest. For comparison, mean values of all physicochemical features presented in Table 1. were very close to the one reported earlier for the surface soils (Tanić et al, 2016) and ash deposits (Kostić et al., 2018) in the vicinity of Tent A.

The values of activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th determined in the samples of ash and soil followed log-normal distribution. The natural radionuclides activity concentrations in soil were at the natural background gamma radiation levels and similar to the values reported earlier for the Tent A (Tanić et al., 2016). Ranges of activity concentrations in ash from the passive lagoons were comparable with the ranges determined in slag sampled below the boiler and in fly-ash samples collected at the electrostatic precipitators in TEK and Tent A (Kandić et al., 2014). The results of the ANOVA test (Table 1) showed that values of activity concentrations were always significantly higher ($p<0.01$) in ash than in soil. This was also concluded by similar study in which soil and disposed ash situated in Kaštela Bay, Croatia was compared (Skoko et al., 2017). However, mean values of activity concentrations (Bqkg^{-1}) found in the ash were about 10 times higher there (1099 for ^{238}U, 1074 for ^{226}Ra, 1165 for ^{210}Pb, 58 for ^{232}Th) than in this study (106 for ^{238}U, 103 for ^{226}Ra, 84 for ^{210}Pb, 74 for ^{232}Th), except for ^{232}Th.

It was noticed that activity concentration (Bqkg^{-1}) in the samples of soil in this study (38 for ^{238}U, 39 for ^{226}Ra, 47 for ^{210}Pb, 46 for ^{232}Th) was comparable to the one in coal used in CFPPs in Serbia (36 for ^{238}U, 37 for ^{226}Ra, 35 for ^{210}Pb, 49 for ^{232}Th) (Kandić et al., 2014). Furthermore, natural radionuclides activity concentration increase of 1.6 to 2.8 times from soil to the samples of ash in this study was similar to the increase of ~2 to ~4 times from coal to the fly-ash and slag after combustion. This indicated that differences of natural radionuclides activity concentrations between used coal and generated ash might to some extent be reflected in the environment as the differences of that between nearby soil and ash from passive lagoon.

To test this, activity concentration ratios within the same (^{238}U/^{226}Ra and ^{210}Pb/^{226}R) and between different decay chains (^{232}Th/^{226}Ra) were examined in this study and compared to the values of that for coal, slag and fly-ash used in Serbia (Kandić et al., 2014). No significant differences at the 95% confidence level were found in this study between soil and ash for ratios of ^{238}U/^{226}Ra approximately equal to ~1 and analogue to that, ^{238}U/^{226}Ra ratio values were only slightly changed from ~1 in coal to ~0.91 in fly-ash and slag. However, values of ^{210}Pb/^{226}Ra and ^{232}Th/^{226}Ra which were ~1 and ~1.3 in coal, respectively, both decreased significantly to the value of approximately 0.65 after combustion. In this study, ^{210}Pb/^{226}Ra was significantly higher ($p<0.05$) in nearby soil (1.25) then in ash from passive lagoon (0.88), but radioactive disequilibrium and its differences could occur in the environment due to additional dry or wet atmospheric deposition of ^{210}Pb of natural origin resulting from ^{222}Rn decay in the atmosphere. Finally, ^{232}Th/^{226}Ra ratio showed significant difference ($p<0.0001$) between nearby soil (1.20) and

ash from passive lagoon (0.74), indicating that this ratio could be used to differentiate unmodified from modified soil due to presence of ash.

^{232}Th/^{226}Ra activity concentration ratio

^{232}Th/^{226}Ra activity concentration ratios (Bqkg^{-1}/Bqkg^{-1}) were calculated in order to observe differences between nuclides activity concentrations of uranium and thorium decay chains in each sample. Range of calculated values was 0.49 to 1.52. It was noticed that those values were increasing with percentages of clay size particles which is why simple linear regression analysis was used to assess this effect. Linear relation between those two variables was found ($p<0.001$), however our analysis showed that two statistically significant linear models could be distinguished, as it is shown at Figure 1.

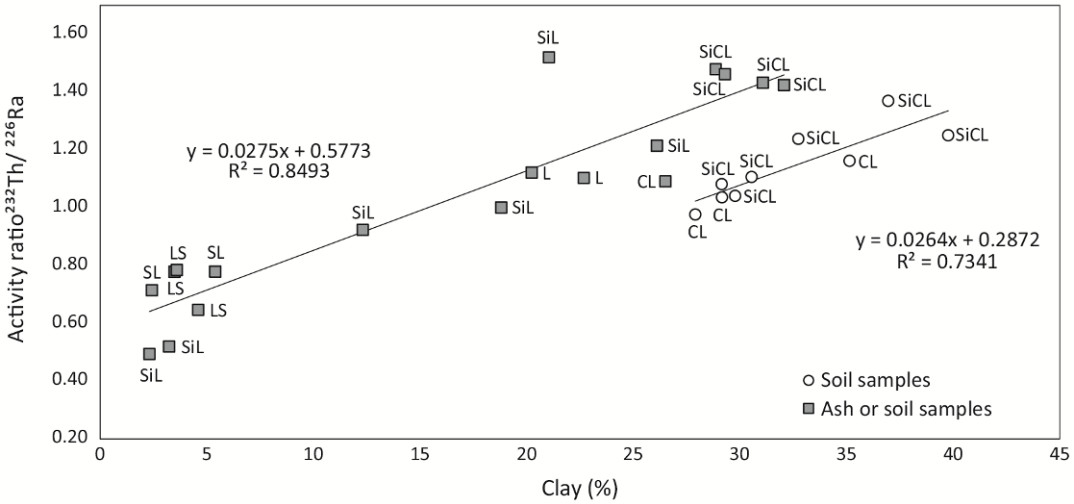

Figure 1. Changes of ^{232}Th/^{226}Ra activity ratios (Bqkg^{-1}/ Bqkg^{-1}) of investigated samples of soil and ash with percentages of clay (%)

Lower line at Figure 1 is fitted through the points which are represented by the soil samples whose mean ^{232}Th/^{226}Ra activity ratio value of 1.14 was close to the natural value of 1.1 observed in the environment (Wang et al., 2021). Based on the balanced ^{232}Th and ^{226}Ra activity concentrations, those samples could be characterised as unmodified i.e., representative soils that surround four coal fired power plants investigated. Their characteristic texture is found to be SiCL (mainly) or CL while clay fraction percentages were in the interval 27.9% to 39.8%.

From Figure 1. can be noticed that upper line is fitted through the points represented by the samples of ash and soil whose ^{232}Th/^{226}Ra ratio values mainly differed from natural being in the broad range from 0.49 to 1.52. First, a group of samples of ash taken from passive lagoon can be noticed with characteristic texture SL or LS while 0.67 was the mean value of their ^{232}Th/^{226}Ra ratio. Ash samples, rich in coarse particles (2000–50 μm), had small amount of clay (2.3% to 5.4%) as expected and there ^{226}Ra was more abounded than ^{232}Th as a consequence of coal combustion process and also reduced mobility of cations (such as Ra^{2+}) due to decreased soluble salt levels of ash (Skoko et al., 2017).

For the second group, which consisted of soil samples of L or SiL texture (Figure 1) with clay percentages from 12.3% to 26.5%, could be assumed that ash particles were incorporated into the soil and consequently changed its texture which became coarser compared to unmodified soil, but it was not the case according to their mean $^{232}Th/^{226}Ra$ ratio value of 1.14 which was the same as for unmodified samples. The rest of the samples were soils with SiCL texture as typical samples from lower regression line, but compared to them higher mean $^{232}Th/^{226}Ra$ value of 1.42 and a bit lower average clay content of ~30% suggested enhanced ^{226}Ra reduction from studied samples relative to ^{232}Th considering its known low mobility in the environment.

It could be noticed that both regression line slopes are approximately the same (α_1=0.0275 vs. α_2=0.0264) and that difference comes from the intercept which is about two times higher for „upper“ line (β_1=0.5773 vs. β_2=0.2872) indicating that $^{232}Th/^{226}Ra$ activity ratios are regularly increasing with clay content while ^{226}Ra removal from surface layers varies between samples. Comparing only samples of soil with SiCL texture, it appears that soils from „upper“ regression line are modified due to proximity of CFPPs and their ash disposal sites based on their higher radium's mobility and hence availability to plants which is different from soils from "lower" line where radium is part of more resistant fractions of soil. In accordance to that, a significant increase in the activity concentration in the samples of vegetation collected from the disposal sites of coal ash and slag was found only for ^{226}Ra and no significant difference was observed for ^{232}Th in comparison with control soil (Skoko et al., 2017). Additionally, unlike redistribution of naturally occurring radionuclides which was most likely took place in this study, no texture changes due to potential incorporation of ashes into the soils couldn't be observed.

CONCLUSION

Differences between samples of soil and ash collected in the proximity of coal fired power plants and coal ash disposal sites according to their physical and chemical properties are analysed in this study. No differences were found between soil and ash regarding $CaCO_3$ and humus content. Ash was weakly alkaline and the mean pH (in H2O) value of ash (7.9) differed significantly from that in soil (7.3). Mean percentage of sand (2000–50 μm) was significantly higher in ash (56%) than in soil (12%), but coarse silt (50–10 μm) and fine silt (10–2 μm) were not statistically different. Clay (<2 μm) mean content was significantly higher in soil (28.3%) than in ash (3.6%). Activity concentrations (Bq kg^{-1}) of naturally occurring radionuclides were also determined and means of ^{238}U, ^{226}Ra, ^{210}Pb and ^{232}Th were always significantly higher in ash (106.3, 102.9, 73.7 and 83.5, respectively) than in soil (38.2, 38.7, 45.9 and 46.5, respectively), as it was expected. Even though texture of investigated samples was predominantly silty clay loam, soils could be distinguished, according to our analyses, based on their different $^{232}Th/^{226}Ra$ activity concentration ratio: unmodified soil with the mean value of 1.14 almost equal to the natural value (~1.1) and modified soil with higher mean value of 1.42 suggesting enhanced ^{226}Ra reduction form surface layers due to proximity of CFPPs and ash disposal sites.

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