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ORIGINAL ARTICLE

The in situ natural radionuclide (^{238}U,^{232}Th and^{40}K) concentrations in Gümüşhane granitoids: implications for radiological hazard levels of Gümüşhane city, northeast Turkey

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Abstract

We have carried out 48 in situ measurements on radioactivities of ^{238}U, ^{232}Th and ^{40}K isotopes at the Gümüşhane granitoid plutons employing gamma-ray spectrometer with a NaI(Tl) scintillation detector. The radionuclide activity concentrations of ^{40}K, ^{238}U and ^{232}Th ranged from 62.6 to 1680.8 Bq kg^{-1}, 2.5 to 119.9 Bq kg^{-1} and 3.3 to 92.4 Bq kg^{-1}, respectively. The mean concentration of natural radionuclides (^{40}K, ^{238}U and ^{232}Th) was found to be 638.5 ± 421.6 Bq kg^{-1}, 40.8 ± 27.4 Bq kg^{-1} and 33.5 ± 25.5 Bq kg^{-1}, respectively. The mean values of the radium equivalent and the external hazard index were 137.9 ± 80.6 Bq kg^{-1} and 0.37 ± 0.22, respectively. The value of annual effective dose equivalent value (81.8 µSv year^{-1}) is lower on average than in the world average of 460 µSv year^{-1}. The external hazard index acquired in this study did not exceed the international safety standard levels, which means that Gümüşhane pluton does not produce any radiation hazards to the dwellers.

Keywords Radium equivalent activity · Natural radioactivity · Radiological hazard · Granitic pluton · Gümüşhane

Introduction

The radionuclides of the ^{40}K, ^{238}U and ^{232}Th are the fundamental sources of the outer gamma radiation. This radiation contributes significantly to the background doses received by human beings. Brai et al. (2002) and Arafa (2004) presented that these radionuclides concentrations are usually high in granite rocks within the continental crust. Magmatic rocks of granitic composition are evidence for greatly enriched in U–Th isotopes related to the continental Earth's crust and rocks of basaltic or ultramafic composition (Mason and Moore 1982; Rudnick and Gao 2003; Tzortzis and Tsertos 2004).

U and Th are present in relatively high concentrations in accessory minerals concentrated in granitic rocks such as allanite or orthite, apatite, monazite, sphene and zircon (Bowie and Plant 1988; Valkovic 2000). Potassium, which is a major element in the composition of K-feldspars such as biotite, microcline, orthoclase and muscovite, is available in most of the granitic rocks. The K_2O content of K-feldspars changes between 11 and 15% while in biotite and muscovite it ranges from 10 to 11% and varies between 8 and 10%, respectively.

The method of gamma-ray spectrometry has been widely utilized in natural radiation level of soils and sediments (Myrick et al. 1983; Karahan and Bayülken 2000; Sroor et al. 2001; Chiozzi et al. 2002; Karakelle et al. 2002; Merdanoğlu and Altınsoy 2006; Kurnaz et al. 2007; Saleh 2012), natural radionuclide contents of the granites for building materials (Chen and Lin 1996; Zikovsky and Kennedy 1992; Pavlidou et al. 2006), radionuclide concentrations of volcanic rocks and granitic plutons (Örgün et al. 2005; Aydın et al. 2006; El-Arabi 2007; Maden and Akaryalı 2015a), mineral exploration (El-Sadek 2009; Maden and Akaryalı 2015b), and environmental radiation monitoring (Sanderson et al. 1995; Ford et al. 2000; Lahti et al. 2001).

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Örgün et al. (2005) determined the radioactivity level of the Sivrihisar and Kaymaz granitoid plutons investigating the 14 granitic samples. Yang et al. (2005) studied the natural radioactivity of soils to evaluate the radiation hazard at the Xiazhuang granite computing the values of the radium equivalent activity, the absorbed dose rate, the annual effective dose rate, and the external hazard index. Pavlidou et al. (2006) carried out the natural radioactivity and dose exposure based on the activity concentrations of radionuclides for granites used as building products in construction industry to evaluate the relationship between radioactive minerals and the natural radioactivity. Kurnaz et al. (2007) have estimated the levels of natural radioactivity in sediment and soil samples in Fırtına Valley. Maden and Akaryalı (2015b) used the gamma-ray spectrometry method to characterize the zone of potassic alteration developed in epithermal gold deposit in the Gümüşhane. Also, Maden and Akaryalı (2015a) used gamma-ray spectrometry data of the Gümüşhane granitoid to delineate geodynamic setting of the Eastern Pontides.

Gümüşhane has a total surface area of 6575 km^2. The population of Gümüşhane has increased from 116,008 in 2000 to almost 162,748 in 2018. The population density equals to 25 inhabitants per square kilometer. Life expectancy at birth for the Gümüşhane province is 79.8 years (TURKSTAT 2018). The main goal of this study is to evaluate the radiological hazards indices at the Gümüşhane province using the concentrations of ^{40}K, ^{238}U and ^{232}Th isotopes estimated by the γ-ray spectrometer with a NaI(Tl) scintillation detector. It is also carried out as an assessment of the radium equivalent activity (Ra_{eq}), the absorbed dose rate (Dabs), the annual effective dose equivalent (AEDE) and the external hazard index (Hex), based on concentrations of Gümüşhane granitoids.

Geological setting

The Eastern Pontides magmatic Belt, which geographically occupies the northeastern portion of Turkey, is situated in the Alpine–Himalayan system. This belt can be subdivided into three different geological subregions as Axial, Southern and Northern from south to north based on tectonic characteristics, facies changes and distribution of the principal lithological units (Bektaş et al. 1995; Eyüboğlu et al. 2006; Fig. 1a).

The study region is situated in the southern side of the Eastern Pontides Volcanic Belt (Fig. 1b). The oldest rock groups are Late Carboniferous Kurtoğlu metamorphic complex including gneiss, schist, phyllite, amphibolite (Topuz et al. 2007) and Gümüşhane–Köse granitoids (Topuz et al. 2011; Dokuz 2011). The Gümüşhane granitoid cuts the ≥ 323 Ma aged Kurtoğlu metamorphites with amphibolite facies along the southern boundary and is overlain unconformably by the Liassic aged volcanoclastics. Granite, granodiorite and felsic rocks (microgranite, spherolytic dacite and rhyolite) are the main rock types forming granitoid. The granite and granodiorite are exposed in the middle and northern parts of the pluton that contain greater amounts of hornblende, biotite, plagioclase, K-feldspar and lesser amounts of magnetite, apatite and zircon. On the other hand, the felsic rocks consisting of quartz and feldspar are exposed in the south and southeastern parts of the basement. The boundary between granite/granodiorite and felsic rocks is gradual transition. The granites containing plenty of discontinuities have completely altered to display the arenas in places. In addition, apliteic dykes are seen in granitic units. The ^{40}Ar–^{39}Ar biotite/hornblende and U–Pb zircon ages of the Gümüşhane granitoids range from 320 ± 4 Ma to 324 ± 6 Ma (Topuz et al. 2011).

The igneous rocks of Gümüşhane pluton are richer in radioactive accessory minerals such as monazite, apatite and zircon, which could be the source of high Th abundances. The radioelemental concentrations of granite are in the order of 1–2 magnitudes, which is greater than those of basaltic–ultrabasic rocks. Granitic rocks give highest U, Th, and K contents with a mean value of 5.9 ± 2.7 ppm, 22.1 ± 5.6 ppm, and 4.4 ± 0.7%, respectively (Maden and Akaryalı 2015a). Data from Topuz et al. (2010) obtained that samples from the Gümüşhane pluton have U in the range of 1.8–9.2 ppm (mean value of 3.8 ppm), Th 6.9–36.8 ppm (mean value of 14.5 ppm) and K_2O 1.8–5.2% (mean value of 3.5%).

Material and methods

Gamma-ray spectrometric analysis

In situ gamma-ray spectrometry is a powerful technique that can identify and potentially quantify radionuclides directly at the measurement site. We have utilized three windows, 512-channel Gamma Surveyor instrument made by GF Instruments (Czech Republic) to measure the concentrations of ^{40}K, ^{238}U and ^{232}Th radionuclides. The detector in the instrument is a bismuth germanate oxide with a volume of 21.2 in^3. To count photons that have the energies of interest, the spectrometers are designed with four windows. Three important gamma-ray energies from higher to lower are 2.62 MeV for thorium (^{208}Tl), 1.76 MeV (^{214}Bi) for uranium and 1.46 MeV (^{40}K) for potassium. The ^{40}K activity concentration measurements were determined directly from the 1.460 MeV emissions. The ^{238}U and ^{232}Th activities for measurements were determined from daughter product ^{214}Bi and ^{208}Tl, respectively (Ray et al. 2008; IAEA 2003).

The NaI(Tl) scintillation detector, housing the sensor, was mounted directly on the fresh outcrops with flat areas. During the measurements, it is kept away from the environmental

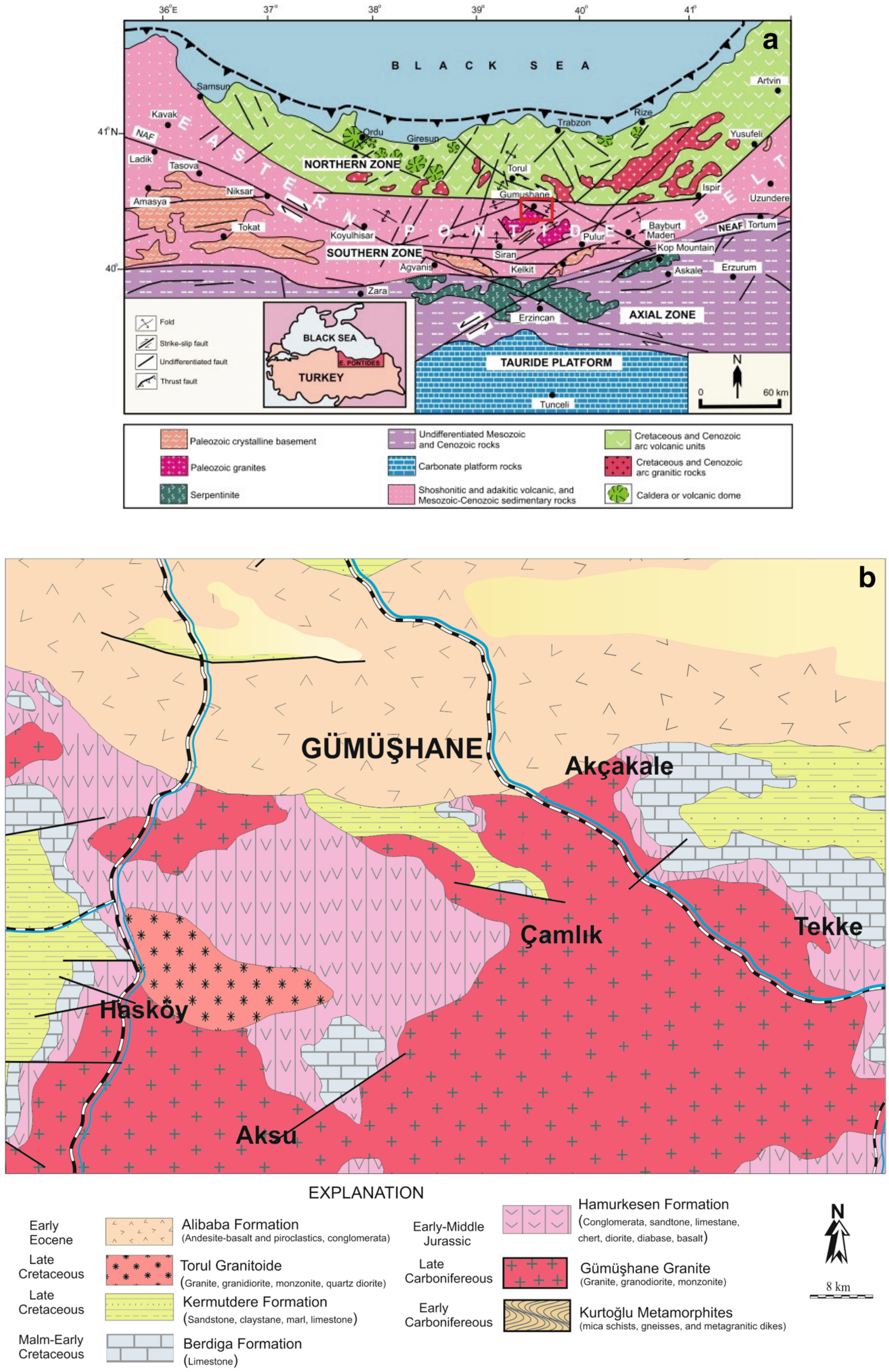

Fig. 1 **a** Tectonic map showing the main lithological units and zones of the eastern Pontides orogenic belt (Eyüboğlu et al. 2006). The red rectangle shows the study region. **b** Geological map of the study area (Güven, 1993; Maden and Akaryalı, 2015a)

effects causing the erroneous reading (Loevborg et al. 1971; Ray et al. 2008). The duration of measurement is adjusted with requested measurement precision and the estimated radiation strength. An optimum measurement period for certain situations is chosen experimentally to provide precise results. If a 2 × 2″ NaI(Tl) detector is utilized and the measuring period is fixed to 3 min, the result will have ensure order of 1 ppm for U and Th and of 0.1% for K concentrations. Shorter period of measurement provokes minimized stability of attained results (Maden and Akaryalı 2015b;

IAEA 2003). In the present study, in situ radioelemental (^{238}U, ^{232}Th and ^{40}K) measurements have been carried out at 48 sites (Fig. 2) through Gümüşhane city where the population density is high.

Results and discussion

The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K series in the Gümüşhane Province

The gathered ^{40}K, ^{238}U, ^{232}Th concentration values whose survey locations shown in Fig. 2 diversify widely, depending on the lithological and geochemical properties of the rocks within the study region (Table 1; Fig. 3a–d). In Gümüşhane province, concentration of ^{40}K ranges from 62.6 to 1680.8 Bq kg^{-1} that of ^{238}U changes from 2.5 to 119.9 Bq kg^{-1}, and ^{232}Th values vary from 3.3 to 92.4 Bq kg^{-1}. The mean values of ^{40}K, ^{238}U, and ^{232}Th for the Gümüşhane province listed in Table 2 were found to be 638.5 ± 421.6 Bq kg^{-1}, 40.8 ± 27.4 Bq kg^{-1}, and 33.5 ± 25.5 Bq kg^{-1}, respectively. The determined results indicate that the average activity values of ^{40}K and ^{238}U are higher when inspected with worldwide mean value (580 Bq kg^{-1}for ^{40}K and 40 Bq kg^{-1} for ^{238}U) of this radionuclide in the Gümüşhane province (UNSCEAR 2000). On the other hand, the average ^{232}Th values are lower than the worldwide mean value of 40 Bq kg^{-1}. Since the majority of the measurements have granitic composition, plenty of the values are approached near to the K-apex in the triangular K–Th–U plot (Fig. 4). The diversity of the concentration of radionuclide activity in the investigated region might be due to the mineral content. In the Gümüşhane Pluton, the radioactive accessory mineral assemblage of igneous rocks such as zircon, apatite, monazite and titanite mostly contain high contents of radioactive elements of Th and U (Pagel 1982).

The Th/U values change from 0.1 to 13.0 with a mean value of 1.5 ± 2.2 for the Gümüşhane granitoid (Table 2). Th versus U plot demonstrates that Th content increases with increasing U content for the study region (Fig. 5a). In Fig. 5b, Th/U versus Th plot represents Th increases with increasing Th/U ratio. The obtained results represented that Th was lesser enriched particularly in the subalkaline Gümüşhane granitoids relative to U. The U versus Th/U plot remarks decreasing U values with increasing Th/U ratios shown in Fig. 5c. The Th/U ratio reveals the enrichment or relative depletion of radioisotopes. The high radioactivity content in Gümüşhane pluton could be clarified by the geochemical and mineralogical composition of the granitoid that produced from the fractional crystallization of magma and contamination in the lower continental crust (Papadopoulos et al. 2013).

Evaluation of radiological hazard indices

Absorbed gamma dose rate in air (Dabs)

The contribution of natural radioelements to the absorbed dose rate in air (Dabs) is associated with the radioelement concentrations in the rocks and soil. The most important component of the gamma radiation is produced from radionuclides. There is a linear relationship between radioelement

Fig. 2 The radioelemental survey locations gathered from Gümüşhane pluton. The coloured circles show the intensity of the *K* [Bq kg^{-1}] concentration values for the study region

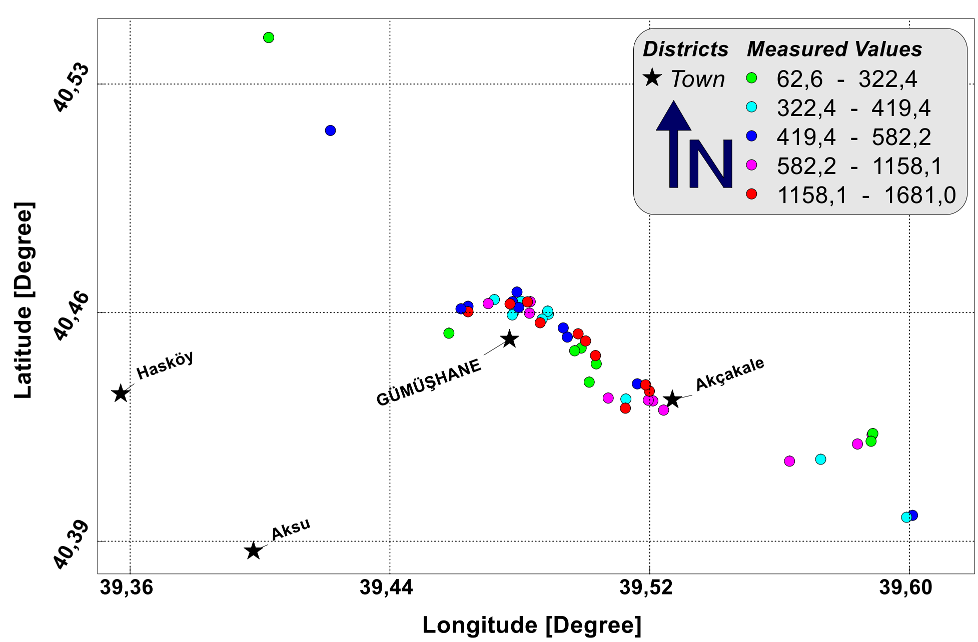

Table 1 The potassium, uranium and thorium concentrations, absorbed dose rate, radium equivalent activity and the activity concentration index for Gümüşhane granitoids

Number	K (Bq kg^{-1})	U (Bq kg^{-1})	Th (Bq kg^{-1})	Th/U	Dabs (nGy h^{-1})	Ra_{eq} (Bq kg^{-1})	AEDE (μSv $year^{-1}$)	Hex
1	150.2	63.4	15.8	0.2	45.1	97.6	55.3	0.26
2	419.4	29.9	13.8	0.5	39.6	81.9	48.6	0.22
3	460.1	58.5	16.0	0.3	55.9	116.8	68.6	0.32
4	350.6	63.5	15.8	0.2	53.5	113.1	65.6	0.31
5	331.8	26.8	20.0	0.7	38.3	80.9	47.0	0.22
6	582.2	8.9	26.8	3.0	44.6	92.1	54.7	0.25
7	341.2	52.0	19.2	0.4	49.8	105.7	61.1	0.29
8	701.1	20.6	23.4	1.1	52.9	108.0	64.9	0.29
9	472.6	18.3	17.3	0.9	38.6	79.4	47.3	0.21
10	435.1	3.2	17.7	5.5	30.3	62.0	37.2	0.17
11	341.2	17.7	8.0	0.5	27.2	55.4	33.4	0.15
12	416.3	37.5	29.0	0.8	52.2	111.0	64.0	0.30
13	450.7	11.9	20.5	1.7	36.7	75.9	45.0	0.21
14	394.4	13.8	26.6	1.9	38.9	82.2	47.7	0.22
15	322.4	26.2	10.8	0.4	32.1	66.5	39.4	0.18
16	225.4	37.7	7.3	0.2	31.2	65.5	38.3	0.18
17	363.1	26.8	20.0	0.7	39.6	83.4	48.6	0.23
18	566.5	38.8	47.5	1.2	70.2	150.3	86.1	0.41
19	989.1	40.3	66.2	1.6	99.8	211.1	122.4	0.57
20	144.0	3.0	17.6	5.9	18.0	39.3	22.1	0.11
21	510.2	52.1	19.3	0.4	57.0	119.0	69.9	0.32
22	400.6	34.8	13.6	0.4	41.0	85.1	50.3	0.23
23	694.9	14.7	36.0	2.4	57.5	119.7	70.5	0.32
24	591.6	52.7	28.6	0.5	66.3	139.2	81.3	0.38
25	162.8	36.2	10.4	0.3	29.8	63.6	36.5	0.17
26	147.1	2.5	8.4	3.4	12.4	25.8	15.2	0.07
27	391.3	32.7	7.5	0.2	36.0	73.6	44.2	0.20
28	1039.2	66.1	74.7	1.1	119.0	252.9	145.9	0.68
29	992.2	18.4	91.6	5.0	105.2	225.8	129.0	0.61
30	854.5	90.2	33.7	0.4	97.7	204.2	119.8	0.55
31	1079.9	24.1	48.2	2.0	85.3	176.2	104.6	0.48
32	1158.1	119.9	76.2	0.6	149.7	318.0	183.6	0.86
33	1680.8	38.2	51.1	1.3	118.6	240.7	145.5	0.65
34	1167.5	29.4	78.9	2.7	109.9	232.1	134.8	0.63
35	72.0	64.2	3.3	0.1	34.7	74.5	42.6	0.20
36	1411.6	97.7	92.4	0.9	159.8	338.5	196.0	0.91
37	538.4	26.9	20.1	0.7	47.0	97.1	57.6	0.26
38	425.7	65.0	65.2	1.0	87.2	191.0	106.9	0.52
39	203.5	32.6	7.4	0.2	28.0	58.9	34.3	0.16
40	62.6	51.1	9.9	0.2	32.2	70.1	39.5	0.19
41	816.9	42.2	19.7	0.5	65.5	133.3	80.3	0.36
42	1261.4	74.5	46.7	0.6	115.2	238.4	141.3	0.64
43	1486.8	54.0	90.7	1.7	141.7	298.2	173.8	0.80
44	1173.8	100.8	64.4	0.6	134.4	283.3	164.8	0.77
45	1314.6	61.5	53.3	0.9	115.4	238.9	141.5	0.65
46	472.6	36.9	19.8	0.5	48.7	101.6	59.7	0.27
47	1192.5	5.2	67.4	13.0	92.8	193.4	113.8	0.52
48	1214.4	75.3	34.3	0.5	106.1	217.9	130.1	0.59

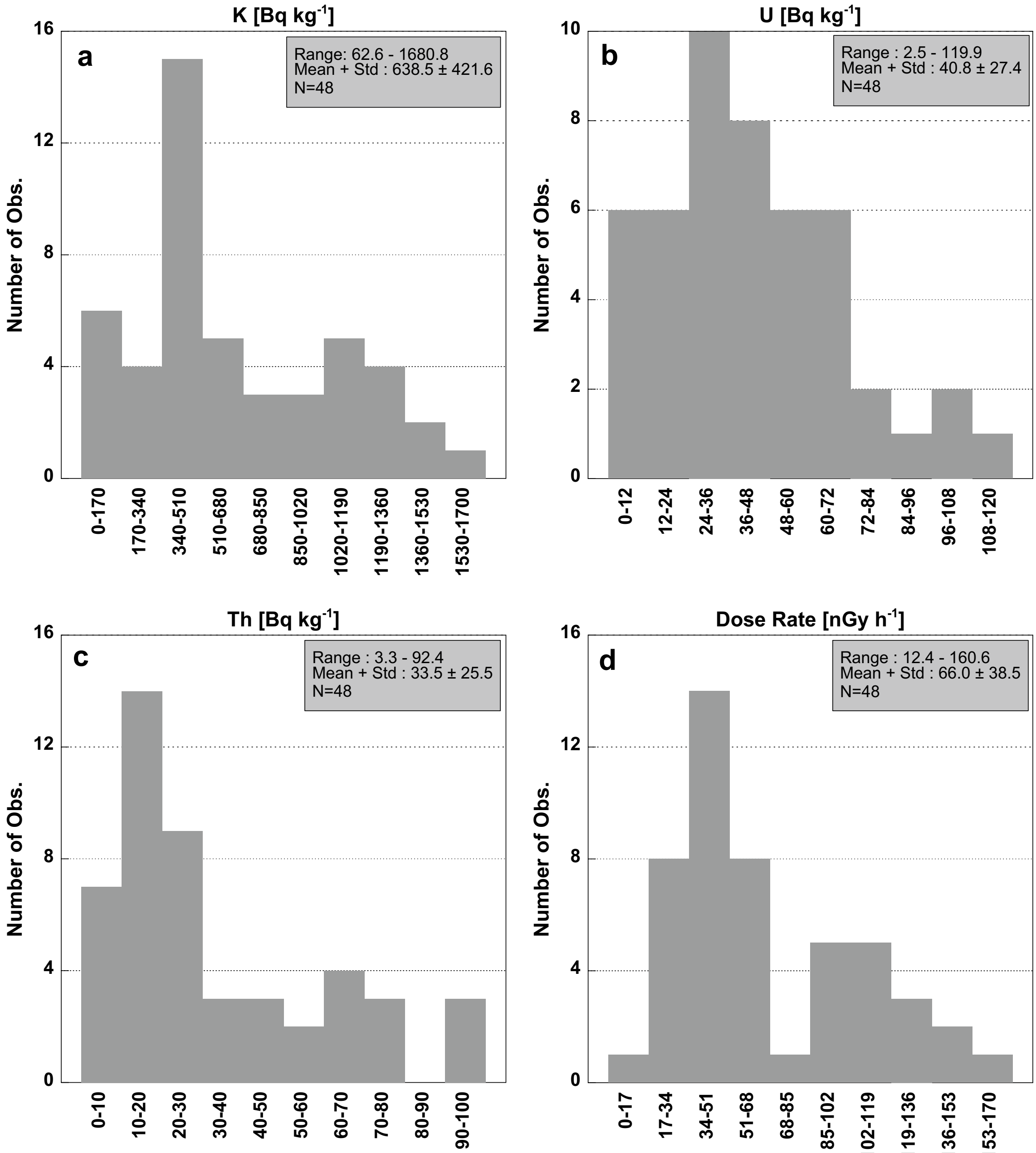

Fig. 3 The distribution-specific activity concentration and external gamma absorbed dose rates for 48 in situ measurements

concentration and terrestrial gamma radiation in rocks and soil (Beck 1972). The absorbed dose rates in air at 1 m above the ground could be computed using the equation suggested by UNSCEAR (2000).

$$\mathrm{Dabs}\,(\mathrm{nGy\ h^{-1}}) = 0.462C_{\mathrm{U}} + 0.604C_{\mathrm{Th}} + 0.0417C_{\mathrm{K}}, \quad (1)$$

where C_{U}, C_{Th} and C_{K} are the concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq kq^{-1}, respectively. Absorbed gamma dose rate changes between 12.3 nGy h^{-1} and 159.8 nGy h^{-1}with a mean value of 65.7 ± 38.3 nGy h^{-1} (Table 2, Fig. 6a). The mean value of absorbed gamma dose rate from Gümüşhane pluton is 1.15 times higher than that of the worldwide of 57 Bq kg^{-1} given in the UNSCEAR (2000) report. There is strong relationship between the mineral content such as orthoclase, feldspar and the radioactivity levels of the Gümüşhane granitoid.

Table 2 Average activity concentrations of U, Th and K (Bq kg^{-1}) in the study region

	Range	Mean ± STD	World average[a]
U (Bq kg^{-1})	2.5–119.9	40.8 ± 27.4	40
Th (Bq kg^{-1})	3.3–92.4	33.5 ± 25.5	40
K (Bq kg^{-1})	62.6–1680.8	638.5 ± 421.6	580
Th/U	0.1–13.0	1.5 ± 2.2	–
Dabs (Bq kg^{-1})	12.3 –159.8	65.7 ± 38.3	55
Ra_{eq} (Bq kg^{-1})	25.8–338.5	137.9 ± 80.6	370
AEDE (μSv year^{-1})	15.2–197.0	81.8 ± 47.3	460
Hex	0.07–0.91	0.37 ± 0.22	1

[a]UNSCEAR (2000)

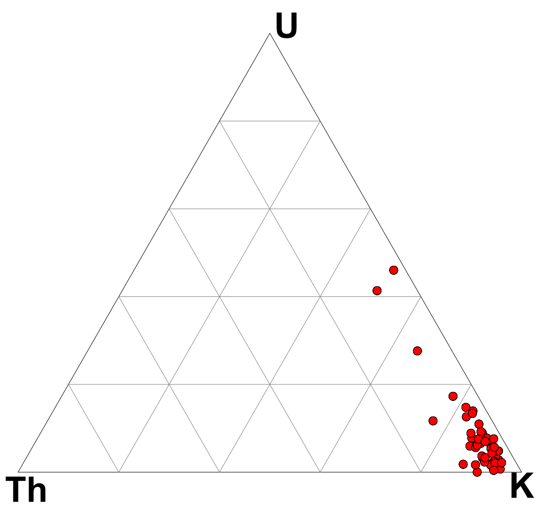

Fig. 4 Plots of K–U–Th ternary diagram showing the concentration of K, U and Th of the Gümüşhane Pluton

Radium equivalent activity (Ra_{eq})

The radium equivalent activity (Ra_{eq}) index provides guideline for adjusting the safety standards of radiation protection for the general public dwelling in the investigated region. The Ra_{eq} index reveals a weighted sum of the activities of the natural radionuclides. In granites, the distribution of radionuclide concentrations (^{40}K, ^{238}U and ^{232}Th) is not stationary. Ra_{eq} in Bq kg^{-1} defines the homogeneity with respect to the exposure to radiation to compare the specific activity of materials including various radionuclide quantities. Radium equivalent activity is computed with the following equation proposed by Beretka and Matthew (1985):

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K, \tag{2}$$

where C_U, C_{Th} and C_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1}respectively. The values of Ra_{eq} varied between 25.8 Bq kg^{-1} and 338.5 Bq kg^{-1} with a mean value of 137.9 ± 80.6 Bq kg^{-1} (Fig. 6b). Both the maximum and average radium equivalent activity values are lower than the recommended permissible value of 370 Bq kg^{-1} suggested by the Beretka and Mathew (1985) and UNSCEAR (2000) report.

Annual effective dose equivalent (AEDE)

The conversion coefficient from the absorbed dose in air to the effective dose (0.7 Sv/Gy) and the outdoor occupancy factor (0.2) suggested by UNSCEAR (2000) was utilized to compute the annual effective dose rates. Annual effective dose rate can be given by the following equation (UNSCEAR 2000; Tzortzis et al. 2003):

$$\begin{aligned}&\text{Effective dose rate } (\mu \text{ Sv year}^{-1})\\&\quad = \text{Dose rate(nGy h}^{-1}) \times 8760 \text{ h} \times 0.2 \times 0.7 \times 10^{-3}.\end{aligned} \tag{3}$$

The annual effective dose equivalent originating from external exposure to terrestrial radionuclides in the studied granitoids ranges from 15.2 to 197.0 μSv year^{-1} with a mean value of 81.8 ± 47.3 μSv year^{-1} (Fig. 6c). The values of external annual effective dose rates do not exceed the average worldwide exposure due to natural sources (UNSCEAR 2000).

External radiation hazard indices (Hex)

The external hazard index is an assessment of the natural gamma radiation hazard. The main objective of the index is to constrain the radiation dose to the acceptable dose equivalent limit of 1 mSv year^{-1}. The external hazard index (Hex) was computed from the following formula (Huda 2011):

$$\text{Hex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \le 1, \tag{4}$$

where C_U, C_{Th} and C_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The estimated external hazard index values ranged from 0.07 to 0.91 with a mean value of 0.37 ± 0.22, which is lower than the desirable value of 1 (Fig. 6d). It is clear from this study that the results show low radionuclide concentrations for the Gümüşhane province.

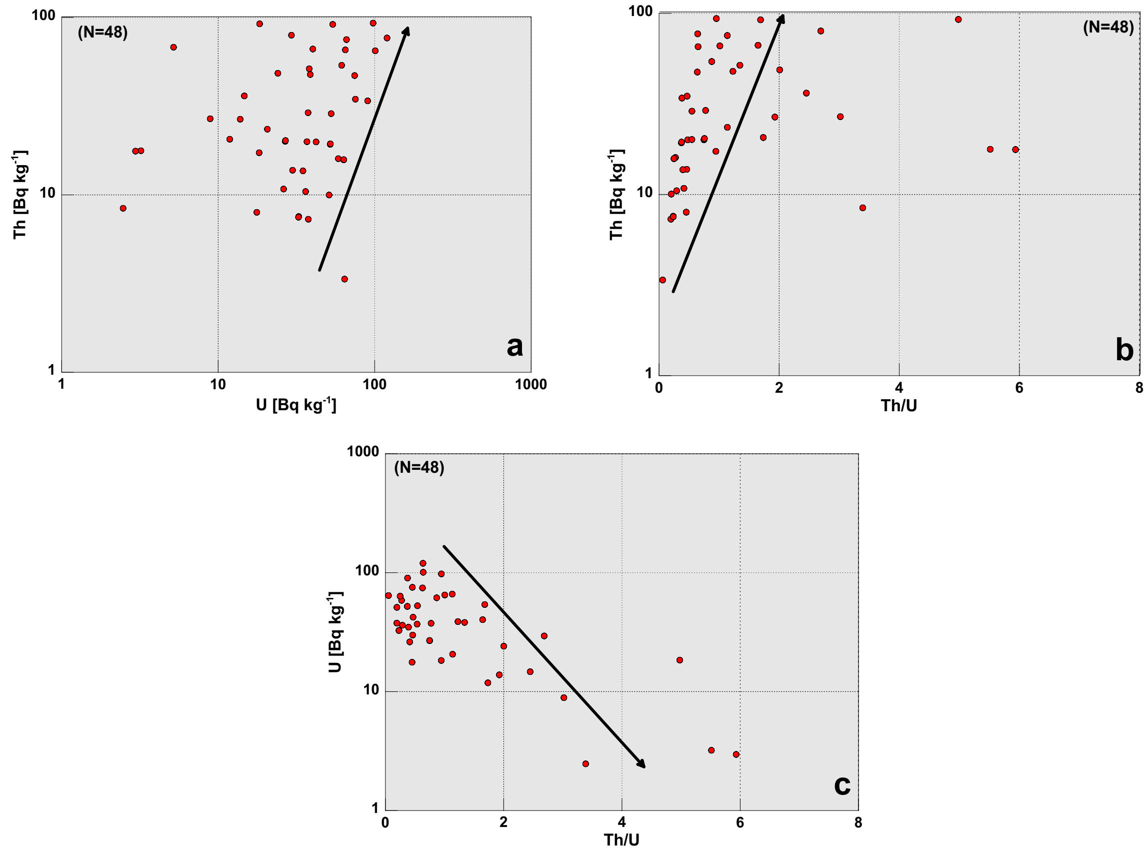

Fig. 5 Variation diagrams of the Gümüşhane province: **a** Th versus U **b** Th versus Th/U, and **c** U versus Th/U. The black arrows on the plots denote the increasing and decreasing trend orientation

Conclusion

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil samples from Gümüşhane province have been inspected with NaI (Tl) gamma-ray spectrometry. The obtained results indicated that the spreading of natural radionuclides in the study region was not uniform. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K estimated to be 40.8 ± 27.4 Bq kg^{-1}, 33.5 ± 25.5 Bq kg^{-1} and 638.5 ± 421.6 Bq kg^{-1} are higher than the world average value of 40 Bq kg^{-1} and 580 Bq kg^{-1}, respectively, in the Gümüşhane pluton (Table 2). The high-level radioactivity seen in Fig. 2 is related to the K-rich minerals (K-feldspars and biotite) and accessories (zircon, apatite, monazite, allanite and titanite) which are primarily found in granitic rocks at the center of the Gümüşhane city.

The average values of absorbed dose rate and annual effective dose are lower than the average worldwide exposure of 2.4 mSvy^{-1}. The value of radium equivalent activity (137.9 ± 80.6 Bq kg^{-1}) was fairly lower than maximum recommended value of 370 Bq kg^{-1} (UNSCEAR 2000; Fig. 6b). The computed average external hazard index (Hex) of 0.37 ± 0.22 in the study region is lower than allowable limit of 1, which means that Gümüşhane province is trustworthy for the human activity (Fig. 6d).

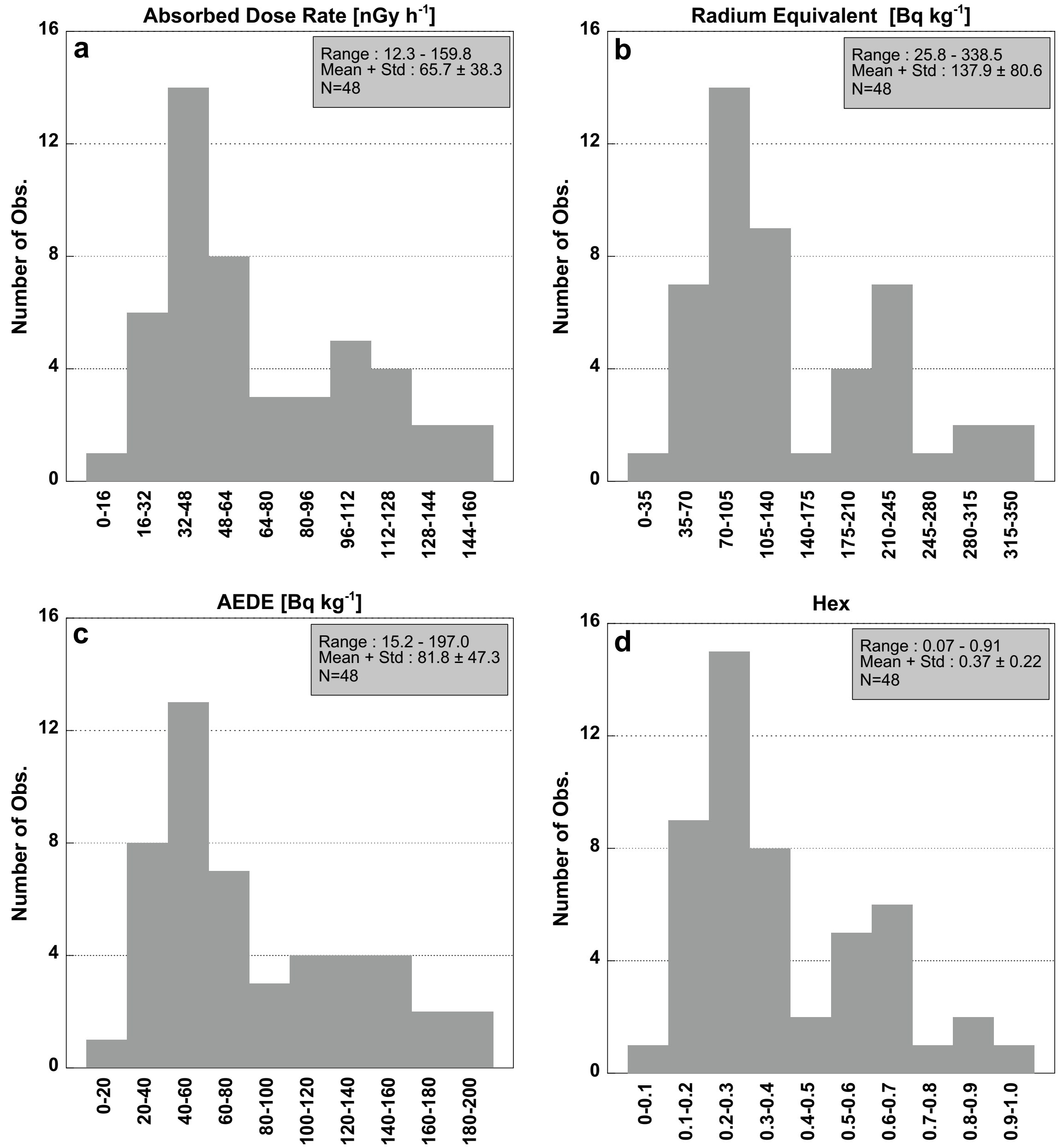

Fig. 6 The distribution-specific activity concentration and external gamma absorbed dose rates for 48 in situ measurements

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