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Radioactivity characteristics of lignite and hard coals: Case study of Zonguldak (Kozlu), Konya (Karapınar) and Antalya (Pamucakyayla) in Türkiye

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Abstract. In this study, the radioactivity contents of Carboniferous Zonguldak-Kozlu hard coals, Carboniferous Antalya-Pamucakyayla hard coals and Pliocene Konya-Karapınar lignites were determined, then compared with the limit values determined by the authorized institutions and their hazard indexes were determined. The range of measured activity concentrations was from 16.2 Bq·kg^{-1} to 227.6 Bq·kg^{-1} for ^{238}U, 20.6 Bq·kg^{-1} to 67.5 Bq·kg^{-1} for ^{232}Th and 211.9 Bq·kg^{-1} to 515.5 Bq·kg^{-1} for ^{40}K. The calculated mean absorbed gamma dose rate (D), radium equivalent activity (Ra_{eq}) and annual equivalent dose (AED) were 105.7 nGy·h^{-1}, 227.9 Bq·kg^{-1} and 129.6 μSv·h^{-1}, respectively. Although ^{238}U and ^{232}Th radionuclide activity concentrations are comparable to literature values, ^{40}K activity concentrations were around three or four times higher than in UNSCEAR (2000) reports. As it poses a radiological risk, it is necessary to take the necessary precautions to reduce the negative effects on the environment and human health due to use of coals from Konya-Karapınar, Antalya-Pamucakyayla basins and to burn in a controlled manner.

Keywords: Coal • Hard coal • HPGe • Lignite • Radioactivity • Türkiye

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Introduction

Coal, one of the fossil-based energy sources in the world, is important because it is more easily accessible and versatile than petroleum [1]. Türkiye has a total of 20.8 billion tons of coal resources (1.8% of the world's coal reserves), including 1.5 billion tons of hard coal and 19.3 billion tons of lignite (7.1% of the world's reserves) [2, 3]. In Türkiye, lignites are produced from underground and open pit mining operations. Lignites have low calorific value and are generally used as fuel in thermal power plants (TPP). Most of the lignite basins of Türkiye are located in the Western Anatolia, Central Anatolia and Thrace regions. In this study, radioactivity contents of lignites in Pliocene Konya-Karapınar, hard coals in Carboniferous Antalya-Pamucakyayla and Zonguldak-Kozlu sites were evaluated (Fig. 1).

The three elements naturally occurring in coal, which also have radioactive isotopes, are uranium (U), thorium (Th) and potassium (K). It is the K major element in lignites and is the main element of many rocks, and almost all of the potassium in coal shows an inorganic relationship [4]. Swaine

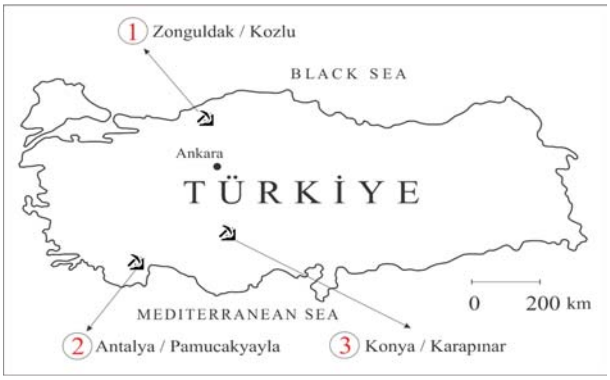

Fig. 1. Location map of coalfields.

[5], Dai *et al*. [6] emphasized that the modes of occurrence of uranium in coal are diverse, but that organic origin appears to be common. A substantial amount of thorium found in coal originates from commonly abundant phosphate minerals such as monazite and apatite [5].

The extraction, transportation and combustion processes of coal can be hazardous to human health and the environment because of the heavy metal and naturally occurring radionuclide content, which vary based on the geological formation of the material [7–11]. Furthermore, it has been documented that the separation of volatile components during coal combustion enriches radioisotope concentrations by up to 10 times [12–16]. For the protection of public health and environmental quality, it is necessary to know the radionuclide contents of the feed coals when using coal wastes for the production of cement and other building materials, as aggregate in stabilizing roadways or filling underground cavities, building road/rail embankments and reinforced earth walls, mining filling, agriculture, etc. [12–14]. In addition to handlers (miners and TPP employees), this information is crucial for evaluating the radiological danger to the general public near TPP and final users. Approximately 12–15% of Turkey's lignite and hard coal production is produced from these three fields, and no data on activity concentrations and analyses of radiological risk for these coalfields were available in the literature. This study was carried out to determine the radionuclide activity concentrations and radiological risks that may arise from the uranium, thorium series and potassium (in terms of the health of both the miners and the population living around the TPP and coal mine sites).

Geological background

Zonguldak-Kozlu coalfield is located in the Istanbul Zone, north of Türkiye [17]. The zone is divided into two subdivisions and the study area is situated in the Istanbul-Zonguldak zone. The Istanbul-Zonguldak zone developed on the southern margin of Laurasia during the Devonian-Carboniferous (Fig. 2) [21]. The carbonate sedimentation starting from the Middle Devonian was interrupted [22, 23]. Coal formation started with the abundance of plants in Namurian. A new transgression occurred in the Early Barremian and most of the coal units in the Zonguldak basin remained below sea level [24]. With the compression tectonics developing in the region, the Zonguldak basin started to rise again. With the complete closure of the Neotethys, the Zonguldak basin rose completely above sea level and attained its present form. The erosion period that started in the Lutesian is still continuing [24].

The Konya-Karapınar coalfield is located in the Konya Plain in the north of the Taurus Orogenic Belt. The region is defined as 'Central Anatolian Plains Region' based on its tectonic position [23]. Konya region is morphologically characterized by the dominant NW–SE and N–S trending elevations in the north and the basins formed by grabens. Sultandağları, Emirdağları and Ekecikdağları to the east of Tuz Lake constitute important elevations; Akşehir graben, Beyşehir graben, Cihanbeyli plateau and Aksaray basin constitute important tectonic depressions [25]. N–S and E–W trending Bozdağlar in the west of the study area and Obruk Plateau in the north are important morphological elements. In the south of the region, the Taurus Mountains, which have a high elevation compared to the region, are located. In this section, the NE–SW trending Bolkar Mountains and NW–SE trending Ozyurt Mountains are the main elevations [25]. The main morphology of the region has been shaped by normal faults with different trends and dominant lateral thrusts that bound the elevations and basins [25]. As a result of these block faults, the uplifted sections formed mountainous areas and the collapsed sections formed basins. While the age of basement rocks ranges between Palaeozoic and Eocene periods, deformed and fractured due to the Palaeotectonic regime surface on the elevations, the depression basins between the elevations are dominated by Miocene-Holocene aged lacustrine, terrestrial and volcanic rocks (Fig. 2) [25].

The study area is located within the Antalya Nappes in the south and southwest of Antalya. In the north of Kumluca (southwest of Antalya), the unit named as the Kumluca zone and Kumluca complex found Halobia and Daonella fossils in the stratified cherts, which were suggested to be Jurassic--Cretaceous in age, and its age was determined as Middle-Upper Triassic (Ladinian–Norian) [26]. Palaeozoic-Lower Mesozoic carbonate-clastic rocks of the Antalya Union mainly contain calcite, dolomite, quartz, feldspar, goethite and phyllosilicate minerals, and rarely co-occurred with jarosite, hematite, goethite, gypsum, barite or siderite. Silicate (analcime, chlorite, chlorite-smectite, tetranatrolite, tetranatrolite, hydroxyapophyllite), carbonate (calcite) and feoxide/oxy-hydroxide (hematite, goethite) minerals were observed in the matrix of volcanic rocks or matrix observed in the form of intrusions and lenses within the Silurian-Triassic aged sedimentary units. The amount of quartz and feldspar increases in the Ordovician and dolomite in the Cambrian and Permian-Lower Triassic. Barite, siderite and jarosite were found in Cambrian (Çaltepe), Ordovician (Sarıyardere) and Permian (Pamucakyayla), respectively. The amount of kaolinite increases in

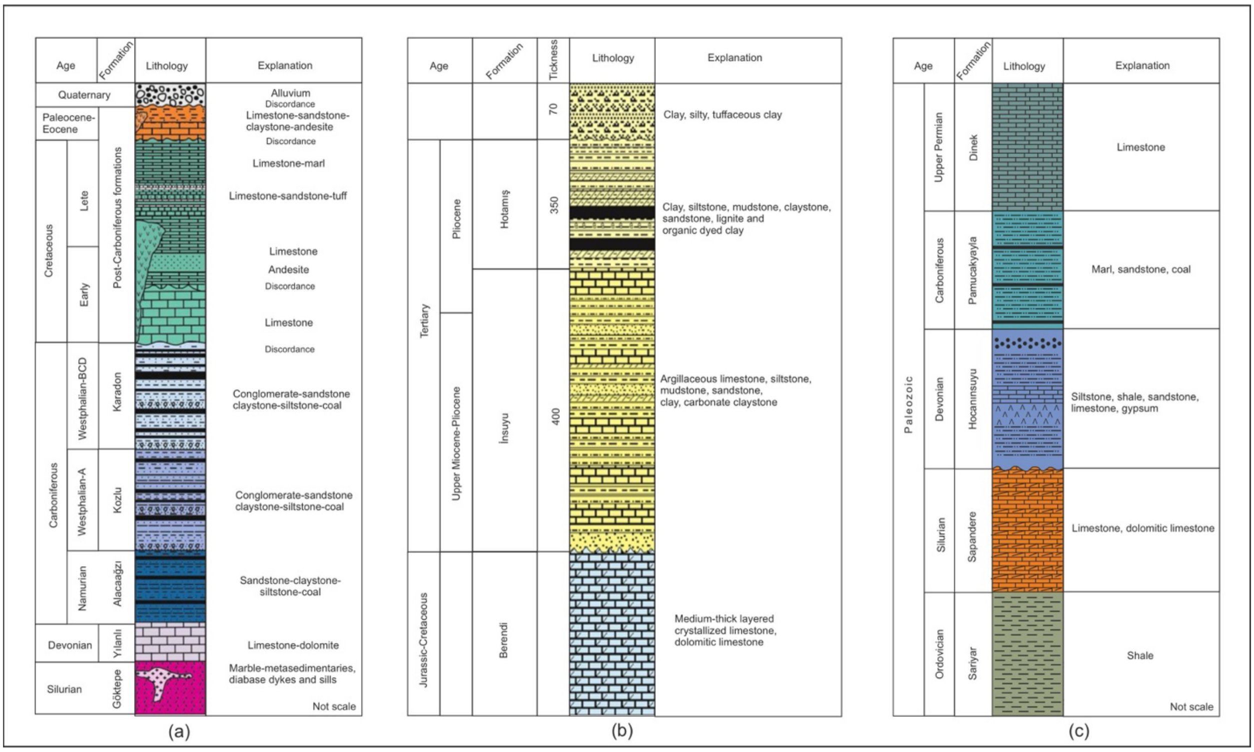

Fig. 2. General stratigraphic section of study area (a) Zonguldak-Kozlu [18], (b) Konya-Karapınar [19], (c) Antalya--Pamucakyayla [20].

Permian, illite-smectite and smectite in Devonian--Triassic aged units [27] (Fig. 2).

Material and methods

The samples in the Konya-Kar apınar basins were taken as core samples from the drillings selected to represent the basin, and hard coal samples of Zonguldak-Kozlu and Antalya-Pamucakyayla were taken from mine galleries. Twelve samples were taken from each field to represent the general characteristics of the coalfields.

The coal samples were numbered and labelled after they were transferred to the sample preparation laboratory of Akdeniz University, Faculty of Science, Department of Physics. Foreign substances and impurities in each sample were removed. All samples were homogenized with the grinding machine and then sieved through a 2-mm mesh in the sample preparation laboratory. The sieved samples were then filled into hermetically sealed (6 cm × 5 cm) 150 ml polyethylene cylindrical containers, labeled, weighed and stored for 4 weeks in order to reach secular equilibrium between ^{226}Ra and ^{222}Rn prior to counting.

Radioactivity measurement was conducted by using a n-type, coaxial, high-purity germanium (HPGe) gamma-ray detector AMETEK-ORTEC with 768 eV full width half maximum (FWHM) at 122 keV for ^{57}Co and 1.85-keV FWHM at 1332 keV for ^{60}Co. It is connected to a digital electronic unit HEXAGON-CAEN and a computer. The detector was placed into a 10-cm thick lead shield with an inner surface covered by a 2-mm thick copper foil to shield it from the X-rays originating from lead. Data acquisition and analysis were carried out with MC^2 software.

All samples were placed on the front face of the detector and counted for 50 000 s. Background intensities were obtained with an empty beaker for 50 000 s under the same conditions before and after the samples' measurement. Then, the average of the background counts was subtracted from the sample spectrums. ^{238}U and ^{232}Th activity concentrations were indirectly determined from their daughter products, while ^{40}K were determined directly by their gamma-ray peaks. To determine the activity concentration of the ^{238}U nuclide, daughter nuclides ^{214}Pb and ^{214}Bi were used, while ^{228}Ac concentration was chosen for the parent ^{232}Th. 351.9 keV ^{214}Pb and 609.3 keV ^{214}Bi energy peaks were used to determine the concentrations of ^{238}U. 911.2 keV ^{228}Ac energy peak was used to determine the concentration of ^{232}Th and 1461.0 keV energy peak was used to determine the concentration of ^{40}K. IAEA-375, IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1 reference materials were used for the energy and efficiency calibrations (Table 1). Details of the activity and dose calculations were presented by [28].

Two-way ANOVA analyzes were performed to compare statistically radionuclide concentration between regions, using Orange software [29].

Table 1. Summary of the analysis of standard materials

Standard	Reference value (Bq·kg^{-1})	Measured value (Bq·kg^{-1})
RGU-1	4 940 ± 30	4 958 ± 70
RGTh-1	3 250 ± 90	3 280 ± 67
RGK-1	14 000 ± 400	14 543 ± 459

Table 2. Descriptive statistics for ^{238}U, ^{232}Th and ^{40}K activity concentrations of coal samples

Site: Zonguldak-Kozlu (12 samples)		^{238}U (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
Min		16.2	20.6	283.9
Max		25.5	39.2	375.7
Mean SEM		19.5 ± 3.1	29.5 ± 3.4	321.7 ± 54.1
	25	17.6	23.2	290.5
Percentile	50	18.9	29.3	319.2
	75	20.7	34.6	351.5
Site: Konya-Karapınar (12 samples)		**^{238}U (Bq·kg^{-1})**	**^{232}Th (Bq·kg^{-1})**	**^{40}K (Bq·kg^{-1})**
Min		109.4	30.5	308.8
Max		227.6	52.0	515.5
Mean SEM		181.2 ± 18.0	43.7 ± 5.5	432.3 ± 65.5
	25	156.3	35.5	389.6
Percentile	50	186.2	46.4	436.5
	75	203.4	51.0	475.5
Site: Antalya-Pamucakyayla (12 samples)		**^{238}U (Bq·kg^{-1})**	**^{232}Th (Bq·kg^{-1})**	**^{40}K (Bq·kg^{-1})**
Min		113.1	28.2	211.9
Max		219.5	67.5	373.1
Mean SEM		159.6 ± 12.3	45.0 ± 6.4	302.1 ± 50.2
	25	138.0	35.3	261.7
Percentile	50	160.8	45.9	320.5
	75	181.7	50.7	339.2

Result and discussion

Measurement results of the coal samples indicate only the existence of ^{238}U, ^{232}Th and ^{40}K radionuclides. ^{238}U, ^{232}Th and ^{40}K radionuclide activity concentrations of hard coal and lignite samples collected from Zonguldak (Kozlu), Konya (Karapınar) and Antalya (Pamucakyayla) were measured. The calculated ^{238}U activity concentrations ranged from 16.2 ± 0.9 Bq·kg^{-1} to 227.6 ± 13.4 Bq·kg^{-1}, ^{232}Th activity concentrations ranged from 20.6 ± 1.3 Bq·kg^{-1} to 67.5 ± 3.8 Bq·kg^{-1} and ^{40}K activity concentrations ranged from 211.9 ± 8.4 Bq·kg^{-1} to 515.5 ± 25.6 Bq·kg^{-1}. Descriptive statistics for ^{238}U, ^{232}Th and ^{40}K activity concentrations of coal samples were presented in Table 2.

When the ^{238}U concentrations of the coalfields are compared with the literature, the Zonguldak-Kozlu coalfield is comparable to Ankara-Beypazarı (16 Bq·kg^{-1}) [30], Amasya-Suluova (22 Bq·kg^{-1}) [30], Bursa-Orhaneli (15 Bq·kg^{-1}) [31], Çorum-Alpugat (11 Bq·kg^{-1}) [30], Çanakkale-Çan (15 Bq·kg^{-1}) [31], Erzurum (17 Bq·kg^{-1}) [31] and Istanbul-Kemerburgaz (14 Bq·kg^{-1}) [32] and is at a level well below the worldwide results of Greece-Megapolis (530 Bq·kg^{-1}) [33], Brazil-Figueira (321 Bq·kg^{-1}) [34], Bangadesh-Barapukuria (54.3 Bq·kg^{-1}) [35] and Greece-Megapolis (173 Bq·kg^{-1}) [36]. On the other hand, it is observed that in Antalya-Pamucakyayla and Konya-Karapınar coalfields ^{238}U concentration levels are higher than the fields in Türkiye except for Muğla-Kemerköy (213 Bq·kg^{-1}) [37], at the same level as Greece-Megapolis (173 Bq·kg^{-1}) [36] and lower than Brazil-Figueira (321 Bq·kg^{-1}) [34] and Greece-Megapolis (530 Bq·kg^{-1}) [33] in the world.

It was determined that ^{232}Th activity concentration levels of the coalfields were generally comparable to the results given in the literature for Türkiye: Ankara-Beypazarı (10 Bq·kg^{-1}), Çorum-Osmancık (13 Bq·kg^{-1}), Kayseri-Seyit Ömer (37 Bq·kg^{-1}) [30], Bolu-Göynük (12 Bq·kg^{-1}), Bursa-Orhaneli (40 Bq·kg^{-1}), Afşin-Elbistan (49 Bq·kg^{-1}), Manisa-Soma (47 Bq·kg^{-1}), Tekirdağ-Saray (30 Bq·kg^{-1}) [31] and for the world: Greece-Megapolis (13 Bq·kg^{-1}) [33] and Greece-Megapolis (31 Bq·kg^{-1}) [36].

When coal fields were examined in terms of ^{40}K activity concentration level, it was determined that the findings were approximately 3–4 times higher than the studies conducted in Türkiye: Muğla-Kemerköy (130 Bq·kg^{-1}) [37] and Erzurum (113 Bq·kg^{-1}) [31]; and were generally higher than the values in the literature around the world: China-Baqiao (105.7 Bq·kg^{-1}) [38] and Romania-Oltenia (154 Bq·kg^{-1}) [39] and were at a level comparable to the study conducted in Greece-Megapolis (372 Bq·kg^{-1}) [36] and Bangadesh-Barapukuria (241 Bq·kg^{-1}) [35].

In terms of ^{238}U concentration, ANOVA analysis revealed a statistically significant difference (ANOVA: 139.811 [P = 0.000, N = 36]) between Zonguldak-Kozlu basin and the other basins.

Since the probability of the presence of these elements in organic matter is low when their close relationship with geological events is examined, their appearance at the determined level suggests that they may probably be caused by the geochemical processes that occurred during coalification.

Table 3. Descriptive statistics of radiological risk parameters

Site	Samples	D (nGy·h^{-1})			Ra_{eq} (Bq·kg^{-1})		
		Min	Max	Mean	Min	Max	Mean
Zonguldak-Kozlu	12	33.3	47.9	40.7 ± 5.8	40.8	58.8	49.9 ± 8.9
Konya-Karapınar	12	90.9	173.8	140.7 ± 39.3	111.6	213.2	172.5 ± 39.7
Antalya-Pamucakyayla	12	107.5	144.1	128.2 ± 27.4	131.9	176.7	157.2 ± 23.5

Site	Samples	Annual equivalent dose (μSv·year^{-1})			H_{in}		
		Min	Max	Mean	Min	Max	Mean
Zonguldak-Kozlu	12	69.7	103.4	86.4 ± 20.2	0.23	0.35	0.32
Konya-Karapınar	12	192.7	381.3	304.1 ± 64.1	0.82	1.59	1.13
Antalya-Pamucakyayla	12	234.3	311.5	276.9 ± 56.6	0.94	1.40	1.25

Hökerek [40], Altunsoy *et al.* [41] and Koca *et al.* [42] state that the elements K, U and Th are positively correlated with ash and negatively correlated with total organic carbon, and are generally associated with silicate and clay minerals.

By using the radionuclide activity concentrations, radiologic risk parameters: the absorbed gamma dose rate (D), the radium equivalent activity (Ra_{eq}), the annual equivalent dose (AED), external hazard index (H_{ex}) and internal hazard index (H_{in}) of the coal were also calculated and descriptive statistics of radiological risk parameters estimated based on activity concentration of lignite and hard coal samples were presented in Table 3.

The calculated mean absorbed gamma dose rate (D), were ranged from 33.3 nGy·h^{-1} (Zonguldak-Kozlu) to 173.8 nGy·h^{-1} (Konya-Karapınar), radium equivalent activity (Ra_{eq}) was from 40.8 Bq·kg^{-1} (Zonguldak-Kozlu) to 213.2 Bq·kg^{-1} (Konya-Karapınar), AED levels were from 69.7 μSv·h^{-1} (Zonguldak-Kozlu) to 381.3 μSv·h^{-1} (Konya-Karapınar). For Zonguldak-Kozlu, all internal (H_{in} <1.0) and external (H_{ex} <1.0) hazard indices were less than unity. Gamma dose rate values of all samples were in between the 10 nGy·h^{-1} and 200 nGy·h^{-1} range, radium equivalent activity and annual effective dose values of all samples were lower than the worldwide average levels 370 Bq·kg^{-1} and 460 μSv·year^{-1}, respectively [43]. But For Konya-Karapınar and Antalya-Pamucakyayla basins, external (H_{ex} <1.0) hazard indices were less than or equal to unity, but internal (H_{in} >1.0) hazard indices were higher than unity.

Conclusion

In this study, lignite and hard coal samples of Zonguldak-Kozlu, Konya-Karapınar and Antalya-Pamucakyayla basins were analyzed for radioactivity with regard to ^{238}U, ^{232}Th and ^{40}K isotopes. The measured radioactivity levels of coal samples from Zonguldak-Kozlu were compatible with those reported from Turkey and neighbouring countries. But measured radioactivity levels of coal samples from Konya-Karapınar and Antalya-Pamucakyayla basins were two- to four-fold higher than the world average (^{238}U: 35, ^{232}Th: 30 and ^{40}K: 400 Bq·kg^{-1}) of UNSCEAR (2000) report [43]. The possible source of relatively high radionuclide concentrations is mineral deposits in the vicinity of the region. The calculated (D) gamma dose rate values for the investigated region are in between the 10 nGy·h^{-1} and 200 nGy·h^{-1} range. Similarly calculated mean radium equivalent activity (Ra_{eq}) 80.03 Bq·kg^{-1} and annual effective dose with an average 46.29 μSv·year^{-1} are lower than the worldwide average values (370 Bq·kg^{-1} and 460 μSv·year^{-1}) which implies that the radiation hazard is insignificant for the population living in the investigated area.

Although the radiological risk parameters are below the recommended limit values, radioactivity concentrations of some coal samples are above the world average. Because it may pose a radiological risk, it is necessary to take necessary precautions to reduce the negative effects on the environment and human health from the use of coal in the Konya-Karapınar and Antalya-Pamucakyayla basins and to burn it in a controlled manner. It is important to keep the coal mines, where a significant portion of the fuel is used for electricity production, under control at regular intervals, considering that these values may increase further due to the geological conditions (layers enriched in U, Th and K concentrations).

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Conflicts of interest. The authors declare that there is no conflict of interest.

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