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Evaluation of radionuclides concentration in Brazilian coals

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Abstract

Raw material samples of coal used by the main power plants in Brazil (RS; SC; PR) were evaluated. Radiological relevant radionuclides of the natural series, such as ^{238}U, ^{228}Ra, ^{226}Ra, ^{210}Pb and ^{40}K were analyzed by direct gamma spectrometry. The results showed that the uranium concentration in coal from (PR) was at least three times higher than concentration found in other coal samples in Brazil (RS and SC). In spite of the high uranium concentration in the PR coal, the small (10 MW_e) local power plant currently operating in (PR) produced low amount of ashes, however more detailed studies should be done for the new power plant (140 MW_e) in project.

Keywords: Coal; Radionuclides; Brazil

1. Introduction

Coal plays an increasingly important role to cover the energy needs of Brazil, representing 66% [1] of the national energetic resource. Environmental issues are important for the development of new coal-fired power plants. The uncontrolled release of coal burning byproducts can increase the concentration of toxic metals and radionuclide in the environment, therefore the evaluation of the amount of radioactivity in coal is important.

During the 70s and 80s Brazilian coal distribution and occurrence were evaluated. The coal deposits are concentrated in South of Brazil 90.9% in Rio Grande do Sul State (RS) (Candiota coalfield is the largest one with 8 billion t), 8.5% in Santa Catarina State (SC), 0.5% in Paraná State (PR) and only 0.04% in São Paulo State [2].

The radioactivity of components of the earth's crust is mostly due to the presence of ^{40}K and the radionuclides that form the three natural radioactive series (^{238}U, ^{235}U and ^{232}Th). These radionuclides are distributed uniformly and their variations depend on the rock type. The average concentration of ^{238}U, ^{232}Th and ^{40}K in coal is estimated to be 20, 20 and 50 Bq kg^{-1}, respectively, based on analysis of coal samples from 15 countries, with a variation of more than two orders of magnitude [3].

The combustion of the coal in coal-fired power plant (CFPP) leads to an increase of the natural radionuclides, and non-combustible elements, with enrichment factors for the ashes of 5–10 times [13].

The increase in specific activities of naturally occurring radionuclides in combustion products, fly ash and bottom ash, compared to that of the original coal, depends primarily on the inorganic fraction of the coal (ash content). Eisenbud and Petrow [4] first pointed out that the radiation dose from the use of fossil fuel for power generation could be a significant addition to the natural radiation dose.

According to UNSCEAR [5] published data, the annual collective effective equivalent dose to public estimated for the coal combustion cycle was approximate 54,000 man Sv $year^{-1}$. The world nuclear energy generation caused collective effective equivalent dose to public of 200 man Sv GW_e^{-1}. Considering that the world nuclear energy generation in 1987 was 189 GW $year^{-1}$, the annual collective effective equivalent world dose to public was 37,800 man Sv $year^{-1}$. This data showed that coal energy

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generation can cause effective collective equivalent doses that need attention. Therefore doses produced by uranium and thorium present in coal cannot be ignored and need more detailed studies about the concentration of radionuclides in coal used in coal plants.

Although several papers have been published concerning to radioactivity in coal around the world [6–9], Brazil has few data published so far about the radioactivity in their main coal mines. The purpose of this study is to evaluate the radioactivity concentration of coal from the main Brazilian coal mines and to estimate the radioactivity concentration of ashes produced by the operation of the coal power plants.

1.1. Geological and geographical context

The Brazilian deposits of energetic coal occur at the eastern border of the Paraná Sedimentary Basin, within the Rio Bonito Formation, of Permian age, in a variable number of layers at the different occurrence areas, as well as with varied thickness.

The South Brazilian carboniferous belt occurs in a sinuous strip, of width of some dozens of kilometers, of direction east–west in the State of São Paulo, inflecting to the South, in Paraná, outlining the Crystaline Basament at the Ponta Grossa Arch. Deeping in the south of Santa Catarina, in Torres Syncline, it outcrops in Rio Grande do Sul in an arch that outlines the Riograndense Shield. The outcropping area of the Gondwanic layers, in which it is the Rio Bonito Formation, which contains the coal layers [10].

The coal layers occur in sub-horizontal position, being, sometimes, affected by faulting and by intrusive diabase bodies. The mining of the bodies, depending on the local conditions, is done in open pits, hillside galleries and underground mines of, in the maximum, 300 m of depth. According to the carbonification degree, the south-Brazilian coals are classified as sub-bituminous coal high volatile types, with high tenor of ashes and variable tenor of sulfur.

It should be emphasized that uranium occurrences are known in Permian rocks of Paraná Basin from Brazil to Uruguay, being Figueira the most important one. The uranium is thought to have come from the weathered rocks of the basement and from reworked sediments of the Itararé Formation, which contains boulders of crystalline basement. The uranium was probably transported in its oxidized state by slightly alkaline ground waters to reducing environments. The uranium is associated with sandstones, siltstones, clays and carbonaceous sediments, including coal. In the sandstones the uranium occurs in the form of uraninite in carbonate cement filling the interstices. In the siltstones, carbonaceous clays and coals the uranium is present as organic complexes, classified as uranocircite. The uranium deposit at Figueira, with an average grade of 0.148% U_3O_8, occurs in sedimentary beds of calcitic sandstone above the coal seam.

2. Experimental part

To evaluate the level of radioactivity of the Brazilian coals, ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K contents were determined in ROM coal samples (coal run of mine, without treatment) from the beds Candiota, Iruí and Leão/Butiá Mines, in Rio Grande do Sul State (RS); São Geraldo Mine – Siderópolis, in Santa Catarina State (SC); and Cambuí Mine, Figueira, in Paraná State (PR). The raw coal samples (ROM) were taken from mine output. A pulverized coal of Figueira power plant (PR) was also measured. These coals are used as fuel in the thermoelectric plants Presidente Médici, São Jerônimo and Charqueadas, in Rio Grande do Sul; Jorge Lacerda, in Santa Catarina; and Figueira in Paraná.

Sixteen samples of coal from each region were ground to 200 mesh, homogenized, air dried and hermetically sealed in a cylindrical polyethylene vessel of 5 cm diameter and 5.5 cm height. ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K contents were measured by gamma spectrometry, with a coaxial germanium detector, EGNC 15-190-R, from Eurisys, with a relative efficiency of 15% for the photopeak of ^{60}Co at 1332 keV. The detector was calibrated using standard soils with radionuclides activities certified by Amersham. The samples were sealed for about four weeks prior to the measurement in order to ensure that equilibrium has 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 keV and 351.9 keV, and ^{214}Bi at 609.3 keV. The ^{228}Ra content of the samples was determined by measuring the intensities of the 911 keV and 968 keV gamma-ray peaks of ^{228}Ac, the intensity of the 238 keV gamma-ray peak of ^{212}Pb and the intensity of the 583 keV gamma-ray peak ^{208}Tl. ^{40}K content was measured by the 1460 keV gamma-ray peak. The concentration of ^{210}Pb was carried out by measuring the activity of its low energy peak (46.5 keV) and that of ^{238}U by measuring the ^{234}Th photopeak at 63.3 keV. Self-absorption correction was applied to the 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 et al. [11].

The total metal content of uranium and lead in pulverized coal was determined by wavelength dispersive X-ray fluorescence spectrometer RIX 3000 (Rigaku Co, Osaka, Japan). Around 0.9 g of coal samples were mixed with boric acid (9:1) and grounded carefully to 200 mesh. The powder was pressed with hydraulic press to obtain a double-layer pressed pellets.

3. Results and discussion

The results obtained for the activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K in Brazilian coals from different regions (PR, SC, RS) are listed in Table 1. The

Table 1
^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K concentration in Brazilian coal (Bq kg^{-1})

	^{238}U	^{226}Ra	^{210}Pb	^{228}Ra	^{40}K
ROM-PR					
Average	356 ± 304	321 ± 199	808 ± 650	22 ± 7	191 ± 77
Range	159–807	135–698	249–1745	15–32	136–245
ROM-SC					
Average	107 ± 70	98 ± 32	340 ± 328	58 ± 36	556 ± 584
Range	42–181	42–144	91–712	36–100	208–1230
ROM-RS					
Average	32 ± 15	31 ± 4	75 ± 30	26 ± 5	267 ± 144
Range	18–48	27–35	41–98	20–30	181–433
Pulverized coal-PR					
Average	1111 ± 183	995 ± 178	2252 ± 283	32 ± 8	313 ± 84
Range	882–1325	813–1251	1859–2609	24–41	200–450

range obtained for the activity concentration of ^{238}U, ^{228}Ra and ^{40}K in ROM Brazilian coals is compared with world coals data from UNSCEAR [12]. The results obtained in this study, present higher concentrations for natural uranium series (from 18 to 807 Bq kg^{-1}) and for ^{40}K (from 181 to 1230 Bq kg^{-1}) compared to the world range for ^{238}U 10–600 Bq kg^{-1} and for ^{40}K 30–100 Bq kg^{-1} [13]. The uranium and lead X-ray fluorescence technique evaluation of coal showed average value of 1016 ± 230 Bq kg^{-1} and 80 ± 20 mg kg^{-1}, respectively.

The $^{238}U/^{232}Th$ ratio for coal around the world was calculated using data from UNSCEAR [12], for the majority of the coal samples this range varied between 1 and 3.5. Some world coals showed higher ^{238}U concentration with $^{238}U/^{232}Th$ ratio ranging from 5 to 33 (examples: coal from Austria, Greece, Hungary and Yugoslavia [12]). This data was compared with the results obtained in this paper for the $^{226}Ra/^{228}Ra$ ratio. The observed ratio for Brazilians coals from RS and SC (Table 1) showed normal world values, but PR coal showed higher values ($^{226}Ra/^{228}Ra$ ratio around 13 for ROM coal and 33 for pulverized coal). In this coal, uranium is enriched over thorium, probably due to two reasons: higher mobility of uranium compared to thorium during ground and surface water transport and the vicinity of a uranium mine.

To evaluate the asymmetry distribution of the activity concentration values obtained for the several radionuclides in coal of Brazil, a Box Plot graphic representation was applied (Fig. 1). This methodology allows a visualization of the results dispersion in different samples, the range of data variation, the average and the median, as well as comparison between different radionuclides.

From the results presented in Table 1, it can be concluded that the coal from PR showed the highest concentration among the Brazilian coals. The ROM coal is grounded and washed to separate the pyrite fraction, before its use in the power plant. The pulverized coal of PR was measured and the uranium series concentration showed an increase by a factor of around 3. It is known that coal from this region has 26% of ash content, and also a high quantity of pyrite (7%). Thorium series and ^{40}K had

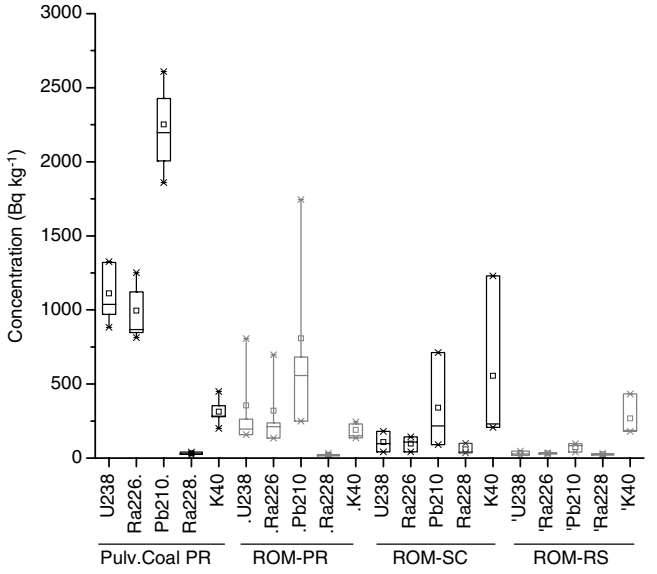

Fig. 1. Box plot representation of the activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K (Bq kg^{-1}) for the ROM coal from Figueira (PR), Santa Catarina (SC), Rio Grande do Sul (RS) and pulverized coal from Figueira (Pulv.Coal PR). □ : the average (–) the median.

smaller enrichment factor, about 1.5 and 1.6, respectively. This factor suggests that uranium and thorium series are distributed differently in coal and probably have different geochemical properties.

ROM coal and its associated mineral matter contain most of the naturally occurring elements at significant concentration levels. Coal is cleaned by gravity separation to reduce ash and pyrite content. To separate coal from pyrite (and other heavy metals and minerals), it is desirable to take advantage of the large difference in their specific gravities: the specific gravity of coal is 1.2, while the specific gravity of pyrite is 5.0. During the physical cleaning of coal, elements are partitioned among the clean coal (pulverized coal), the coal waste and the process water used for cleaning. Certain elements associated with the organic fraction of coal should be enriched in the clean coal, whereas water-soluble minerals should become enriched in process water. Conzemius et al. [14] evaluated the partitioning factors of elements during cleaning operation of coals and observed an enrichment factor of 2.7 for uranium in clean coal.

According to Breger [15], uranium enters coal bed from ground water as the soluble sodium uranyl decarbonate ($Na_2[UO_2(CO_3)_2]$) or the sodium uranyl tricarbonate ($Na_3[UO_2(CO_3)_3]$) complex. Breger also suggests that this slightly acidic environment of the coal decomposes theses complexes, and the uranium quickly becomes absorbed by the coal with subsequent reduction to uraninite (UO_2). If sufficient silica is present in solution, the mineral coffinite ($U(SiO_4)_{1-x}$–$(OH)_{4x}$) is formed instead of uraninite. Since the uranium is quite dispersed in the coal, it must have migrated from cracks and joints for some distance into the coal matrix. Uranium can be expected to reside in coal

in a very highly dispersed state, and be mineraralized as both uraninite and coffinite, depending on the composition of the original mineralizing.

Pires et al. [16] studied the geochemical distribution of trace elements in coal. Data from coal of Pittsburgh, Pocahontas and Spain listed in this work showed that uranium tends to be associated to organic fraction and non-sulfide fraction of coal.

The ratios $^{238}U/^{226}Ra$ and $^{210}Pb/^{226}Ra$, for uranium series in ROM and pulverized coal was calculated considering the data of Table 1. The average ratio for $^{238}U/^{226}Ra$ in pulverized and ROM coal were 1.2 ± 0.2 and 1.1 ± 0.2, respectively. These values suggest an approximate equilibrium between the parent and the ^{226}Ra in the uranium series, in coals from different locations. If the results of the ratio $^{210}Pb/^{226}Ra$ (average values of 2.3 ± 0.2 and 2.6 ± 0.9 in pulverized and ROM) are taken into account, it is seen that there is not a secular equilibrium in the sub series, probably due to emanation of ^{222}Rn that migrates into or out of the coal seam, the half-time of ^{222}Rn is short (3.8 days) and decays to ^{210}Pb. These appear to represent atypical situation, generally ignored in most assessments. Similar behavior was also found in few report [9,17,18] .

The Pearson correlation matrix ($p < 0.05$) for all radionuclides in the ROM and pulverized coal showed a good correlation ($r > 0.80$) for the uranium series. The correlation between ^{210}Pb and stable Pb showed a very poor correlation ($r \sim 0.1$), suggesting that ^{210}Pb and stable Pb occurrence in coal have different minerals origin. The ^{238}U concentration in the pulverized coal obtained by gamma spectrometry ($U_{\gamma spec}$) was compared with the uranium concentration obtain by wavelength dispersive X-ray fluorescence spectrometry (U_{XRF}) for the same samples. The ratio $U_{\gamma spec}/U_{XRF}$ was around 1.1, with very good correlation ($r \sim 0.90$). These values confirm that both techniques are suitable for the U determination.

In order to estimate the amount of radioactivity present in the waste produced by the coal power plant in Brazil, data concerning capacity, coal consumption, ash content, enrichment factor and ash production for each power plant in Brazil are showed in Table 2. The data concerning capacity, coal consumption and ash content were taken from literature [1] and the enrichment factor was calculated by taking into account the average ash content of each coal.

The activity concentration present in the ashes produced in the Brazilian power plants was calculated multiplying the enrichment factor (EF) presented in Table 2 by the activity concentration of each radionuclide in the coal of the locations studied (Table 1). The results for the activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra, and ^{40}K (Bq kg^{-1}) in ash produced by coal power plants of Brazil are presented in Table 3. This results showed that the activity concentration of the uranium series in coal ashes from RS, SC and PR, ranged from 62 to 162 Bq kg^{-1}; from 294 to 1020 Bq kg^{-1}; and from 1284 to 3232 Bq kg^{-1}, respectively. It can be seen that the ash produced by the PR coal plant presents activity concentrations for the uranium series at least 3 times higher than the other power plants in Brazil. However the activity concentrations of ^{228}Ra (174 Bq kg^{-1}) and ^{40}K (1668 Bq kg^{-1}) were two times higher in the SC ash. According to UNSCEAR [12], the world average concentrations in fly ashes are 265 Bq kg^{-1} for ^{40}K, 200 Bq kg^{-1} for ^{238}U, 240 Bq kg^{-1} for ^{226}Ra, 930 Bq kg^{-1} for ^{210}Pb and 70 Bq kg^{-1} for ^{232}Th. Comparing the present results with the UNSCEAR [12] data, PR ash presented higher activity concentration for the uranium series and SC ash presented higher values for ^{228}Ra (^{232}Th series) and ^{40}K.

A recent evaluation of PR ashes [19] presented high concentrations for uranium series, following the same pattern that in coal. The ashes showed activity concentration ranging from 1459 to 5198 Bq kg^{-1} for ^{238}U; from 1442 to 3773 Bq kg^{-1} for ^{226} Ra; from 3292 to 14,641 Bq kg^{-1} for ^{210}Pb; from 43 to 124 Bq kg^{-1} for ^{228}Ra (^{232}Th series) and from 471 to 1144 Bq kg^{-1} for ^{40}K. The highest concentrations were observed at the finest ashes particles. ^{210}Pb appears to be the most volatile radionuclide measured and the lead tends to be enriched on the finer fly ash particles.

Table 2
Brazilian power plants capacity, coal consumption and ash content from (SUMÁRIO MINERAL-DNPM; 1999) [1]

Power plant	Capacity (MW_e)	Coal consumption ($\times 10^3$ t year^{-1})	Ash content (%)	Enrichment factor (EF)	Ash production ($\times 10^3$ t year^{-1})
RS-state					
Charqueadas	72	432	Average: ~50% Range: 40–57%	2	216
Pres. Médici	446	2676			1338
São Jerônimo	20	120			60
Total	538	3228			1614
SC-state					
Jorge Lacerda	857	5142	Average: ~35% Range: 40–30%	3	1543
PR-state					
Figueira	10	60	Average: 25% Range: 20–30%	4	15

Table 3
Activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K in ash produced by coal power plants of Brazil (Bq kg^{-1})

	RS-ash	SC-ash	PR-ash	PR*-ash
^{238}U	64	321	1424	2222
^{226}Ra	62	294	1284	1990
^{210}Pb	150	1020	3232	4504
^{228}Ra	52	174	88	60
^{40}K	534	1668	764	626

PR* ash produced from pulverized coal.

The Brazilian regulatory agency published recently a guideline concerned with the radiological protection of activities which may lead to enhanced concentrations of radionuclides (CNEN-NN-4.01) [20]. Such activities may include, for instance, the mining and processing of ores or coal burning as well as storage of raw material, products, by-products, residues and wastes containing radionuclides of the U and Th series, simultaneously or separately, which may incur undue exposures of members of the public and occupationally exposed. All the power plants studied present in their ashes activity contents below 10 Bq g^{-1}, limit adopted by the regulation for category III. These values should exempt such installations from further control by the Brazilian regulatory agency, if the doses to the workers occupationally exposed are below 1 mSv per year.

The coal used in power plants is pulverized and burned inside the boiler, producing bottom ash (15–20%), which falls inside the boiler, and fly ash (80–85%). The combustion of coal leads to an increase of the natural radionuclides. 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 [12]. The installation of ash collectors in Brazilian power plants like cyclone and bag filters reduced significantly the emission of radionuclides to the atmosphere. In contrast, treatment and disposal of power plant ashes continues to be a serious problem, mainly in Brazil where the coal have a high ash content (20–50%) (Table 2), the coal combustion residues are stored at the plant sites or on-site disposal, and only a small part of them have commercial use (30%).The main application of coal ash in Brazil is the manufacture of cement and concrete (30%). It is also used in small percentage as a road-bed stabilizer, in asphalt mix, building materials, glass composites and ceramic. The large fraction of coal ash of Brazilian power plant that does not find a commercial application is usually stored in piles, or dumped in the vicinity of the power plant.

An evaluation of data about the amount of ash produced in Brazilian coal plants in Table 2 showed that the RS e SC power plants ($\sim 15 \times 10^5$ t year^{-1}) produced 100 times more ashes than PR (15×10^3 t year^{-1}). In spite of the low amount of ashes produced per year by the PR power plant of Figueira, Table 3 showed that the coal used in this power plant is 3 times more radioactive than the coal from SC and can be a problem for the surrounding environment, and for the disposal and reused application of these ashes.

The current power plant in Figueira is a small one (10 MW$_e$) and so the amount of ash production is low. However, since a larger power plant (140 MW$_e$) is in project, a more detailed study of the environmental impact should be carried out.

4. Conclusions

The results obtained in this work showed that the uranium series concentration in coal from Figueira (PR) reached values until 1745 Bq kg^{-1} and was at least three times higher than concentrations found in other coals samples in Brazil (RS and SC). Owing to the high ash content (20–50%) of Brazilian coal, the estimation of the activity concentration in ashes showed high activity concentration for the uranium series predominantly for PR coal ashes, reaching values from 1284 to 3232 Bq kg^{-1}. In spite of the high uranium concentration in the PR coal, the small (10 MW$_e$) local power plant currently operating in Figueira (PR) produced low amount of ashes, however more detailed studies should be done for the new power plant (140 MW$_e$) in project.

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