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The influence of a coal-fired power plant operation on radionuclide concentrations in soil

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Abstract

Fifty-two soil samples in the vicinity of a coal-fired power plant (CFPP) in Figueira (Brazil) were analyzed. The radionuclide concentration for the uranium and thorium series in soils ranged from <9 to 282 Bq kg^{-1}. The range of ^{40}K concentration in soils varied from <59 to 412 Bq kg^{-1}. The CFPP (10 MWe) has been operating for 35 years and caused a small increment in natural radionuclide concentration in the surroundings. This technologically enhanced natural radioactivity (TENR) was mainly due to the uranium series (^{234}Th, ^{226}Ra and ^{210}Pb) and was observable within the first kilometer from the power plant. The CFPP influence was only observed in the 0–25 cm soil horizon. The soil properties prevent the radionuclides of the ^{238}U-series from reaching deeper soil profiles. The same behavior was observed for ^{40}K as well. No influence was observed for ^{232}Th, which was found in low concentrations in the coal.

Keywords: Radionuclides; Soil; Coal power plant

1. Introduction

Mineral coal is composed of about 1% of trace elements and radionuclides. According to UNSCEAR (2000), the mean natural radionuclide concentrations in

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coal are 35 Bq kg^{-1} (range: 16–110) for ^{238}U, 35 Bq kg^{-1} (range: 17–60) for ^{226}Ra, 30 Bq kg^{-1} (range: 11–64) for ^{232}Th and 400 Bq kg^{-1} (range: 140–850) for ^{40}K.

Coal, burned as fuel material in power plants, produces energy and solid wastes, such as heavy and light ashes. The ashes tend to be enriched in inorganic elements (metals and radionuclides). The heavy ash fraction is deposited at the bottom of the furnaces, and is called bottom ash. The light ash (fly ash) is carried through the furnaces with the gases flow toward the stack. Depending on the emission control system of the stack, most of the ash is collected and any leftover is released into the atmosphere and deposited on the soil around the coal-fired power plant.

The amount of ash released into the atmosphere is variable: in old plants around 10% is released, while in modern plants with sophisticated emission control equipment, the releases can be reduced to 0.5% of fly ash (UNSCEAR, 1988). During the coal burning process radon, a gas decay product of the U and Th series, is released into the atmosphere and is dispersed into the surrounding environment. Besides the potential radon inhalation problem, neighborhood soil contamination by the long-lived decay products, ^{210}Po and ^{210}Pb, has to be considered.

Concern over the environmental impact of the use of coal and oil in large-scale electrical power plants is not new. Beck (1989) observed that in most cases the activity released does not significantly impact the surrounding environment. However, recent publications have been concerned with the impact of environmental radioactivity from fossil fuel combustion (Papastefanou, 1996; Man-yin & Leung, 1996; Ayçik & Ercan, 1997; Jasinska, Mietelski, & Pociask-Karteczka, 1998; Bem & Bem, 1998). In Brazil, no data have been published so far about Brazilian coal-fired power plants. In addition, the increase of the Brazilian population necessitates new investments in alternative sources of energy other than hydro-energy. Coal-fired power plants are one possible solution, since 66% of the Brazilian natural mineral reserves of fuel are coal. Most of these reserves are located in the south of Brazil. An evaluation of Brazilian coal radioactivity showed values ranging from 24 to 35 Bq kg^{-1} for ^{238}U-series, 27–48 Bq kg^{-1} for ^{232}Th-series and 351–447 Bq kg^{-1} for ^{40}K (Moraes, Flues, & Mazzilli, 2000). One exception is the coal from the Figueira region, which contains higher radioactivity, 250 Bq kg^{-1} for ^{238}U-series; 30 Bq kg^{-1} for ^{232}Th-series; and 450 Bq kg^{-1} for ^{40}K. Fly ash contains 350 Bq kg^{-1} for ^{238}U-series (Moraes, Flues, & Mazzilli, 2000).

The present work evaluates the influence of fly ash deposition from the Figueira coal-fired power plant on radionuclide concentrations in soil profiles up to 50 cm in depth. The coal-fired power plant is considered old and small (10 MWe) and is located in Figueira county, in the North of Paraná State (Fig. 1). The plant has been operating for more than 35 years and, only recently, a filter system (cyclone and filtering bag–coupled filter system) was installed (July, 1998) to reduce the particulate emission through the 40-m stack. The coal used in the plant is from a nearby mine. An unexplored uranium mine also exists in the neighborhood (Fig. 1).

The evaluation of the environmental impact of this old installation is very important, since there is a new coal-fired power plant under construction in the same region and no data are available about the radioactive impact. This information is

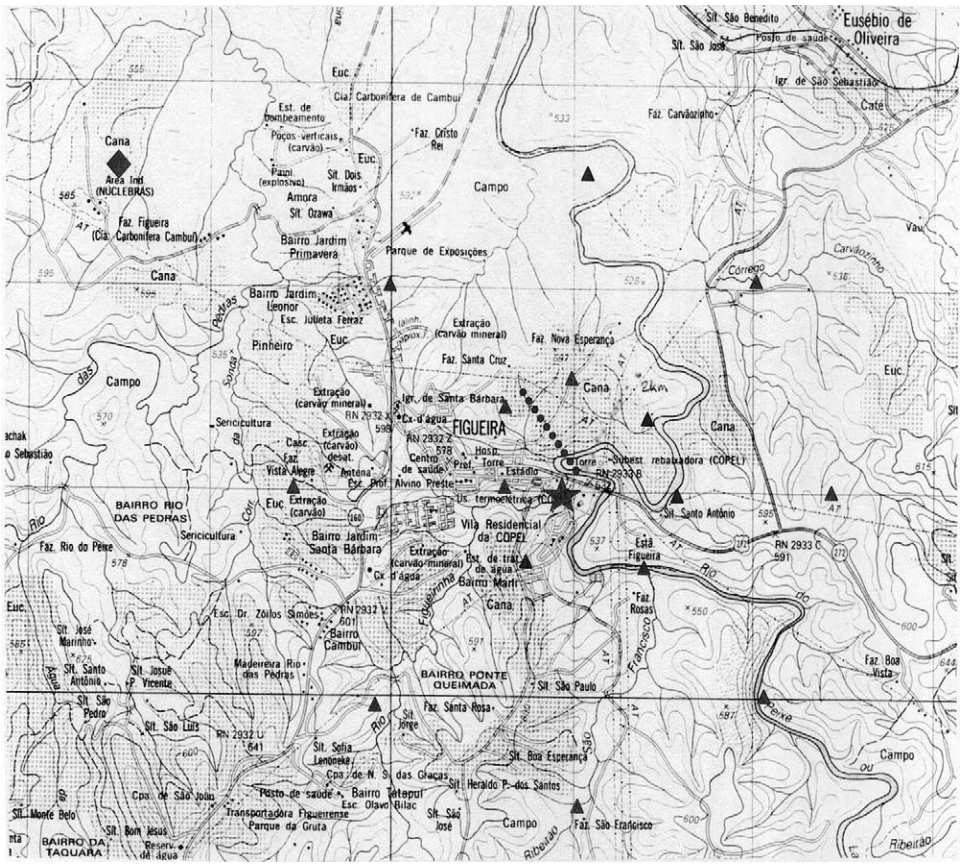

Fig. 1. 1. Map of Figueira region, with the location of the uranium mine, CFPP and soil sampling points. ★, coal-fired power plant (CFPP); ▲, 1 and 3 km transect sampling points; ●, <1 km transect sampling points; ◆, uranium mine. Scale=1:50,000; wind direction: NW.

important for authorities in their decisions about the viability of such an undertaking in Brazil.

2. Material and methods

Two soil-sampling runs were performed. Initially, 32 soil samples were collected (in December 1996) around the coal-fired power plant, at a distance of 1 and 3 km, in the principal compass directions (Fig. 1). A second sampling (December 1997) was performed along the northwest transect, considered the preferential wind direction (Fig. 1). These latter soil samples were collected at every 100 m from the coal-fired power plant up to 1 km. At each sampling location, soil samples were collected from two different profiles, named horizon A (depth from 0 to 25 cm) and horizon

B (depth from 25 to 50 cm), and approximately 1 kg of soil was collected. The samples were identified by compass direction, distance from the CFPP, and soil horizon (A or B).

All samples analyzed were grounded to 2 mm, homogenized, air dried and hermetically sealed in a cylindrical polyethylene vessel of 5 cm diameter and 5.5 cm height. ^{234}Th, ^{226}Ra, ^{232}Th and ^{40}K contents were measured by direct gamma spectrometry, with a coaxial germanium detector, EGNC 15-190-R, from Eurisys, with a resolution of 1.83 keV and relative efficiency of 19.1%, both measured at 1332 keV. The detector was calibrated using standard solutions with radionuclide activities certified by Amersham. The samples were sealed for about 4 weeks prior to measurement in order to ensure that equilibrium had been reached between^{226}Ra and its decay products of short half-life. The ^{226}Ra activities were determined by taking the mean activity of three separate photopeaks of its decay product radionuclides: ^{214}Pb at 295.2 and 351.9 keV, and ^{214}Bi at 609.3 keV. The ^{232}Th content of the samples was determined by measuring the intensities of the 911 and 968 keV gamma-ray peaks of ^{228}Ac, and the ^{40}K content from its 1460 keV gamma-ray peak. The concentrations of ^{210}Pb were obtained by measuring the activity of its low energy peak (46.5 keV), and of ^{234}Th by measuring the photopeak at 63.3 keV and 92.4+92.8 keV. Self-absorption correction was applied to low energy peaks (^{210}Pb and ^{234}Th), since the attenuation for low energy gamma rays is highly dependent upon sample composition. The self-absorption factor was calculated by the method suggested by Cutshall, Larsen, and Olsen (1983).

3. Results and discussion

Radionuclide (^{234}Th, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K) concentrations obtained in the two soil samplings, horizon A and B, are presented in Table 1 for the 1 and 3 km transects and in Table 2 for the <1 km transect. The radionuclide concentrations for the uranium and thorium series ranged from <9 to 282 Bq kg^{-1}. The range of ^{40}K concentrations varied from <59 to 412 Bq kg^{-1}. The mean values obtained for the activity concentration of all radionuclides analyzed showed a high coefficient of variation (28–63%), indicating that the results' dispersion was high.

There are few studies published about the radioactivity in Brazilian soil. Perez, Saldanha, Moreira, and Vaitsman (1998) found uranium concentrations from 0.01 to 26.4 Bq kg^{-1} and thorium from 0.01 to 971 Bq kg^{-1} in 15 different Brazilian soil samples. Assuming that radioactive equilibrium in the uranium series (^{238}U) had been established in the sealed samples, ^{238}U concentration could be deduced from the ^{234}Th concentration. Our results for soils collected at 1 and 3 km (Table 1) agree well with the data presented by Perez et al. (1998). Radionuclide concentrations from points located within 1000 m of the CFPP (Table 2), however, were higher than the average values, indicating an increase of radioactivity due to the power plant. Beck and Miller (1980) observed that usually the influence of air activity concentration at ground level is extended over about 1 km for short (50 m) stacks and over 5–10 km for taller (150 m) stacks.

Table 1
Concentrations of ^{234}Th, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K in soil samples collected at 1 and 3 km from the coal-fired power plant of Figueira, Brazil (Bq kg^{-1}). Sample locations defined by compass direction and depth of soil profile (A=0–25 cm; B=25–50 cm)

Sample	Th-234 1 km	*K*[a]	Th-234 3 km	*K*	Ra-226 1 km	*K*	Ra-226 3 km	*K*	Pb-210 1 km	*K*	Pb-210 3 km	*K*	Th-232 1 km	*K*	Th-232 3 km	*K*	K-40 1 km	*K*	K-40 3 km	*K*
N A	73±3	1.1	27±2	0.7	72±1	1.3	30±1	0.7	119±4	1.8	38±2	1	38±1	0.6	29±1	0.5	176±13	0.9	71±10	1.3
B	65±3		36±2		54±1		42±1		68±3		39±1		59±1		54±1		203±12		55±11	
NE A	50±3	1.5	24±2	0.3	41±1	1.3	31±1	0.6	60±3	1.5	51±3	1.4	43±1	1	24±1	3	176±12	0.7	85±11	1.4
B	33±2		<9.1		31±1		24±1		39±2		37±2		44±1		<8		256±12		<59	
NW																				
A	56±2	0.8	34±2	0.9	84±1	1.2	39±1	1	90±3	1.2	34±3	0.8	18±1	1	51±1	0.9	206±11	1.2	225±15	0.8
B	71±3		39±2		69±1		38±1		74±3		42±3		18±1		58±1		176±10		289±15	
E A	28±2	2.8	29±2	1.4	18±1	1.2	34±1	0.9	42±3	4.2	56±3	0.9	14±1	1.1	35±1	0.8	93±10	1.2	104±12	0.7
B	10±2		21±2		15±1		37±1		10±2		60±3		13±1		43±1		74±9		146±14	
S A	64±3	1.5	25±2	0.7	37±1	1	26±1	1.3	95±3	2.3	62±3	1.3	32±1	0.9	31±1	3.1	184±11	0.9	107±11	0.6
B	42±2		33±2		37±1		20±1		41±3		47±1		37±1		10±1		205±11		174±12	
SE A	48±2	1.4	39±2	1.1	34±1	0.9	29±1	1.1	61±3	1.7	26±2	2.4	31±1	0.9	23±1	1	158±11	1	211±14	1
B	33±2		36±2		37±1		26±1		36±3		<11		35±1		23±1		158±10		215±12	
SW A	61±3	1.2	66±3	2.4	49±1	1.2	72±1	1.4	42±3	0.8	99±3	1.3	40±1	0.8	21±1	1.2	225±12	0.8	145±13	1.1
B	50±2		27±2		42±1		52±1		50±3		75±3		52±1		18±1		291±12		126±12	
W A	78±3	1	31±2	1	60±1	1	49±1	0.8	79±3	0.8	42±3	1	31±1	1.3	26±1	0.7	184±11	1	328±15	1.2
B	81±3		32±2		65±1		47±1		96±3		43±2		24±1		35±1		179±11		281±14	
Mean±standard deviation																				
(A)	56±16		34±14		50±22		39±15		80±27		51±22		31±10		30±10		190±56		161±90	
(B)	48±24		28±13		44±18		36±11		52±27		43±20		35±16		30±21		182±60		161±102	

[a] *K*=radionuclide concentration in horizon A/radionuclide concentration in horizon B.

Table 2
Concentrations (Bq kg^{-1}). of ^{234}Th, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{40}K in soil samples collected in Northwest (<1 km) transect from the Figueira CFPP, in Brazil

Sample[a]	Th-234	*K*[b]	Ra-226	*K*	Pb-210	*K*	Th-232	*K*	K-40	*K*
200A	273±5	22.7	199±2	12.4	282±6	11.7	51±2	2.1	412±19	2.7
B	12±1		16±1		24±2		24±1		155±10	
300A	222±4	2.9	270±3	1.8	252±5	1.6	41±2	1	353±16	1.4
B	75±2		154±2		160±3		41±2		259±12	
400A	136±3	3.2	132±2	3.1	165±4	2.7	31±1	1	245±13	0.9
B	42±2		42±1		62±2		31±1		258±12	
500A	83±3	1.8	81±1	1.6	134±3	1.9	18±1	0.8	208±13	1.1
B	47±2		51±1		69±3		24±1		192±12	
600A	173±4	2.4	133±2	1.7	204±4	2	42±2	1.1	299±15	2.2
B	73±2		78±1		100±3		38±2		138±12	
700A	96±3	1.5	92±2	1.2	153±4	2	40±2	0.8	188±15	1.1
B	63±3		74±2		77±3		52±2		174±13	
800A	111±3	1.5	91±2	0.9	172±4	1.3	41±2	0.7	120±13	0.6
B	76±3		107±2		132±3		55±2		209±13	
900A	113±3	2	119±2	1.7	158±4	1.8	43±1	1	129±13	1.5
B	55±2		71±1		86±3		44±2		88±11	
1000A	104±3	1.8	125±2	1.9	169±4	2.2	47±2	1	211±14	1.7
B	56±2		65±1		77±3		45±2		126±12	
1100A	74±3	1.6	93±2	1.8	126±3	1.5	37±1	0.8	164±13	0.9
B	46±2		51±1		82±3		44±1		185±12	
Mean±standard deviation										
mean A	139±65		133±59		182±50		39±9		233±96	
mean B	55±19		71±38		87±38		40±11		178±55	

[a] The samples were named according to the distance from the CFPP and the horizon A and B.
[b] *K*=radionuclide concentration in horizon A/radionuclide concentration in horizon B.

Table 3
Physical and chemical soil characterization of horizon A

Soil sample	1–3 km range	mean±SD	<1 km range	mean±SD
% sand	23–82	51±15	24–61	39±11
% silt	11–44	25±9	28–42	36±5
% clay	3–38	23±9	6–40	24±11
% OM	0.7–3.9	2.1±0.9	2.7–6.4	4.7±1.2
pH	3.3–5.0	4.2±0.4	3.8–5.2	4.4±0.4
CEC	5–25	16±6	12–28	22±5

OM—organic matter, CEC—cationic exchange capacity (meq. 100 g^{-1}).

The soil of the region is classified as yellowish-red podzolic and considered as an acid soil. The physical and chemical characterization assays are presented in Table 3. It is well known that soil properties such as texture, pH, cationic exchange capacity (CEC) and organic matter contents may influence the adsorption of metals and radionuclides (Van Driel & Nijssen, 1988).

To evaluate the distribution of the activity concentrations obtained for the several radionuclides in soils collected from the <1, 1, and 3 km transects, a box plot graphic representation was applied (Fig. 2). This methodology allows a visualization of the radionuclide concentrations among different sampling locations, the range of data

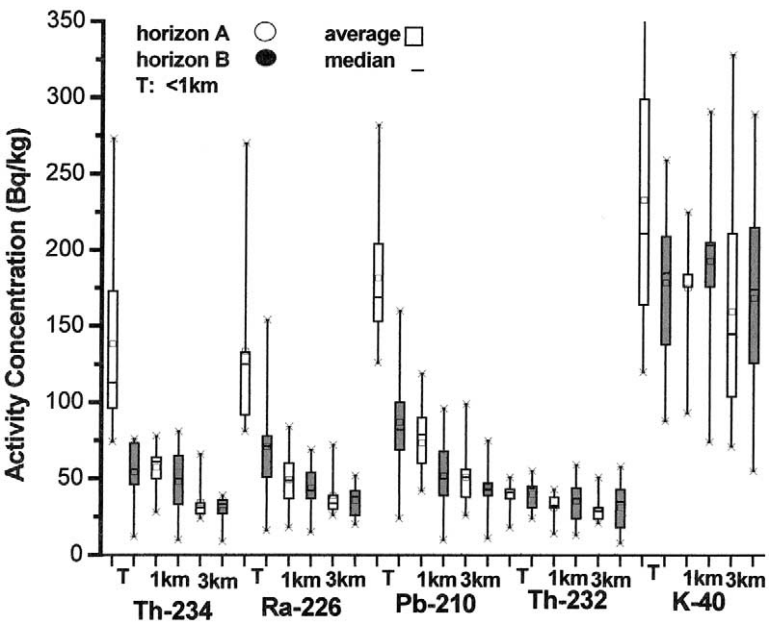

Fig. 2. "Box plot" graphic representation of soil natural radionuclides concentration.

variation, average and median concentrations, as well as comparisons among different radionuclides.

The results presented in Fig. 2 show higher concentrations for the radionuclides of the uranium series (^{234}Th, ^{226}Ra and ^{210}Pb). Lower concentrations were observed for ^{232}Th. It can also be seen in Fig. 2 that the mean concentrations for ^{234}Th, ^{226}Ra, ^{210}Pb and ^{40}K decrease with the distance from the discharge point up to 1 km. No influence was detected for ^{232}Th. This radionuclide was also observed in low concentration in the composition of the coal. The application of a Student "*t*" test showed that there was no statistical difference between ^{232}Th activity concentrations of the samples taken 1–3 km away from the CFPP ($p<0.05$), i.e., the concentrations were statistically equal, indicating that in this case the influence of the CFPP activity was insignificant.

The concentration of radionuclides ^{234}Th, ^{226}Ra and ^{210}Pb should follow the same pattern since they belong to the same decay series. However, in Fig. 2 higher concentrations were found for ^{210}Pb compared with other radionuclides of the uranium series (^{234}Th, ^{226}Ra). Besides the transport of Pb-210 adsorbed in fly ashes, there is an additional concentration explained by the direct emanation and escape of radon through the stack. The half time of ^{222}Rn is short (3.8 days), decaying to ^{210}Pb, which can be adsorbed more easily and precipitate with dust particles. This behavior explains why ^{210}Pb presents higher activity concentrations in soil in the vicinity of the coal-fired power plant.

For samples collected <1 km from the CFPP, concentrations of ^{234}Th, ^{226}Ra, ^{210}Pb and ^{40}K in horizon A (empty box) were significantly greater than those in horizon B (full box, Fig. 2). No statistical difference was observed between the two horizons for samples collected at 1 and 3 km.

With data of Tables 1 and 2, the ratio between the radionuclide concentrations of horizon A and B (K=A/B) were calculated. The mean ratios obtained for samples collected <1 km from the CFPP were 1.8±0.7 for ^{234}Th, 1.6±0.6 for ^{226}Ra, 1.7±0.5 for ^{210}Pb, 0.9±0.1 for ^{232}Th and 1.3±0.5 for ^{40}K. The ratios confirmed an increase in the radionuclide concentrations of the uranium series and ^{40}K in the upper soil layer (horizon A) within the close vicinity of the CFPP. The result of point 200 A was not considered in the ratio value determination. It was verified that this point represents an alluvial sediment of a river located in the region. Therefore, the samples collected at point 200 are not representative of horizon A and B, as the other points. The presence of higher radionuclide concentrations in sample "A" compared with sample "B" indicates that the fly ash being continuously deposited was washed out by each river flood and according to Echevarria, Sheppard, and Morel (2000) the strong acidity of the soil (pH=4.4±0.4) reduces the migration of uranium, preventing infiltration to the lower layers.

Available data from CFPP-Figueira showed that the amount of coal burned in the power plant between 1986 and 1997 (before the installation of filters) was 25,440 t year^{-1}. No data are available concerning the amount of ash generated. However, it is known that coal from this region has 26% ash content, so this information indicates a production of 6614 t year^{-1} of ash. Only a small part of this ash is considered bottom ash and the remainder is fly ash, with different particle sizes. Due to the lack

of fly ash found, it can be assumed that 50% (3307 t $year^{-1}$) was carried out from the 40 m stack and was deposited in the surrounding soil. Beck (1989) observed that old plants with a 50 m stack presented an increase in the soil radioactivity up to 1000 m. Indeed, it was observed that the soil was greyish in colour between 200 and 600 m, indicating possible contamination of the soil in the vicinity of the CFPP.

To estimate the discharge dispersion of the fly ash a computer model based on the Pasquille equation was applied. The conditions assumed for the application of the model were stack height: 40 m; discharge of fly ash: 3.307 t $year^{-1}$; wind speed: 1 m s^{-1}, deposition rate: 1 cm s^{-1} (Beck & Miller, 1980) and dispersion sector: 22.5°. By applying the model, a maximum deposition of 2.1 kg m^{-2} $year^{-1}$ at 400 m was obtained.

The experimental results obtained for the concentration ratio (K=A/B in Table 2) at point 400 m was greater than the other points, with the exception of anomalous point 200 m, thus confirming the conclusion obtained by using the computer model.

A Pearson correlation matrix with all the calculated K ratios from Tables 1 and 2 was constructed. The correlation coefficient of K ratios for the data from the <1 km transect are given in Table 4. The highest correlation coefficients were obtained for the radionuclides of the uranium series, indicating that the transect point contamination probably was caused by the same pollutant source, fly ash deposition during the 35 years of plant activity. In the case of ^{40}K, the correlation values suggest that the contamination was probably not only from one source; the potassium-rich fertilizer and the soil composition (feldspar) should be considered in addition to the fly ash from the CFPP. The Pearson correlation for K ratios of the 1 and 3 km data (Table 1) reached maximum values of r=0.62.

4. Conclusions

Thirty-five years of operating the 10 MWe coal-fired power plant, as studied in this work, caused a two- to three-fold increase in the natural radionuclide concentrations, mainly of the uranium series (^{234}Th, ^{226}Ra and ^{210}Pb), in soils within 1 km of the facility. At a distance greater than 1 km, statistically significant increases were not observed. The coal-fired power plant influence was more observable in horizon A soil. The same behavior was also observed for ^{40}K.

In conclusion, significantly increased concentrations within the upper soil profile

Table 4
Correlation coefficients (r) of samples from the <1 km transect

	Th-234	Ra-226	Pb-210	Th-232
Ra-226	0.99	–	–	–
Pb-210	0.99	0.99	–	–
Th-232	0.95	0.93	0.93	–
K-40	0.66	0.64	0.74	0.66

were detected within 1 km of the CFPP for the technologically enhanced radionuclides ^{234}Th, ^{226}Ra, ^{210}Pb and ^{40}K, but not ^{232}Th. The latter was found in low concentrations in coal. This information should be considered by authorities in the planning of a newer and larger coal-fired power plant in the region.

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