

The Investigation of Radionuclide Concentrations in Soil and Lifetime Cancer Risk Due to Gamma Radioactivity in Zonguldak, Turkey

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The distributions of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs activity concentrations were determined using a high-purity germanium detector in uncultivated surface soil samples collected from Zonguldak. The range of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs activity concentrations varied from 10.5 ± 2.0 to 98.0 ± 4.3 Bq kg⁻¹ (mean value 28.8 ± 4 Bq kg⁻¹), from 12.9 ± 3.4 to 53.2 ± 5.1 Bq kg⁻¹ (mean value 29.6 ± 3 Bq kg⁻¹), from 190 ± 22 to 808 ± 43 Bq kg⁻¹ (mean value 395 ± 31 Bq kg⁻¹) and from < 0.5 to 47.1 ± 3.8 Bq kg⁻¹ (mean value 12.5 ± 2 Bq kg⁻¹), respectively. In addition, the mean outdoor absorbed dose rates in air (terrestrial, terrestrial+cosmic), annual effective dose equivalent, radium equivalent activity and external hazard index were found to be 48 nGy h⁻¹ (terrestrial), 56 nGy h⁻¹ (terrestrial+cosmic), 58 μSv y⁻¹, 101 Bq kg⁻¹ and 0.27 , respectively. The evaluated excess lifetime cancer risks (ELCRs) varied from 0.13×10^{-3} to 0.36×10^{-3} with a mean value of 0.20×10^{-3} .

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1. Introduction

Environmental natural gamma radiation is formed from terrestrial and cosmic sources. The terrestrial component of the natural background radiation depends on the compositions of soil and rocks containing natural and artificial radionuclides [1, 2]. An important contribution to total dose from natural sources comes from terrestrial radionuclides, such as ²³⁸U, ²³²Th and ⁴⁰K, existing in soil [3, 4]. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soil and rocks, due to the difference in the geological structure of the region, plays an important role in radiation protection and measurement. Besides naturally occurring radionuclides, many radionuclides of artificial origin have been released into the environment by different processes. The isotope ¹³⁷Cs is one of them and it is produced anthropogenically by several types of nuclear activities. Cosmic radiation originates from cosmic rays, whose contribution to background changes mainly with altitude and latitude.

Background radiation measurement in Zonguldak province was previously performed in surface soils around the Çatalağzi coal-fired power plant in Zonguldak, by Aytakin and Baldık [5, 6]. The main objective of this study was to identify and to determine natural radionuclide activity concentrations in soil samples collected from 22 locations around the province of Zonguldak and to evaluate the annual effective dose and lifetime cancer

risks from outdoor terrestrial radiation. In addition, the natural gamma radiation coming from terrestrial radionuclides in air was also investigated in the Zonguldak region. To the researcher's knowledge, this is the first study to assess the level of the background radiation in Zonguldak province.

Zonguldak province is situated between $41^{\circ} 00''$ – $41^{\circ} 35''$ N latitude and $31^{\circ} 18''$ – $32^{\circ} 19''$ E longitude. The Zonguldak Basin, which contains a thick Carboniferous clastic sequence with several coal seams, is located in the northwestern Turkey on the Black Sea coast. It is predicted that the coal reserves in Zonguldak coal basin are about 1.1 billion tons of coal and from a mining operations point of view, Zonguldak Basin is the most important production area in Turkey [7]. These features make Zonguldak an interesting candidate for radiological studies.

2. Materials and methods

The sampling procedure was done as described by Akkaya et al. [8]. Sampling locations are shown in Fig. 1.

Prepared soil samples were placed in acrylic cylindrical containers with dimensions of 6 cm \times 5 cm. The containers were closed and stored for a month in order to achieve the radioactive equilibrium between ²²⁶Ra and its daughter element (²²²Rn) [9]. ⁴⁰K, ²³²Th, ²²⁶Ra and ¹³⁷Cs activity concentrations were measured using a p-type coaxial HP Ge detector (ORTEC/GEM110P4) with a relative efficiency of 107.7% and resolution of 1.92 keV at 1.33 MeV (with associated electronics produced by EG&G Ortec). The detector was surrounded by a 9.25 cm thick lead shield. For activity of ²³⁸U and ²³²Th the following gamma lines were used; ²³⁸U: 186 keV

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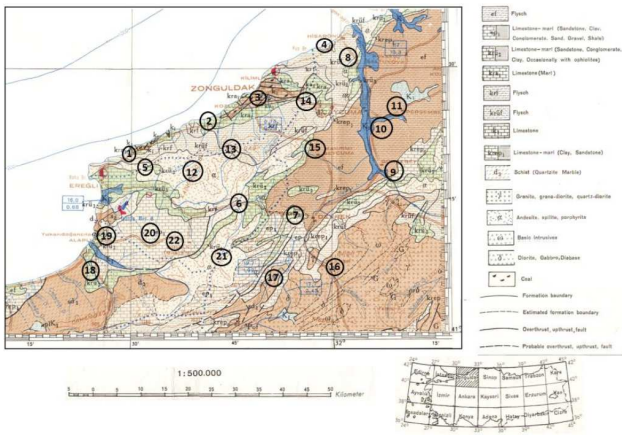


Fig. 1. Study area and sampling sites.

(^{226}Ra), 295 keV and 352 keV (^{214}Pb), 609 keV, 1120 keV and 1764 keV (^{214}Bi); ^{232}Th : 583 keV (^{208}Tl), 911 keV (^{228}Ac). The activity of ^{40}K was determined directly by its 1460 keV gamma line and the activity of ^{137}Cs was determined by its 662 keV gamma transition. The detector calibration was performed using standards in radioactive equilibrium, purchased from International Atomic Energy Agency. The minimum detectable activity of the detector for ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs was found to be 2.2, 6.7, 15.5 and 0.5 Bq kg^{-1} , respectively, for 20 000 s counting time.

The outdoor gamma radiation levels were measured by a portable dose rate meter (Fluke Victoreen survey meter, connected with a 489-55 model $1.5'' \times 1.5''$ NaI(Tl) scintillation detector, optically coupled to PMT, as described by Akkaya et al. [10].

3. Results and discussion

The activity concentrations of the radionuclides in 22 surface soil samples are given in Table I. The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K have varied in the range from 10.5 ± 2.0 to 98.0 ± 4.3 Bq kg^{-1} , from 12.9 ± 3.4 to 53.2 ± 5.1 Bq kg^{-1} , and from 190 ± 22 to 808 ± 43 Bq kg^{-1} , respectively. The average activity concentrations found in the study are around the world averages, reported by UNSCEAR [11] as 35, 30 and 400 Bq kg^{-1} for ^{226}Ra , ^{232}Th and ^{40}K , respectively.

The two highest ^{226}Ra activity concentrations were found as 98.0 and 59.2 Bq kg^{-1} for the samples collected from the Kokaksu and Buruncuk respectively. At the same time, the two biggest measurements for ^{232}Th are 53.2 and 51.4 Bq kg^{-1} , from the Buruncuk and Kokaksu respectively. The reason for the high ^{226}Ra and ^{232}Th activity concentrations can be explained by the fact that there are coaly clastic rocks in these areas. It is known that coals sometimes include high contents of natural radioactive elements (U, Th, and their decay products) and, in some cases, concentrate considerable amounts of the uranium [12, 13]. The four highest ^{40}K activity

concentration are 808, 561, 561 and 541 Bq kg^{-1} , which were found in soil samples collected from the Soğanlıyörük, Beycuma, İsabeyli and Alpaslan respectively. The reason of the high ^{40}K activity concentrations can be explained with the existence of limestone and clay in this region. In general, limestones, clays and shales contain relatively high concentration of potassium [14, 15].

The ^{137}Cs activity concentration ranges from < 0.5 to 47.1 ± 3.8 Bq kg^{-1} . It was found that ^{137}Cs activities of the soil samples collected from Kokaksu, Düzpelit and İsabeyli are higher than activities of the others. ^{137}C is a fission product. It has been spread by a nuclear explosion or a reactor accident. The northwest of Turkey was also contaminated because of Chernobyl reactor accident. Due to the lack of data before the Chernobyl accident, it is not possible to compare ^{137}Cs concentrations originating from the previous nuclear weapon tests and Chernobyl accident by means of the data obtained for Zonguldak.

Absorbed dose rate in air (ADRA) depends on the activity concentrations of ^{238}U , ^{232}Th and ^{40}K natural radioisotopes ADRA (nGy h^{-1}) was calculated using the following relation [11]:

$$\text{ADRA} = 0.462C_{\text{U}} + 0.604C_{\text{Th}} + 0.041C_{\text{K}}, \quad (1)$$

where ADRA is the dose rate in air at 1 m above the ground, and C_{U} , C_{Th} and C_{K} are the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq kg^{-1} of the soil sample, respectively.

ADRA values and the measured dose rates are given in Table I. The obtained mean absorbed dose was found to be 48 nGy h^{-1} . The average measured outdoor gamma dose rate (terrestrial and cosmic ray) is 56 nGy h^{-1} , which is higher than the average of the calculated. This difference is to be expected since the absorbed doses are calculated only for outdoor terrestrial gamma radiation, whereas the measured doses have cosmic contributions as well. The result obtained by the study is near the world average value (~ 51 nGy h^{-1} [11]).

$$\text{AEDE} = \text{ADRA} \times \text{DCF} \times \text{OF} \times \text{T}, \quad (2)$$

where, DCF is the dose conversion factor (0.7 Sv Gy^{-1}), OF is the outdoor occupancy factor (0.2) and T is the time factor (8760 h y^{-1}) [11]. AEDE values are given in Table I. The calculated annual mean effective dose equivalent was found to be 58 $\mu\text{Sv y}^{-1}$.

To assess the radiological risk, it is useful to calculate an index called the Radium equivalent activity, Ra_{eq} in Bq kg^{-1} . It is calculated through the following relation [16].

$$\text{Ra}_{\text{eq}} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}}, \quad (3)$$

where C_{Ra} , C_{Th} and C_{K} are the mean activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K (in Bq kg^{-1}) of the soil sample, respectively. Values of Ra_{eq} , calculated using Eq. (3) are given in Table I. The calculated mean radium equivalent activity was found to be 101 Bq kg^{-1} . The recommended maximum value of Ra_{eq} is 370 Bq kg^{-1} [16]. All Ra_{eq} values calculated in this study are lower than the recommended value.

H_{ex} , is defined as the external hazard index, and it is given by the following equation.

$$H_{\text{ex}} = \frac{C_{\text{Ra}}}{370} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810}, \quad (4)$$

C_{Ra} , C_{Th} and C_{K} have the same meaning as in Eq. (3). Calculated H_{ex} values are given in Table I. The value of H_{ex} must be less than unity [17–19]. All calculated values are lower than 1.

TABLE I

^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs activity concentrations, absorbed dose rates in air, AEDE, radium equivalent activity, external hazard index and excess lifetime cancer risk in soil samples collected from Zonguldak.

(Sampling code) Sampling areas	^{226}Ra [Bq kg $^{-1}$]	^{232}Th [Bq kg $^{-1}$]	^{40}K [Bq kg $^{-1}$]	^{137}Cs [Bq kg $^{-1}$]	ADRA (nGy h $^{-1}$)		AEDE [$\mu\text{Sv y}^{-1}$]	Ra $_{\text{eq}}$ [Bq kg $^{-1}$]	H_{ex}	ELCR terrestrial
					Terrestrial	Terrestrial and cosmic				
(1) Buruncuk	59.2 ± 3.2	53.2 ± 5.1	451 ± 31	12.0 ± 2.6	78	80	96	170	0.46	0.34 × 10 $^{-3}$
(2) İlkısu	18.4 ± 2.0	19.7 ± 3.2	205 ± 23	< 0.5	29	52	36	62	0.17	0.12 × 10 $^{-3}$
(3) Kokaksu	98.0 ± 4.3	51.4 ± 5.2	343 ± 30	20.9 ± 3.0	91	80	111	198	0.53	0.39 × 10 $^{-3}$
(4) Türkali	11.2 ± 2.0	13.8 ± 3.5	279 ± 25	< 0.5	25	37	31	52	0.14	0.11 × 10 $^{-3}$
(5) Soğanlıyörük	35.4 ± 2.8	43.8 ± 5.0	808 ± 43	13.8 ± 2.8	77	82	94	160	0.43	0.33 × 10 $^{-3}$
(6) Düzpelit	15.9 ± 2.2	13.1 ± 3.7	238 ± 27	47.1 ± 3.8	25	48	31	53	0.14	0.11 × 10 $^{-3}$
(7) İsabeyli	32.7 ± 2.7	38.8 ± 4.8	541 ± 36	26.8 ± 3.1	61	64	75	130	0.35	0.26 × 10 $^{-3}$
(8) Alpaslan	28.5 ± 2.6	30.9 ± 4.6	484 ± 30	13.5 ± 2.4	52	58	64	110	0.30	0.22 × 10 $^{-3}$
(9) Gaziler	16.1 ± 2.2	20.9 ± 4.3	310 ± 27	6.7 ± 2.1	33	46	40	70	0.19	0.14 × 10 $^{-3}$
(10) Geriş	24.2 ± 2.4	31.2 ± 4.4	399 ± 31	15.7 ± 2.4	47	49	57	100	0.27	0.20 × 10 $^{-3}$
(11) Kerimler	26.6 ± 2.5	22.2 ± 4.5	417 ± 32	10.8 ± 2.5	43	49	53	90	0.24	0.18 × 10 $^{-3}$
(12) Dağlıca	30.5 ± 2.5	31.8 ± 4.5	202 ± 27	9.9 ± 2.4	42	52	51	92	0.25	0.18 × 10 $^{-3}$
(13) Balçıklı	21.7 ± 2.4	31.7 ± 4.6	332 ± 30	7.8 ± 2.4	43	53	53	93	0.25	0.18 × 10 $^{-3}$
(14) Sofular	38.4 ± 2.7	43.0 ± 4.7	440 ± 30	14.7 ± 2.5	62	84	76	134	0.36	0.27 × 10 $^{-3}$
(15) Beycuma	29.2 ± 2.5	32.8 ± 4.3	561 ± 33	5.9 ± 2.2	57	54	70	119	0.32	0.24 × 10 $^{-3}$
(16) Gürbüzler	15.2 ± 1.9	12.9 ± 3.4	190 ± 22	8.0 ± 2.0	23	29	28	48	0.13	0.10 × 10 $^{-3}$
(17) Eğerci	15.1 ± 1.9	15.9 ± 3.5	289 ± 25	14.3 ± 2.1	29	37	35	60	0.16	0.12 × 10 $^{-3}$
(18) Fındıklı	10.5 ± 2.0	19.2 ± 3.8	341 ± 28	7.6 ± 2.1	31	38	38	64	0.17	0.13 × 10 $^{-3}$
(19) Hüseyinli	17.8 ± 1.9	19.0 ± 3.5	454 ± 29	< 0.5	39	50	47	80	0.22	0.17 × 10 $^{-3}$
(20) Işıklı	30.5 ± 2.4	36.4 ± 4.3	483 ± 33	9.5 ± 2.2	56	59	69	120	0.32	0.24 × 10 $^{-3}$
(21) Uludağ	31.1 ± 2.5	33.5 ± 4.2	454 ± 31	14.9 ± 2.4	54	70	66	114	0.31	0.23 × 10 $^{-3}$
(22) Ormanlı	26.7 ± 2.3	35.2 ± 3.9	468 ± 32	15.9 ± 2.3	53	62	65	113	0.31	0.23 × 10 $^{-3}$
Mean	28.8 ± 4	29.6 ± 3	395 ± 31	12.5 ± 2	48	56	58	101	0.27	0.20 × 10 $^{-3}$

Excess lifetime cancer risk (ELCR) was calculated using Eq. (5)

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF}, \quad (5)$$

where DL is duration of life (70 year) and RF is risk factor (Sv $^{-1}$), fatal cancer risk per Sievert. For stochastic effects, ICRP 60 [20] uses values of 0.05 for the public. ELRC values calculated from Eq. (5) are given in Table I.

When life expectancy was taken as 70 years, the mean lifetime outdoor gamma radiation was calculated as 4.06 mSv (terrestrial) and 4.81 mSv (terrestrial and cosmic). These values yielded a lifetime cancer risk of 0.20 × 10 $^{-3}$ (terrestrial) and 0.24 × 10 $^{-3}$ (terrestrial and cosmic). According to UNSCEAR [11], the average ELRC value in the world is 0.25 × 10 $^{-3}$ (terrestrial) and 0.29 × 10 $^{-3}$ (terrestrial and cosmic). The results obtained by the study are less than the world average values.

4. Conclusions

The activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs were determined in uncultivated surface soil samples collected from Zonguldak. From the measured values, the mean outdoor gamma ADRA, annual effective gamma dose equivalent (AEDE), radium equivalent activity (Ra $_{\text{eq}}$) and external hazard index (H_{ex}) were calculated. The results presented in this study are at normal levels, except for some locations.

In order to evaluate the health hazard, especially various types of cancer, it is important to determine the environmental radioactivity level. This research indicates that the average lifetime risks of cancer of the Zonguldak are below the worldwide range, even though there are some extreme values. This study contributes to the database of the radioactivity level and lifetime risks of

cancer in the Province of Zonguldak. The results may also be used as a reference data for monitoring possible radioactivity pollution in future.

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