ICCESEN 2017



Determination of natural radioactivity levels in soil and travertine of the region of Tokat and Sivas, Turkey

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Received: 15 November 2017 / Accepted: 28 February 2018 / Published online: 22 March 2018 \odot Saudi Society for Geosciences 2018

Abstract

In this study, the environmental radioactivity measurements for Tokat and Sivas provinces in the northeast of Turkey were performed. Using gamma ray spectrometry, the activity concentrations of natural radionuclides in soil and travertine samples (232 Th, 226 Ra, and 40 K) were determined. The annual effective dose equivalent, the absorbed doses rate in air, the radium equivalent, and the external hazard index were obtained from these activities. The activity concentrations vary from 9.09 to 17.04 Bq kg⁻¹ for 232 Th, from 36.53 to 76.95 Bq kg⁻¹ for 226 Ra, and from 216.56 to 576.59 Bq kg⁻¹ for 40 K in soil samples. The activity concentrations in travertines vary from 15.99 to 21.01 Bq kg⁻¹ for 232 Th, from 19.89 to 67.71 Bq kg⁻¹ for 226 Ra, and from 179.89 to 314.43 Bq kg⁻¹ for 40 K. The average dose rate in air for soil and travertine samples was 43.41 and 41.05 nGy h⁻¹ respectively. The obtained results are presented and compared with other studies, and the results of this study are lower than the international recommended value (55 nGy h⁻¹) given by UNSCEAR (2000). The results show that the region has a background radiation level within the natural limits.

Keywords Natural radioactivity · Soil and travertine · Gamma spectrometer

Introduction

Natural radioactivity for many years has been one of the popular research areas for scientists since it has harmful effects on human beings and the environment. Naturally occurred radioactive materials (²³⁵U, ²³²Th, and ⁴⁰K isotopes and their decay products) are the main source of radiation where we live and exposed everyone to ionizing radiation (UNSCEAR 2000; Kucukomeroglu et al. 2009). The contribution of ²³²Th, ²²⁶Ra, and ⁴⁰K radionuclides to the level of background radiation is about 14, 55.8, and 13.8% respectively (Bozkurt et al. 2007; Bavarnegin et al. 2013). It is crucial to measure in different regions the radiation concentrations in soil, water, and air to determine the radiation exposure. For this reason, the literature is rich in regional radiation measurement issue (Cetin 2016; Canbazoğlu et al. 2013; Cetin et al. 2016; Degerlier et al. 2008; Karahan and Bayulken 2000; Saad 2017; Gören et al. 2017; Kuluöztürk and Doğru 2015; Turhan and Gündüz 2008; Uyanik et al. 2015; Yigitoglu et al. 2010). This study was done to determine the natural

This article is part of the Topical Collection on *Geo-Resources-Earth-Environmental Sciences*

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radioactivity concentrations in soil and travertine samples in Tokat and Sivas provinces. In addition, absorbed doses (D) in air, radium equivalent (Ra_{eq}), external hazard index (H_{ex}), and the annual effective dose equivalent (AEDE) were obtained. The obtained results were compared with national and international mean values. This study is going to contribute to the establishment of data on natural radiation background levels for the research region and determines the dose rates to the resident in the region.

Tokat and Sivas provinces are situated in the northeast of Turkey (Fig. 1). Tokat lies between the coordinates $39^{\circ} 51'-40^{\circ} 55'$ N and $35^{\circ} 27'-37^{\circ} 39'$ E, and Sivas lies between the latitudes of $38^{\circ} 42'-40^{\circ} 16'$ N and the longitudes of $35^{\circ} 50'-38^{\circ} 14'$ E, and they are located in K1211rmak and Yeşi11rmak River Basins. Until now, there have been few studies reporting the natural radioactivity concentrations in Tokat and Sivas (Yigitoglu et al. 2010).

Locations of Sivas center-1 (S1) and Sivas center-2 (S2) sampling area are between 39° 52′ 48.8″ N latitude and 37° 12′ 10.0″ E longitude and 39° 52′ 31.8″ N latitude and 37° 12′ 16.3″ E longitude. Sivas–Yıldızeli (S3) sampling area is situated in 39° 46′ 25.7″ N latitude and 36° 44′ 24.4″ E longitude.

And, Tokat sampling area (T1) is situated in 40° 19' 13.3" N latitude and 36° 31' 22.7" E longitude.

Experimental procedure

Geology of the area

The research area lay within the high platitudes of Central Anatolian Thrust Zone and rises to the east. It ends with a mountainous and steep section in the north, east, and southeast. The altitude of the region is over 1400 m. The research stations are located in the Yıldız River Basin near Yildizeli town mostly composed of travertine, marble, limestone and volcanic, plutonic and ophiolitic rocks. The basement of this rock formations in this basin consist of Early Upper Cretaceous–Paleocene Akdag Madeni Lithodeme and Upper Cretaceous–Paleocene Tekelidag Melange. The Cretaceous-Paleocene Boztepe Formation overlay irregularly above the mentioned tectonic units (Yılmaz 1982; Yılmaz et al. 1997). In the research area, hydrothermal fluids appeared using



Fig. 1 The location map of Tokat and Sivas City, Turkey, and the distribution of soil and travertine samples

fracture systems in the Late Miocene form the secondary minerals (Yalcin and Bozkaya 2003).

Sample collection and preparation

In this study, four different types of soil and travertine marble were collected from Tokat and Sivas provinces. The samples, prepared in 1-kg pieces, were then dried in a temperature-controlled furnace at 100 °C for 20 h to the complete removal of the moisture of the samples. After moisture removal, each sample was placed in a cylindrical plastic container, which was completely sealed, and stored for 4 weeks until the daughter and parent decay rates of the radionuclides in the sample are in secular equilibrium (Yang et al. 2005; El-Taher et al. 2013).

Radiometric analysis

Activity concentrations of the ²²⁶Ra, ²³²Th, and ⁴⁰K in the samples were analyzed by a Canberra type $3'' \times 3''$ NaI (Tl) gamma spectrometer at the Gamma Spectrometer Laboratory in Physics Department of Suleyman Demirel University. The counting time for each samples and background was 72,000 s to obtain the gamma ray spectrum. The energy calibration of the detector was performed using point sources (⁶⁰Co, ¹³⁷Cs). Background measurements were taken and subtracted in order to get net counts for the sample.

The detected gamma rays with 1760, 2610, and 1461 keV energies belong to 226 Ra, 232 Th, and 40 K radionuclides in the natural γ -ray spectrum respectively are recorded and used for the measurements. The activity concentrations of these natural radionuclides were calculated using the following Eq. (1) (Armani and Tahtat 2001):

$$A(\operatorname{Bq} \operatorname{kg}^{-1}) = \frac{N}{\varepsilon \times P \times M \times t}$$
(1)

where N is peak area of the gammas decay from the excited states of radionuclides, ε is the detector efficiency, P is transition probability of γ -decay, M is the mass (kg) of the measured sample, and t is the counting time.

The minimum detectable activity (MDA) of the gamma ray measurement system was calculated as follows Eq. (2) (Currie 1968);

$$MDA = \frac{F_c \sigma}{\varepsilon I_{\gamma} t M}$$
(2)

where MDA is in becquerel per kilogram, $F_{\rm C}$ is the statistical coverage factor equal to 1.64 (confidence level 95%), σ is the standard deviation of the background in the region of interest and equals square root of the number of counts for the background spectrum, ε is the absolute efficiency of the detector, I_{γ} is the absolute emission probability of the gamma decay, *t* is the counting live time in seconds, and *M* is the dried sample mass. The mean values of MDA calculated for 226 Ra, 232 Th, and 40 K were estimated to be 3.8, 2.8, 17.2 Bq kg⁻¹, respectively.

Results and discussion

The activity concentrations of ²³²Th, ²²⁶Ra, and ⁴⁰K

The activity concentrations in soil samples and travertine samples were for the 232 Th, 226 Ra, and 40 K radionuclides. The activity concentrations in travertine ranged from 15.99 to 21.01 Bq kg⁻¹ for 232 Th, from 19.89 to 67.7 Bq kg⁻¹ for 226 Ra, and from 179.89 to 314.43 Bq kg⁻¹ for 40 K. The activities of 232 Th, 226 Ra, and 40 K in soil varied from 9.09 to 17.04, from 36.53 to 76.95, and from 216.56 to 576.59 Bq kg⁻¹ respectively. The measured activity values are seen in Figs. 2 and 3, which that this figure the concentration values of 40 K are higher than 232 Th and 226 Ra both soil and travertines. The lowest values were generally obtained in Tokat/Zile. The world mean concentrations of 232 Th, 226 Ra, and 40 K in soil samples are 32, 45, and 420 Bq kg⁻¹ respectively (UNSCEAR 2000).

In Figs. 2 and 3, ²³²Th, ²²⁶Ra, and ⁴⁰K activities of travertine and soil samples for Tokat and Sivas regions are demonstrated. The activities, especially belong to travertines, do not show a uniform distribution. This difference may depend on the rocky geological structure of the region: this area mostly composed of travertine, marble, limestone and volcanic, plutonic and ophiolitic rocks, and overlay irregularly above the mentioned tectonic units (Yılmaz 1982; Yılmaz et al. 1997).

Absorbed dose rate in air

Terrestrial radionuclides are the one of the main sources of gamma rays. The exposure rate can be defined as follows (UNSCEAR 2000; Veiga et al. 2006).

$$D(nGy h^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K}$$
(3)

where A_{Ra} , A_{Th} , and A_{K} represent the activities for these isotopes. The coefficients 0.462, 0.604, and 0.0417 nGy h⁻¹ per Bq kg⁻¹ in Eq. (2) are the conversion factors of ²²⁶Ra, ²³²Th, and ⁴⁰K respectively. The absorbed dose rates are shown in Tables 1 and 2. From Tables 1 and 2, the absorbed dose rates for soil samples lie from 39 to 56 nGy h⁻¹ and a mean value of 43.41 nGy h⁻¹, whereas for travertine samples, it is 41.05 nGy h⁻¹. Our data for soil and travertine are in agreement with the UNSCEAR (UNSCEAR 2000) world mean 55 nGy h⁻¹.

Fig. 2 Variation of ²³²Th, ²²⁶Ra, and ⁴⁰K concentrations in travertine samples



Radium equivalent activity and external hazard index

The external hazard index (H_{ex}) and radium equivalent activity (Ra_{eq}) terms are used for the gamma radiation hazard caused by gamma (γ) rays. Ra_{eq} hazard index is defined by Ref. (Beretka and Mathew 1985).

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.77 A_K.$$
(4)

The values of Ra_{eq} are given in Tables 1 and 2. Ra_{eq} varied from 234.80 to 340.31 Bq kg⁻¹, and the mean value is obtained as 313.94 for soil and 268.09 Bq kg⁻¹ for travertine. The measured Ra_{eq} values for Tokat and Sivas provinces lie low than the worldwide mean value 412.6 Bq kg⁻¹ (UNSCEAR 2000). As can clearly be seen from Table 1, the mean value obtained from this work lies low in value when it is compared with the values of the other areas of Turkey and countries given in Table 1, except for Firtuna Valley (Kurnaz et al. 2007), Egypt (UNSCEAR 2000), and Bangladesh (UNSCEAR 2000).

The H_{ex} hazard index is calculated as in Ref. (Krieger 1981) for the samples investigated as follows:

$$H_{\rm ex} = A_{\rm Ra}/370 + A_{\rm Th}/259 + A_{\rm K}/4810 \le 1.$$
(5)

This criterion corresponds to a maximum radium equivalent activity of 370 Bq kg⁻¹ for the materials. This index is used to estimate the level of γ -radiation hazard associated with the natural radionuclides in specific building materials. The values of H_{ex} calculated for soil and travertine samples are presented in column 7 of Tables 1 and 2. They are varied from 0.176 to 0.436. Their mean is 0.241, and less than the critical value of unity.

The obtained average H_{ex} value for the soil is lower than that of the worldwide mean value (0.286) and is also lower than the reported values from other parts of Turkey, i.e., 0.99, southeast part of Eskischir (Orgun et al. 2005); 1.129, Çanakkale (Merdanoglu and Altınsoy 2006); and 0.45, Fırtına Valley (Kurnaz et al. 2007). But, our results are higher according to the values in other areas of Turkey given in Refs.



Fig. 3 Variation of 232 Th, 226 Ra, and 40 K concentrations in soil samples

 Table 1
 Concentrations of the average natural radionuclides, as well as the absorbed doses in air, Ra equivalent, external hazard index, annual effective dose equivalent in soil samples from different parts of the world, compared with those of the present study

Country	Mean activity concentrations in soil (Bq kg^{-1})			Absorbed dose rate in air (nGy h^{-1})	Ra _{eq}	H _{ex}	AEDE (µSv/year)
	²²⁶ Ra (²³⁸ U)	²³² Th	⁴⁰ K				
Belgium (UNSCEAR 2000)	26	27	380	43	357.21	0.175	52.735
Bulgaria (UNSCEAR 2000)	32	38	490	59	463.64	0.309	72.357
China (UNSCEAR 2000)	32	41	440	62	429.43	0.288	76.036
Algeria (UNSCEAR 2000)	50	25	370	53	370.65	0.307	64.999
India (UNSCEAR 2000)	29	64	400	56	428.52	0.083	68.678
Iran (UNSCEAR 2000)	28	22	640	71	552.26	0.092	87.074
Bangladesh (UNSCEAR 2000)	34	_	350	35	303.5	0.631	42.924
Malaysia (UNSCEAR 2000)	67	82	310	93	422.96	0.188	114.055
Egypt (UNSCEAR 2000)	17	18	320	32	289.14	0.298	39.244
Romania (UNSCEAR 2000)	45	30	400	70	395.9	0.079	85.848
China (Yang et al. 2005)	112	71.5	672	112	731.685	0.232	137.356
Greece (Marchetti et al. 2004)	25	21	360	56	332.23	0.400	68.678
West coast of India (Karunakara et al. 2005)	30.6	38.2	394	54	388.606	0.242	66.225
USA (Myrick et al. 1983)	40	35	370	47	374.95	0.102	57.640
Turkey, Çanakkale (Merdanoglu and Altınsoy 2006)	115	192	1207	219	1318.95	1.129	268.581
Turkey, Fırtına Valley (Kurnaz et al. 2007)	50	42	643	77.4	605.17	0.43	88.70
Turkey, Manisa (Erees et al. 2006)	28.5	27	340	43.65	328.91	0.252	53.43
Turkey, Karabük (Baldık et al. 2011)	21	23.5	363.5	39.05	334.5	0.223	47.89
Turkey, Kırklareli (Taskin et al. 2009)	28	40	667	64.9	598.79	0.368	79.59
Turkey, Karaman (Agar et al. 2014)	28.5	21.6	342.4	40.49	323.11	0.232	49.65
Turkey, Batman (Damla et al. 2010)	42	35	524	62.39	495.53	0.357	76.51
Turkey, Samsun (Tufan and Bostancı 2012)	19	22	521	43.79	451.63	0.245	53.70
Turkey, Sinop, Gerze (Kurnaz 2016)	63.87	72.66	609	98.79	636.70	0.579	121.16
Turkey, Zonguldak (Aytekin et al. 2017)	40.7	38.7	470	61.78	457.94	0.357	75.76
Turkey (Tokat-Sivas) present work	45.7	14.3	321.9	43.1	313.94	0.241	52.091
World average (UNSCEAR 2000)	32	40	420	56	412.6	0.286	70

 Table 2
 Concentrations of the average natural radionuclides in travertine and absorbed doses in air from different parts of the world, compared with those of the present study

Country/region	Mean activity concentrations in travertine (Bq kg ⁻¹)			Absorbed dose rate in air	Ra _{eq}	$H_{\rm ex}$	AEDE (µSv/year)
	²²⁶ Ra (²³⁸ U)	²³² Th	⁴⁰ K	(nGy h ⁻¹)			
Italy, (Trevisi et al. 2005)	5.4	13.5	65.6	13.38	75.22	0.067	16.41
(Marchetti et al. 2004)	0.75	0.25	5.4	0.72	5.26	0.003	0.88
(Bruzzi et al. 1997)	-	< 0.2	5.9	0.37	4.83	0.000	0.45
Italy, Vitorchiano	0.8	< MDA	4.4	0.55	4.5	0.003	0.67
Guidonia (Roma)	0.575	0.52	5.75	0.82	5.75	0.004	1.01
Pietrasanta (Lucca) (Marchetti et al. 2004)	5.2	< MDA	10.4	2.84	13.2	0.014	3.48
Iran (El-Taher et al. 2013)	< MDA	< MDA	4.44	0.185	3.39	_	0.23
Iran–Ramsar	138.1	22.8	421	214.03	495	0.46	262.45
(Ghiassi-Nejad et al. 2002)	43.77	16.5	260.69	41.05	268.09	0.182	50.343
Turkey (Tokat–Sivas) [present study]							

(Erees et al. 2006; Baldık et al. 2011; Agar et al. 2014; Tufan and Bostancı 2012). For travertine, the obtained $H_{\rm ex}$ value in this study is higher than that of the results of the other studies given in Table 2, except for Iran–Ramsar (Ghiassi-Nejad et al. 2002).

Annual effective dose equivalent

The AEDE is the tissue weighted sum of equivalent doses in all organs of human body caused by radiation in a year. AEDE is defined with the following expression

$$AEDE(mSv/year) = D(nGy/year) \times 8760(h/year)$$
$$\times 0.2 \times 0.7(Sv Gy^{-1}) \times 10^{-3} \qquad (6)$$

where 0.2 is the outdoor occupancy factor, 0.7 Sv Gy⁻¹ is the conversion coefficient from absorbed dose in air. The AEDE results calculated in this study were given in the column 8 of Tables 1 and 2. The AEDE values range from 47.83 to 68.68 μ Sv year⁻¹. The mean value is 52.09 mSv year⁻¹ for AEDE and this value is lower according to the world mean (70 mSv year⁻¹) (UNSCEAR 2000). In the present work, the obtained values lie low from the world average value and other values given in Tables 1 and 2, except for Egypt and Bangladesh.

Conclusion

In this study, the environmental radioactivity levels in soil and travertine samples are determined for the Tokat and Sivas regions located in the northeast region of Turkey. The obtained results indicate that the area, which was investigated in this work, is within the natural limits. The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K isotopes in the soil in Tokat and Sivas regions lie in the safe limits according to the reported limits and the mean values of the UNSCEAR (UNSCEAR 2000). In addition, the AEDE is lower than the world mean. The measurement results obtained in these regions show no significant departure from the other parts of the country and world means. In this respect, it may be concluded that the natural radiation from soil and travertine in the Tokat and Sivas provinces does not pose a radiological hazard and health risk. So far, there were only a few study in the region (Yigitoglu et al. 2010). This study will enable the expansion of the environmental radioactivity worldwide database and the assessment of dose rates to which the people in this region are exposed.

Funding information This work is supported by Gaziosmanpasa University Scientific Research Projects Department (BAP) under project no. 24/2013.

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