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HİDROTERMAL ALTERASYON SAHALARINDAN KAYNAKLI DOĞAL RADYASYONA MARUZ KALMA RİSKİ: CANCA SAHASI (GÜMÜŞHANE, TÜRKİYE) ÖRNEĞİ

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Öz

Bu çalışmanın amacı, yoğun bir şekilde hidrotermal alterasyona maruz kalmış sahalarda doğal radyasyona maruziyet riskinin var olup olmadığının araştırılmasıdır. Bu kapsamda Türkiye'nin Kuzeydoğu Karadeniz Bölgesi'nde bulunan Gümüşhane İli'nin batı-güneybatısında yer alan Canca hidrotermal alterasyon sahası örnek çalışma sahası olarak seçilmiştir. Canca (Gümüşhane, Türkiye) hidrotermal alterasyon sahasından toplanmış olan iki yüzden fazla toprak örneğinde toryum (^{232}Th), uranyum ^{238}U ve potasyum (^{40}K) gibi radyoaktif elementlerin analizleri Çift Eşleştirilmiş Atomik Emisyon Spektrometresi ve Çift Eşleştirilmiş Kütle Spektrometresi (ICP-AES ve ICP-MS) cihazları kullanılarak gerçekleştirilmiştir. Analiz verileri kullanılarak sahadan toplanan toprak örneklerindeki radyoaktif elementlerin (Th ve K) aktivite değer ve dağılımları değişik parametreler hesap edilerek ve bu parametrelerin küresel ortalama değerleriyle kıyaslanarak değerlendirilmiştir. Elde edilen sonuçlar dikkate alındığında bahse konu elementler için sahadaki çoğu örnek alım noktalarında genellikle bu element aktivitelerin küresel ortalama değerleri göre daha yüksek olduğunu tespit edilmiştir. Canca (Gümüşhane, Kuzeydoğu Türkiye) hidrotermal alterasyon sahası bağlamında gerçekleştirilen bu çalışma hidrotermal alterasyon sahalarının radyasyona maruz kalma riski açısından incelenmesi gereken sahalar olduğunu göstermiştir.

Anahtar Kelimeler: Hidrotermal alterasyon sahaları, Doğal radyoaktivite, Radyolojik tehlike, Radyasyon maruziyeti, Gümüşhane.

THE RISKS OF EXPOSURE TO NATURAL RADIATIONS INDUCED HYDROTHERMAL ALTERATION SITES: CASE OF CANCA SITE (GÜMÜŞHANE, TÜRKİYE)

Abstract

The main purpose of this study is to investigate the risk of exposure of natural radiation in areas with intensive alteration development, using various methods. As a case study, Canca hydrothermal alteration site located on the west-southwest of Gümüşhane, Northeast of Turkey, was selected. Above two hundred soil samples were obtained from the Canca (Gümüşhane, Türkiye) hydrothermal alteration site and analyzed for some radioisotope elements, such as, potassium (^{40}K), uranium ^{238}U and thorium (^{232}Th) by ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometry) and ICP-MS (Inductively Coupled Plasma-Mass Spectrometry). The distributions of the activities of the radioactive elements in examined soil samples belonging to the Canca field were assessed with the help of some parameters and compared the global average concentrations of those elements. The obtained results for those elements are generally for lots of sample locations greater than the global average values of them. This study conducted in the context of the Canca (Gümüşhane, Türkiye) hydrothermal alteration site showed that the hydrothermal alteration areas should be examined for the risk of exposure to radiation.

Keywords: Hydrothermally alteration areas; Natural radioactivity; Radiological hazard; Radiation exposure; Gümüşhane.

1. INTRODUCTION

Rocks undergo alteration when exposed to hydrothermal influences. Thanks to the alteration, the original chemical content of the rocks changes with the addition and removal of new elements. Thus, while the environment is enriched in some elements, some elements become poor in the environment. If the elements with economic value are to be enriched at a desired level, the environment can be regarded as an ore deposits or mineralization area. If there is an increase over the average values of undesirable elements due to side effects, it can be assumed that the environment is contaminated by those elements (1,2). For this reason, hydrothermal alteration sites are not only important for mineral exploration studies but also for environmental risks/contaminations etc (3–7). These risks are natural radiation exposure risks, heavy metal (HM) and/or trace element contaminations (TECs) etc. in soils. The studies/researches on these kind of risks are increasing day by day (8–12). Generally, soils, especially derived from hydrothermal alteration areas, have high radiation doses as a result of primordial and/or cosmogenic radionuclides being present naturally.

Radioisotopes reach to soil through natural radioactive elements or their daughters. The ^{40}K, ^{238}U and ^{232}Th radioisotopes are mostly known as the main sources of the outer gamma radiations. This natural radiation contributes significantly to the background doses to all living creatures, especially human beings (13–15). People living in such an environment for a long time, face the risk of exposure to extreme radiation risks. Especially recent studies have shown that, especially granitic rocks have high these radionuclides concentrations (16,17). The K, U and Th concentrations of different geological environments are given in Table 1.

As mentioned above, hydrothermal alteration occurs predominantly as a result of magmatic processes and some elements are enriched in the environment due to alteration. The elemental composition of the environment changes due to the new elements coming into the environment with magmatic processes and some elements that are moving away due to the new physicochemical conditions. Therefore, as a result of alteration, these radionuclides can be enriched in the environment.

Table 1. Concentration of K, U and Th radioactive elements (ppm) and the ratios of Th/U and K/U in the Earth (18)

	K	U	Th	Th/U	K/U
CI Carbonaceous chondrites	545	0.0074	0.029	3.92	73 649
Primitive mantle	250	0.021	0.085	4.05	11905
Bulk silicate earth	240	0.023	0.0795	3.46	10435
Continental crust Upper	27500	2.5	10.5	4.20	11 000
Middle (Archaean)	17500	2.2	8.4	3.82	7954
Lower (Archaean)	8333	0.05	0.42	8.40	166660
Lower and Middle (post Archaean)	20000	1.25	6	4.80	16000
Average (1)	17500	1.3	5.7	4.38	13461
Average (2)	12500	1.25	4.8	3.84	10000
Oceanic crust Normal Mid-Ocean Ridge Basalt (NMORB)	600	0.047	0.12	2.55	12766
Ocean Island Basalt (OIB)	12000	1.02	4	3.92	11765

In determining of natural radiation level of soils, the gamma-ray spectrometry method has been widely used. However, there is a method to determine of natural level of soil. In this method, concentrations of elements are converted into respective concentration to Becquerel per kilogram (Bq kg^{-1}) using associated equations (19–21)

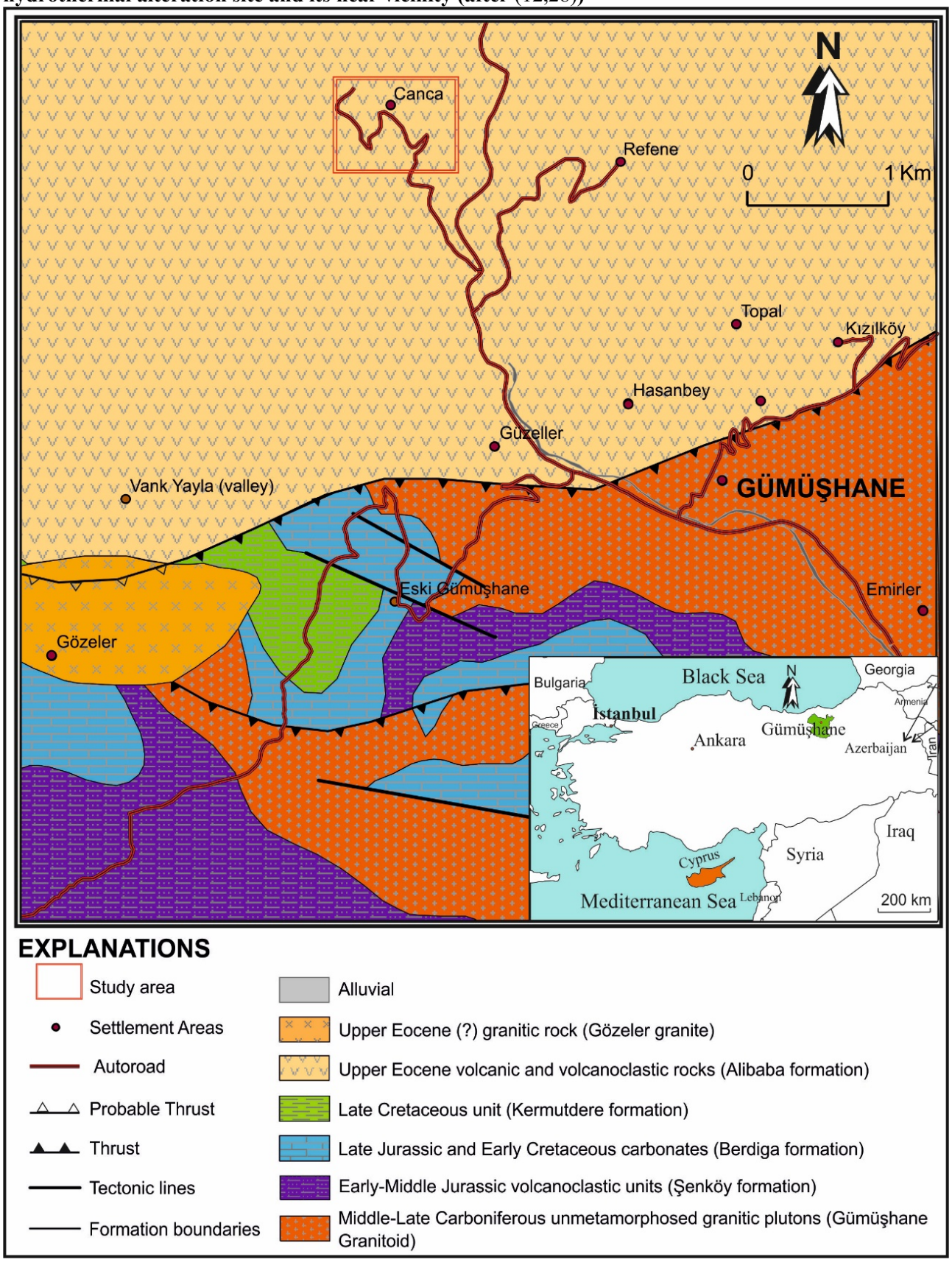
The aim of this study is to examine/researche/investigate the risk of exposure of natural radiation in sites with intensive hydrothermal alteration development, depending on concentrations converting into respective concentration to Becquerel per kilogram (Bq kg^{-1}) using associated equations for natural radioactive element and using related parameters. For this purpose, Canca (Gümüşhane, NE Turkey) hydrothermal alteration site was examined as a case study, which is located on the southwest of Gümüşhane, NE Türkiye.

The study site covers an area of approximately 1.5-2 km^2. The Middle-Late Eocene Alibaba formation outcrops in the examined site. The unit is formed by basalt and andesite in the study area and is occasionally cut by dykes of andesitic and/or doleritic which are the final products of the Eocene volcanism in the region. In close vicinity of the study area the Gözeler granite, which is considered to be Upper Eocene (?) is observed (22) (Fig.1). The alteration in the area possibly might have developed due to the Upper Eocene granitic intrusive rocks which outcrop in many areas such as Upper Eocene Gözeler (Old Gümüşhane), Avliya (Torul-Gümüşhane), Demirören-Sarıçiçek-Dölek (Gümüşhane-Center) in the region and the Upper Cretaceous granitic rocks in northwest of the study area (22–25). Hydrothermal fluids, mainly the products of the Upper Eocene magmatic processes, have reached shallow geochemical environments along the tectonic lines developed as a result of the neotectonic evolution of the region and altered the predominantly andesitic rocks in the Canca area (4,26). The intense hydrothermal alteration (silicification, argillization, limonitization, pyritization, and hematitization) is observed along/within fracture lines. As mentioned earlier, alteration process reorganizes the geochemistry of the environment. According to geochemistry and fluid inclusion geochemistry researches, major hydrothermal fluid solutes are cations of Ca, Mg, K, Na, Mn, Ba, Fe, anions of C, Si, S, Cl, P, N, metals such as Pb, Cu, Zn, Ag, Au, U, gaseous species (H_2S, CO_2, SO_2, CH_4, N_2) and hydrocarbons (27). Potassic alteration is especially common in epithermal and porphyry mineralizing processes. The minerals characteristic of this hydrothermal alteration are K-bearing feldspar and biotite in these systems. The type of phyllic/sericitic hydrothermal alteration is the assemblage of quartz-sericite-pyrite minerals. As can be seen, in both types of alteration, K is added to the original rocks. Because U and Th have large size atom and positive valences, they are not capable of entering into the common igneous rock minerals. Therefore, these elements are concentrated in late-stage silicate melts or hydrous and gaseous fractionates and tend to be taken up by secondary minerals (e.g. rutile, apatite, titanite or monazite. Considering these processes in the hydrothermal alteration, it was concluded that the alteration sites are important place in terms of natural radiation of elements such as, especially U, Th and K. So, in this study, the radioactive properties and exposure risks of this kind of site have been examined.

2. METHODS

Above two hundred soil samples were obtained from the Canca hydrothermal alteration site (Gümüşhane-Turkey). The samples were taken from the B zone of soil profile (about up to 25 cm) which the soil is ideally developed, and element enrichment occurred. The samples were dried under natural conditions in a non-humidity environment, then the product was dried at 60°C for 2 days and the natural moisture was removed. After that, the samples were passed under 2 mm polyethylene sieve and the samples passing through the sieve were milled with a ring mill. They then passed through 80-mesh sieve to obtain 10-15 g packages from soil samples in ideal sizes and analyzed for uranium, thorium (^{238}U and ^{232}Th) and potassium (^{40}K) by Inductively Coupled Plasma-Atomic

Figure 1. Study sites location map and general geological map of the Canca (Gümüşhane, NE Türkiye) hydrothermal alteration site and its near vicinity (after (12,28)**)**



Emission Spectrometry (ICP-AES) and Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) at Gümüşhane University, Central Research Laboratory (Gümüşhane, Türkiye) and at Bayburt University, Central Research Laboratory (Bayburt, Türkiye). In accordance with the procedure recommended by Bulut et al. (29), dried and milled soil samples were digested in a closed microwave digestion system (Milestone ETHOS D, Italy) to obtain limpid solutions before determining element concentrations of the samples using ICP-AES and ICP-MS.

The digestion procedure and analysis of the elements can be summarized as follows; 0.25 g of soil sample was weighed into the teflon vessel of the microwave oven, 1.5 mL HNO_3 and 4.5 mL HCl were added on it. This soil and acid mixture in the vessel was digested by microwave at a 45 bar pressure. After digestion process, soil suspension was filtered, and the supernatant was completed to 50 mL with ultrapure water. The detection limits for the elements were calculated as three times the standard deviations of 20 replicate measurements of blank samples as summarized in (10,30).

The concentrations of elements are in percent (%) for K and parts per million (ppm) for U and Th elements. Due to some inconsistencies in the element uranium results, it is planned to analyze the uranium data later on. Therefore, K and Th data were evaluated as initial findings. The K and Th concentrations were converted into Becquerel per kilogram (Bq kg^{-1}) using associated equations and results were assessed. The equations of conversion from concentration to Becquerel per kilogram (Bq kg^{-1}) are in accordance with International Atomic Energy Agency (19) and (2,20) models:

$$1 \text{ part per million (ppm) of } {}^{238}U = 12.35 \text{ Bq kg}^{-1} \quad (1)$$

$$1\% \text{ of } {}^{40}K = 313 \text{ Bq kg}^{-1} \quad (2)$$

$$1 \text{ part per million ppm of } {}^{232}Th = 4.06 \text{ Bq kg}^{-1} \quad (3)$$

In this study, uranium concentrations were not taken into account for the above-mentioned reason.

3. RESULTS AND DISCUSSION

The distributions of the activities of the radioactive elements, ^{232}Th and ^{40}K etc. in soil samples from the study area were evaluated using some statistics parameters and global average values of those elements.

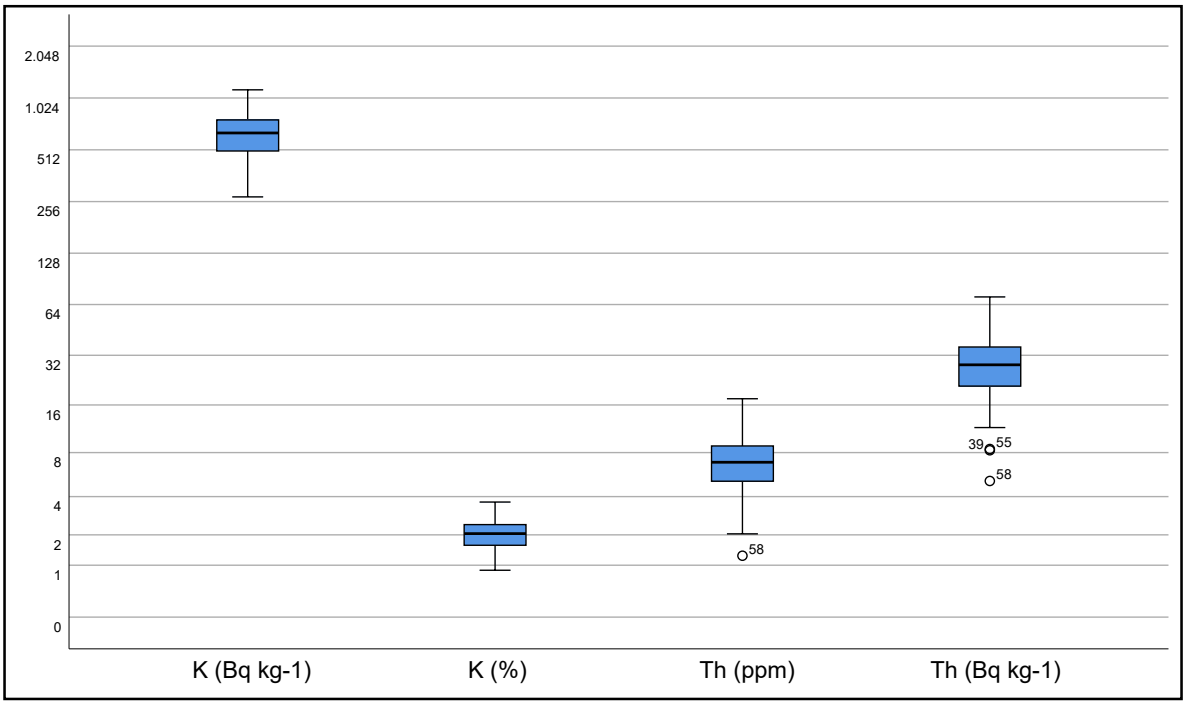
The activity concentrations' descriptive statistics and the converted results (in Bq kg^{-1})'s descriptive statistics are revealed in Table 2 and Fig. 2.

The activities range from 263,17 to 1141,95 Bq kg^{-1} for ^{238}K with an average of 625,15 Bq kg^{-1}, 5,6 to 70,93 Bq kg^{-1} for ^{232}Th with an average of 29,42 Bq kg^{-1} respectively. More than 75% of the results obtained for ^{40}K have higher values than the recommended limit of 400 Bq kg^{-1} as reported by Chandrasankaran et al. (2014). About 25% of the obtained results for ^{232}Th are greater than the global average value of 35 Bq kg^{-1} (31,32). Based on these findings it was seen that the obtained results of ^{232} Th and ^{40}K elements for lots of sample locations are generally higher than the global average concentrations of them.

Table 2. The concentrations of natural radioactivity in soil samples of Canca (Gümüşhane, NE Türkiye) hydrothermal alteration site

	K		Th	
	%	*Bq kg-1*	*pmm*	*Bq kg-1*
Mean	2.00	625.53	7.25	29.42
St.Eror	0.05	16.68	0.27	1.08
Median	1.97	616.79	6.91	28.05
St.Dev.	0.63	198.75	2.98	12.08
Variance	0.40	39502.33	8.85	145.91
Minimum	0.84	263.17	1.27	5.16
Maximum	3.65	1141.95	17.47	70.93

Figure 2. Box plot diagrams of concentrations of natural radioactivity in soil samples of Canca (Gümüşhane, NE Türkiye) hydrothermal alteration site



In the study carried out by Vural (1,2) in the Demirören (Gümüşhane, NE Türkiye) hydrothermal alteration area in the region, it was observed that the activity values obtained for ^{40}K exceeded the recommended 400 Bq/kg. It was determined that the ^{232}Th activities of more than 75% of the samples from the field exceeded the threshold value (30 Bq/kg), while for ^{238}U, more than 25% of the samples exceeded the threshold value of 35 Bq/kg. Researchers concluded that the high natural radioactivity values in the area were caused by intense hydrothermal alteration. Considering the general geological characteristics of the area in question, the geochemical processes associated with the hydrothermal alteration and their results, it is concluded that the effect of hydrothermal processes at high natural radioactivity concentrations cannot be denied. Because hydrothermal processes are effective in the liberation and relative enrichment of K, U and Th elements.

In the study carried out by Kaya et al. (33) on the natural and artificial radioactivity of the mineralization and hydrothermal alteration areas in the Gümüşhane region, remarkable and

confirming the findings of this study's results were also obtained (13,14,21,33–36). In the studies carried out by the aforementioned researchers at the Eskiköy (Torul, Gümüşhane, NE Türkiye) ore deposit site, it was determined that the natural ^{40}K radio isotope activities exceeded the weighted world average for most sampling points, while the ^{232}Th and ^{238}U activities were above the weighted world average at some sampling points. They determined that the ^{137}Cs activities, which represent the most common artificial radioactivity, exceeded 3 Bq/kg at some sampling points of the field, so they were conclude that there was also an anthropogenic source of radioactivity pollution in the field (13,34).

In the studies conducted by Kaya et al. (33), and Vural and Kaya (14,37) on natural and artificial radio isotope activities in Arzular-Dölek-Yitirmez (Gümüşhane, NE Türkiye) hydrothermal alteration and mineralization sites, it has been determined that natural radioactivity values (for ^{232}Th and ^{238}U) exceed the weighted world averages for most sampling points. The researchers concluded that the high radioactivity values of the site are due to the geological environment triggered by hydrothermal alteration and mineralization processes. They reported that the artificial radioactivity concentrations measured in the field generally exceed 2 Bq/kg, so there is a radioactive contamination of anthropogenic origin - possibly associated with the Chernobyl Nuclear Accident - in the field.

Another study in the region was conducted by Maden et al., (38) on granitic rocks in the region. 48 in situ measurements on radio activities of ^{40}K, ^{232}Th and ^{238}U radioisotopes at the Gümüşhane granitoid were carried out by these researches using gamma-ray spectrometer with a NaI(Tl) scintillation detector. According to their findings, it was seen that the radionuclide activity concentrations are from 62.6 to 1680.8 Bq kg^{-1} for ^{40}K, 2.5 to 119.9 Bq kg^{-1} for ^{238}U, and 3.3 to 92.4 Bq kg^{-1} for ^{232}Th. The results of researchers obtained with gamma ray spectrometry are also remarkable for the region.

It was affirmed from first findings that the radiological hazards occur as a result of contributions from naturally occurring radionuclides (thorium, uranium, potassium etc.) induced from hydrothermally alterated sites. As a concluded that the Canca (Gümüşhane, NE Türkiye) alteration site is contaminated zones for people who situated at the hydrothermal alteration area. Taking into consideration the results obtained, it is suggested to investigate the effect of radiation on people who live in the area by carrying out detailed studies. The findings are the first findings of the area and detailed studies of the area are continuing. The first findings were shared in order to draw attention to the importance of the subject and detailed study results will be given separately.

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Conflict of interests

The author declared no conflict of interest regarding this study.

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