NATURAL RADIOACTIVITY (226Ra, 232-Th AND 40K) AND ASSESSMENT OF RADIOLOGICAL HAZARDS IN THE KEŞTANBOL GRANITOID, TURKEY

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The surveys of natural gamma-emitting radionuclides in rocks and soils from the Ezine plutonic area were conducted during 2007. Direct dose measurement using a survey meter was carried out simultaneously. The present study, which is part of the survey, analysed the activity concentrations of 238U, 232Th and 40K in granitoid samples from all over the region by HPGe gamma spectrometry. The activity concentrations of 226Ra ranged from 94 to 637 Bq kg\(^{-1}\), those of 232Th ranged from 120 to 601 Bq kg\(^{-1}\) and those of 40K ranged from 1074 to 1527 Bq kg\(^{-1}\) in the analysed rock samples from different parts of the pluton. To evaluate the radiological hazard of the natural radioactivity in the samples, the absorbed dose rate (\(D\)), the annual effective dose rate, the radium equivalent activity (\(R_{\text{eq}}\)) and the external (\(H_{\text{ex}}\)) hazard index were calculated according to the UNSCEAR 2000 report. The thorium-to-uranium concentration ratios were also estimated.

INTRODUCTION

Knowledge of the distribution pattern of natural radionuclides is essential in maintaining some sense of control of prevailing radiation levels. Measurement of natural radioactivities in soil gives information on natural sources\(^{(1)}\). All rocks, soils and minerals contain naturally occurring radionuclides. Among the natural radionuclides, 238U, 232Th and 40K make the biggest contribution to the total background dose\(^{(2)}\). The external radiation exposure arises mainly from cosmic rays and from terrestrial radionuclides occurring at trace levels in all soils. While absorbed dose rate in air from cosmic radiation outdoors at sea level is \(~30\) nGy h\(^{-1}\)\(^{(3)}\), the specific levels due to terrestrial background radiation are related to the types of rock from which the soils originate. Therefore, the natural environmental radiation depends on geological and geographical conditions\(^{(4)}\). Higher radiation levels are associated with acidic igneous rocks, such as granite, and lower levels are associated with sedimentary rocks\(^{(3)}\).

In terms of natural radioactivity, granites exhibit an enhanced elemental concentration of uranium (U) and thorium (Th) compared with the very low abundance of these elements observed in the mantle and the oceanic crust of the Earth. Geologists provide an explanation of this behaviour in the course of partial melting and fractional crystallisation of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into the more silica-rich products\(^{(5)}\). For that reason, igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5 ppm of U and 15 ppm of Th), compared with the Earth's crust (average 1.8 ppm for U and 7.2 ppm for Th)\(^{(6)}\), the upper continental crust (average 2.7 ppm for U and 10.5 ppm for Th)\(^{(7)}\) and rocks of basaltic or ultramafic composition (0.1 ppm of U and 0.2 ppm of Th)\(^{(6, 8, 9)}\).

On the other hand, the uranium and thorium enhance in radioactive accessory minerals such as monazite, zircon, allanite,apatite, sphene, thorite etc., and potassium occurs in major minerals such as feldspar and micas\(^{(10)}\). All of these minerals and feldspar also enhance in silica-saturated acidic magmatic rocks, such as granite, rhyolite, syenite and pegmatite compared with intermediate, basic and ultra-basic rocks. Thus, higher concentrations of uranium and thorium are also attributed to the presence of these radioactive accessory minerals associated with granite rocks. Accordingly, the UNSCEAR report\(^{(11)}\) stated that the 226Ra, 232Th and 40K activities are 100–500, 40–350 and 1200–1800 Bq kg\(^{-1}\), respectively in the granites which are rich in uranium and thorium.

The Ezine region is located in northwestern Anatolia where young granitic and associated volcanic rocks are widespread (Figure 1). In this region, the Kestanbol granitoid which is surrounded by hypabyssal rocks to the east and southeast is enclosed by volcanic lava flows and associated volcanioclastics\(^{(12)}\). The study area is located in latitudes 39° 40′N to 39° 46′N and longitudes 26° 13′E to 26° 20′E, at altitudes between 100 and 450 m above sea...
level. Accordingly, the population in the region lives at altitudes below 0.5 km, and the annual effective doses (AEDs) from cosmic radiation are thus not in excess of the population-weighed average of 380 μSv (UNSCEAR, 2000).

The main objective of this study is to assess the risk of public exposure to the natural background radiation by determining the naturally occurring radionuclides in granitic rocks from Ezine plutonic area.

MATERIALS AND METHODS
Geological setting
In western Anatolia, the basement units of the Pontides and Anatolides are intruded by numerous granitoid plutons. Two magmatic phases are distinguished; 1-Eocene granitoids dated at ~40 Ma\(^{15}{}^{14}\) and 2-Oligo-miocene granitoids which were dated between 28 and 21 Ma\(^{15}\). The Kestanbol granitoid which was dated at 21–28 Ma\(^{16}{}^{17}\) is included in the second group. The Kestanbol granitoid, 12 km wide and 18 km long, which intruded into the Rhodope-Serbo-Macedonian Massif\(^{18}\) in Biga Peninsula covers approximately an area of 150 km\(^2\). Along the western border, the pluton is unconformably overlain by Upper Miocene-Pliocene continental coarse-grained clastics and conformably overlying shallow marine carbonates. However, the Kestanbol granitoid is bordered to the east and south by the Early Miocene rhyolite and dacite lavas and associated pyroclastics/epiclastic rocks. The Kestanbol granitoid

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Figure 1. (a) Location map of the study area and (b) geological map of the study area, modified from Örgün.\(^{10}\)
is generally made up of medium- to coarse-grained, holocrystalline equigranular quartzmonzonites and granites. Furthermore, in some places, the brownish-pink coloured orthoclase phenocrysts reaching up to 7 cm in size in an equigranular matrix consisting of plagioclase and quartz causes a porphyritic texture in quartzmonzonitic parts of the pluton. Mineral composition of the Kestanbol granitoid is K-Feldspar (orthoclase) + quartz + plagioclase + Ca-amphibole (hornblende) ± Clinopyroxene ± biotite and allanite ± zircon ± monazite ± ilmenite as accessory phases (The sign '+' indicate the minerals are always present while the sign '+' mean the mineral that may be present). Furthermore, the existence of uraninite and thorite have been reported(18). In quartzmonzonites, the allanite crystals up to 1 cm in size reach up to 8 modal %. On the basis of the isotopic and geochemical data, subduction enriched, mantle-derived magmas, which were contaminated by the upper crustal material, have been suggested by Karacık and Yılmaz(12) for the calc-alkaline and metaluminous Kestanbol granitoid.

Sample treatments and gamma-spectrometric measurements

Ten granitoid samples were collected from the Kestanbol granitoid pluton representing the Ezine area. The rock samples were first dried at room temperature, then oven dried at 105°C until they reached a constant weight. They were crushed to pass through a 2 mm sieve and were then filled in a 1-l Marinelli beaker. The containers were sealed and stored for at least 30 d to allow for secular equilibrium between 226Ra and its decay products, and to permit the Compton region to be stabilised before gamma-spectroscopy measurements were made. Spectrometric measurements of gamma-activity were carried out using a 184 cc p-type coaxial HPGe detector with a relative efficiency of 25 %, and a resolution of 1.85 keV at 1.332 MeV (with associated electronics procured from EG&G Ortec) connected to a PC-based 8 K multichannel. In all cases, the activity of 226Ra was evaluated from the 1764.49 keV peak of 214Bi, and 232Th activity was evaluated from the 2614.53 keV peak of 208Tl while that of 40K activity was determined from its characteristic gamma line of 1460.75 keV peak as shown in Figure 2. The minimum detectable activity for 226Ra, 232Th and 40K was 2, 2 and 4 Bq kg⁻¹, respectively, for a 10 000 s counting time and a sample weight of 1650 g.

RESULTS AND DISCUSSION

The activity concentrations for the 40K, U and Th series are shown in Table 1 and Figure 3, which were calculated from the gamma-ray spectra for these series. Relative uncertainties of the activity concentrations were usually lower than 5 % at the 68 % confidence level.

It is well known that the U-Th bearing accessory minerals (sphene, zircon, allanite, apatite, epidote, thorite, uranothorite; 0.1–4.5 %) are present in relatively high concentrations in the studied granitoid pluton(10, 18). Accordingly, the activity concentrations of 226Ra ranged from 94 to 637 Bq kg⁻¹, those of 232Th ranged from 120 to 601 Bq kg⁻¹ and those of 40K ranged from 1074 to 1527 Bq kg⁻¹ in the analysed rock samples from different parts of the pluton. From the 10 samples examined in this study, ‘G5 from Aladag’ appears to present the highest concentrations for all the naturally occurring radionuclides investigated, reaching levels of 637 Bq kg⁻¹.

Figure 2. Gamma-ray spectrum of Aladağ granitoid sample.
for $\text{Ra}_{226}$, 601 Bq kg$^{-1}$ for $\text{Th}_{232}$ and 1527 Bq kg$^{-1}$ for $\text{K}_{40}$. In addition, ‘G6, G9 and G2’ exhibited relatively highest concentrations according to the rest of the granitoid samples of Table 1.

Potassium is a major element in the composition of $\text{K}$-feldspars (orthoclase, microcline), as well as biotite and muscovite which are the main rock-forming minerals of granitic rocks (19). As expected, the $\text{K}_{40}$ activity concentrations of all examined samples were above the value of 1000 Bq kg$^{-1}$.

The absorbed gamma dose rates (nGy h$^{-1}$) in air at 1 m above the ground surface for the uniform distribution of radionuclides ($\text{U}_{238}$, $\text{Th}_{232}$ and $\text{K}_{40}$) were computed on the basis of guidelines provided by UNSCEAR report (3) with the following formula:

$$D = 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_{\text{K}}.$$  

To assess the radiological impact of granites, the absorbed dose rate $D$ (nGy h$^{-1}$), the AED, the radium equivalent activity $(A_{\text{Ra eq}})$ and the external $(H_{ex})$ hazard index were calculated and also listed in Table 1. Corresponding data obtained from UNSCEAR (1993) for the granites are included for comparison.

To estimate the AED (mSv y$^{-1}$), the conversion coefficient from absorbed dose rate in air to effective dose (0.7 Sv Gy$^{-1}$) and outdoor occupancy factor (0.2) proposed by UNSCEAR report (3) were used. The effective dose rate in units of mSv y$^{-1}$ was calculated by the following formula:

$$AED = D(nGy h^{-1}) \times 8760 h \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6}.$$  

The gamma-ray radiation hazards due to the specified radionuclides were assessed by radium equivalent activity and external radiation hazard. Radium equivalent activity $(A_{\text{Ra eq}})$ and external radiation hazards $(H_{ex})$ were calculated according to Equations (3) and (4), respectively.

$$A_{\text{Ra eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}},$$  

$$H_{ex} = \left(\frac{A_{\text{Ra}}}{370}\right) + \left(\frac{A_{\text{Th}}}{259}\right) + \left(\frac{A_{\text{K}}}{4180}\right),$$

where $A_{\text{Ra}}, A_{\text{Th}}$ and $A_{\text{K}}$ are the average activity concentrations of $\text{U}_{238}, \text{Th}_{232}$ and $\text{K}_{40}$ in Bq kg$^{-1}$, respectively.

The radiological characteristics of granitoid samples are shown in Figure 4 and listed in Table 1. The absorbed dose rates ($D$) of the granitic rocks in air at 1 m above ground level were calculated from Equation (1). Incidentally, it may be useful to remember that the presence of high mineral contents in the rock composition decreases the gamma dose rates due to the self-absorption of gamma rays. As reported by Saito and Jacop (1995) (20), with particular attention to iron abundance in the soil composition, the maximum difference in the absorbed dose rate in air is ~25 % for photon energies below 150 keV while the difference is less than 10 % in the
energy region above 150 keV. Nevertheless, the external exposure to the population is mainly by gammarays emitted from $^{226}\text{Ra}$ daughters, particularly $^{214}\text{Pb}$ and $^{214}\text{Bi}$. The contribution of these gamma rays to external dose is about 98%, considering other gamma-emitting radionuclides, in the $^{238}\text{U}$ decay series. In the case of the $^{232}\text{Th}$ series, $^{208}\text{Tl}$ and $^{228}\text{Ac}$ contribute 90% of the total external gamma dose rate, and $^{212}\text{Pb}$ and $^{212}\text{Bi}$ contribute 9% (20). Taking into consideration that the gamma energies are above 150 keV and most of the gamma energies are also higher than 1 MeV, one can, therefore, roughly estimate the dose rate 1 m above the ground level by using the UNSCEAR 2000(3) conversion coefficient like in this case. Accordingly, the calculated absorbed dose rates in air appeared in the range of 170–721 nGy h$^{-1}$. In addition, field in situ gamma radiation exposure rates using a survey meter are compared with the absorbed dose rate values from radiometric data of rocks and reported in the last column of Table 1 to point out any correlations among them. The results are thus in good agreement with the radiometric data.

Although the outdoor occupancy factor is taken into account to be 0.2 in order to calculate the AED from outdoors, the estimated 20% of time spent outdoors is considered likely to be high for the rocky area. However, there is no basis of changing this value at present due to the lack of data that might apply to the surveyed area of concern to this paper. Thus, this factor is assumed as 0.2, and the AEDs were found to range from 208 to 884 μSv. It follows from Table 1 that the Ra$_{eq}$ activities, calculated from Equation (3), are within 368–1614 Bq kg$^{-1}$ range while the values of the hazard index ($H_{ex}$) based on the criterion formula are higher than unity.

Of the values reported in Table 1 concerning the radiological characteristics of granitoid samples, the results are thus rather high in the worldwide average values for regular soil given in the recent UNSCEAR report(15). Nevertheless, these values are within the range for the granite rocks specified by UNSCEAR 1993 report(11).

Additional study of the Th/U and Th/K concentration ratios presents the following conclusions:

1. The eTh/eU ratios in the granitoid samples varied from 1.98 to 5.12 with a mean value of 2.99. Our measurements for $^{238}\text{U}$ and $^{232}\text{Th}$ are inferred from gamma spectrometric analyses of $^{214}\text{Bi}$ and $^{208}\text{Tl}$ gamma lines, respectively, assuming radioactive equilibrium. In this regard, it is preferable that the results are expressed in terms of equivalent uranium and equivalent thorium concentrations, respectively. For this reason, determinations of $^{238}\text{U}$ and $^{232}\text{Th}$, by gamma spectrometry are denoted by a prefixed ‘e’ (e.g. eU, eTh and the Th/U ratio becomes the eTh/eU)(21). Incidentally, it may be useful to remember that the concentrations of 1 ppm for uranium and thorium are equivalent to 12.3 Bq kg$^{-1}$ of $^{238}\text{U}$ and 4.1 Bq kg$^{-1}$ of $^{232}\text{Th}$, respectively, as well as their decay products under secular equilibrium conditions of these natural series. Accordingly, these values are used by converting the unit of activity concentration to the units of concentration (ppm eU and ppm eTh).

Consequently, the eTh/eU ratios in the granitoid samples are close to 3.5 (Clark’s value), which denotes a slight enrichment of uranium in the area under investigation. A correlation exists between $^{232}\text{Th}$ and $^{238}\text{U}$ in granitoid samples ($R^2 = 0.8979$), which shows that the sample population is dominated by Th and U bearing minerals (Figure 5). This clearly indicates the presence of significant amounts of monazite and zircon minerals in granitoid samples.

2. As seen from Figure 6, the mean eTh (ppm)/K (%) concentration ratio is 4.02 and this value indicates a correlation between Th and K according to Clark (Th (ppm)/K(%) = 4.5 in acid igneous rocks)(22).

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Figure 4. Radiological characteristics of granitoid samples.

Figure 5. The eTh (ppm)/eU (ppm) ratios in the granitoid samples.
REFERENCES


