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Gamma spectrometry measurements of natural and artificial radioactivity of Saklıkent-Antalya and its correlation to quarries

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Abstract

We investigated the natural and artificial radioactivity levels of soil samples from Saklıkent, Antalya due to the radioactive isotopes of ²³⁸*U*, ²³²*Th*, ⁴⁰*K* and ¹³⁷*Cs* via gamma spectroscopy. While ¹³⁷*Cs* is the only contributor of artificial radioactivity, the rest of the isotopes contributes natural radioactivity. Absorbed dose rates, radium equivalent activities, annual effective dose equivalents and internal and external hazard indexes are also calculated from measured levels. Results were compared with reported limit values of literature. Anomalies were observed in the activity concentration of the ^{238}U , ^{232}Th and ^{40}K . It is concluded that these anomalies explicitly come from the quarries. On the other hand, the anomaly observed in ¹³⁷*Cs* is clearly dependent on the fallout from nuclear accidents like Fukushima. This study will be a useful reference for literature, especially after a nuclear plant is established in Mersin, Akkuyu region of Turkey.

Keywords Natural radioactivity · HPGe detector · Soil samples · Gamma-ray spectrometry · Quarry

Introduction

Natural and artificial radioactivity level measurements of soil, sand, food, building materials as well as water have taken a great deal of interest recently and many studies have been published for the different regions of world. Since every human being is affected from radioactivity during their life-cycle, whether its source natural or artificial, it is very important to monitor the levels of environmental radiation (U.N.S.C.E.A.R [2000;](#page-10-0) Gunoglu [2018\)](#page-9-0).

Natural radioactivity mostly comes from the radionuclides with long half-lives like ^{238}U and ^{232}Th which are comparable with the age of earth. It is well known that this kind radioactivity depends on the geological structure.

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Radon, which is a product of almost all natural radioactive series, can be found in all rocks bounded to the minerals beneath the Earth's surface (Krane [2008\)](#page-9-1) and it is probably one of the most dangerous sources for human health. Potassium-40 (^{40}K) is also a naturally occurred radioisotope that can be found in environment. Although ^{40}K is not a product of decay of the radioactive heavy elements, it can be as dangerous as Radon because of its long halflife $(1.2 \times 10^9 \text{ y})$ and its high abundance (NUDAT2 [2020;](#page-9-2) U.N.S.C.E.A.R [2000\)](#page-10-0).

Besides the natural radioactivity, products of nuclear interactions can be also found in environment and they also possess danger for human health (Eke and Boztosun [2015\)](#page-9-3). Nuclear power plant accidents and weapon tests are the source of this kind of radioisotopes $(^{137}Cs, ^{131}I, ^{95}Zr$ and ⁹⁰*Sr*). Due to Chernobyl and Fukushima nuclear power plant accidents, huge amount of artificial radioactivity has been occurred (U.N.S.C.E.A.R [2000;](#page-10-0) Eden et al. [2017;](#page-9-4) Kritidis et al. [2012;](#page-9-5) Povinec et al. [2003;](#page-10-1) Steinhauser et al. [2018\)](#page-10-2). The level of these radionuclides is also important to monitor in order to see the effects of nuclear power plants and nuclear weapon tests to the environment.

Recent studies showed that the most of the natural radioactivity, which is dangerous for human health, comes from the soil or the products of the soil (Kaniu et al. [2018;](#page-9-6) Unal et al. [2018;](#page-9-8) Petrović et al. 2018; Dżaluk et al. 2018; Loan et al. [2018;](#page-9-9) Al-Hamarneh [2018;](#page-8-0) Yalcin and Unal [2018;](#page-10-4) El Samad et al. [2018;](#page-10-5) Turhan et al. 2018; Džolji et al. [2017;](#page-9-11) Goren et al. [2017;](#page-9-12) Shehzad et al. [2019;](#page-10-6) Kuluozturk and Dogru [2015;](#page-9-13) Cetin [2016;](#page-8-1) Kulali et al. [2018;](#page-9-14) Gunoglu and Seckiner [2018;](#page-9-15) Gunay et al. [2020;](#page-9-16) Kuluozturk et al. [2020\)](#page-9-17). Since these rodionuclides pass to plants and foods through soil, radiation levels of foods and plants are also a topic of interest (Khalil et al. [2018;](#page-9-18) Ajayi et al. [2018;](#page-8-2) Turkekul et al. [2018;](#page-10-7) Canbazoglu et al. [2018\)](#page-8-3). Building materials may also possess radionuclides. These materials must be studied very carefully because they are used to create closed environments. If the building materials have high level concentrations and these closed areas do not get enough air, these materials could become very dangerous for human health (Eke and Boztosun [2015;](#page-9-3) Aykamis and Kilic 2011 ; Korkulu and Özkan 2013 ; Ozmen et al. 2014 ; Agar et al. [2014;](#page-8-5) Trevisi et al. [2018;](#page-10-8) Joel et al. [2018;](#page-9-21) Abdellah et al. [2018;](#page-8-6) Kasumovic et al. [2018;](#page-9-22) Altun et al. [2017\)](#page-8-7).

In this study, it has been aimed to determine the natural and artificial radioactivity levels of the Bakırlıtepe Mountain. An important detail about Bakırlıtepe is that a touristic ski-center is placed on top of it. We have seen that although this region attracts many people during the year, it has never been monitored to for the levels of natural and artificial radioactivity.

Methodology

Description of study area

Study area is located at the south of Turkey. Bakırlıtepe (Saklıkent) is one the highest peak in Beydagları Mountain ˘ Range. The height of the Bakırlıtepe is almost 2500 *m*. There is a ski center on the top of the mountain. There is also a national observatory very close to the ski center. Both, the ski center and the observatory attract many domestic and international tourists every year. The area around the mountain is not densely populated but, many people visit the mountain for the reasons given above during a year. So, this study is limited by the mountain itself instead of a wider area.

In this study, 21 soil samples were collected from city level to the top of mountain. The coordinates and their altitudes are given in Table [1.](#page-1-0) The exact locations of coordinates are shown in Fig. [1.](#page-2-0) In addition, the locations of construction sites of two nuclear power plants in Turkey are also shown with the small map in Fig. [1.](#page-2-0) Akkuyu Nuclear Power Plant, which is placed in Mersin City, is close to Saklıkent. The red rectangular indicates the area of the collected samples in Fig. [1.](#page-2-0)

Table 1 Coordinates and their altitudes of the sample locations

Sample number	Latitude (north)	Longitude (east)	Elevation (m)
$\mathbf{1}$	36° 53.850'	30° 31.333'	114
$\sqrt{2}$	36° 53.756'	30° 30.518'	312
3	36° 53.932'	30° 29.535'	501
$\overline{4}$	36° 54.096'	30° 28.385'	614
5	36° 53.988'	30° 27.733'	710
6	36° 53.744'	30° 27.000'	862
7	36° 53.906'	30° 26.579'	1045
8	36° 54.141'	30° 24.039'	1112
9	36° 53.467'	30° 22.224'	1218
10	36° 52.778'	30° 22.068'	1374
11	36° 52.518'	30° 20.960'	1606
12	36° 50.952'	30° 20.318'	1787
13	36° 50.360'	30° 20.030'	1883
14	36° 50.557'	30° 19.830'	1943
15	36° 50.373'	30° 19.716'	1999
16	36° 50.323'	30° 19.604'	2021
17	36° 49.980'	30° 19.561'	2028
18	36° 49.947'	30° 19.579'	2029
19	36° 50.123'	30° 19.488'	2032
20	36° 50.238'	30° 19.502'	2046
21	36° 49.870'	30° 19.731'	2051

Fig. 1 The locations of collected samples. Samples between 14 − 21 were collected at the top of the mountain. Construction Sites of Nuclear Power Plants in Mersin and Sinop are also shown in the small map. The red rectangular shows the Bakırlıtepe

Sample collection

In this paper, we mainly interested in how the natural and artificial radioactivity of soil samples are changed with height at Bakırlıtepe Mountain. 21 soil samples were collected to achieve this purpose.

The samples from Number 1 to Number 14 were collected with 100 − 200 *m* steps. After reaching the top of the mountain at near 2000 *m*, 8 more samples were collected. Since the top of the mountain mostly formed by rocks, these samples were collected as rocks. All the samples were collected from 10−15 *cm* beneath the surface. Also, all the samples were dried before measuring the activity levels. The rocks and other samples were pulverized to get a homogeneous distribution in their containers. The volume of each container is approximately 100 ml. The containers were sealed to prevent air contact with the pulverized samples. Sealing the containers is necessary to reach a secular equilibrium state for ²²⁶*Ra* and ²²²*Rn*. The secular equilibrium can be established after five to seven times of half-life (3.82 d) of the daughter nucleus, ²²²*Rn* (Chiozzi et al. [2000\)](#page-9-23). So, the suitable waiting time must be at least 4 weeks (Gorur et al. [2011;](#page-9-24) Al Rashdi et al. [2021\)](#page-8-8) or more (Al-Hamarneh and Awadallah [2009\)](#page-8-9). Samples were waited for 40 days to reach an equilibrium state in this study.

Analysis

Before analyzing the soil samples, we measured the background radiation of the detection chamber. The background measurement is important for two reasons. First, the background must be subtracted from the sample measurements in order to get radiation counts only from the soils. Second, minimum detection limits must be determined for the activity concentrations. Any measurements that below these limits (*MDL*), will not be included in activity concentrations tables and will be regarded as "*< MDL*".

The minimum detection limits of ${}^{40}K$, ${}^{137}Cs$, ${}^{232}Th$ and ²³⁸*U* were calculated as 32.11 *Bq/kg*, 0.02 *Bq/kg*, 1.10 *Bq/kg* and 2.73 *Bq/kg*, respectively with help of the background spectrum. Since we cannot directly measure the activity concentrations of ²³⁸*U* and ²³²*Th*, their daughter nuclides were used for these calculations. In the gamma spectra of samples, ^{214}Pb and ^{214}Bi lines were used for ^{238}U and ^{228}Ac line was used for ^{232}Th .

All measurements were made by a p-type, electrically cooled, Coaxial High Purity Germanium (*H P Ge*) Detector from AMATEK-ORTEC (*(GEM40P4-83)*) with 40% relative efficiency and an energy resolution of 768 eV for a gamma energy of 122 keV ($57Co$) and 1.85 keV for a gamma energy of 1332.5 keV (${}^{60}Co$). The detector and samples were placed in a 10 cm thick lead shield in order to measure the radioactivity of samples. The inner surface of the shield is also covered with a 2-mm-thick copper foil to minimize the back-scattering of low energy particles (Xrays). The energy and efficiency calibration of this detector were studied by Ozmen et al. [\(2014\)](#page-9-20). Both, the samples and background were counted for 86400 *s* in order to get consistent results. MAESTRO-32 software was used for the analysis of spectra (ORTEC [2006\)](#page-9-25).

Activity concentration and dose evaluations of soil samples

In order to calculate activity concentrations, one must consider Eq. [\(1\)](#page-3-0) (Agbalagba E et al. [2012\)](#page-8-10)

$$
A = \frac{N}{m.t.\epsilon_0.P_\gamma} \tag{1}
$$

In Eq. [\(1\)](#page-3-0), *A* is the activity concentration, *N* is entry or count number of the interested peak after background is subtracted, *m* is the mass of the soil sample, *t* is counting time of sample, ϵ_0 is efficiency of *HPGe* detector and *P*^γ is the gamma emission probability. When *m* is taken as *kg* and *t* as *s*, the activity concentration will be calculated

Table 2 Activity concentrations of soil samples

in *Bq/kg*. Minimum detectable activity (*MDA*) can be calculated using Curie Equation (Currie [1968\)](#page-9-26).

Since the natural radioactivity is caused by existence of Thorium, Radium and Potassium (Krane [2008\)](#page-9-1), it might be useful to calculate absorbed dose rates. With help of the activity concentrations, one can evaluate absorbed dose rate *D* from 1 *m* above the ground level with Eq. [\(2\)](#page-3-1), where A_U , *A_{Th}* and *A_K* are activity concentrations of ²³⁸*U*, ²³²*Th* and ⁴⁰*K* respectively (U.N.S.C.E.A.R [2000\)](#page-10-0).

$$
D(nGyh^{-1}) = 0.462A_u + 0.604A_{Th} + 0.0417A_K \tag{2}
$$

Another dose evolution parameter from U.N.S.C.E.A.R [\(2000\)](#page-10-0) is annual effective dose equivalent (AEDE). This useful dose calculation is only meaningful after adding outdoor occupancy and absorbed dose in air to effective dose coefficients which are 0.2 and 0.7 *(Sv/Gy)*, respectively.

$$
AEDE = D(nGyh^{-1}) \times 8760(h/y) \times 0.2
$$

×0.7(Sv/Gy) × 10⁻³ (3)

We can add one more radiological hazard index which is Radium equivalent activity index (*Raeq*). When calculating "*Raeq* ", it should be considered that the Radium concentrations are not same for all the isotopes. The variations of ²³⁸U, ²³²Th and ⁴⁰K in soil, sand, rock, stone, water, etc. are not constant to evaluate. Therefore, a general

*Not detected

Fig. 2 Distribution of ¹³⁷*Cs* activity concentration with altitude. The maximum value is removed from the graph in order to draw an understandable figure

radiological index which is called radium equivalent activity is required for the actual radioactivity of ^{226}Ra , ^{232}Th and $40K$ in environmental samples. This index ensures beneficial recommendation in arranging the safety standards in radiation protection for human population (Garcêz et al.

[2018;](#page-9-27) Beretka and Matthew [1985\)](#page-8-11). Radium equivalent activity can be calculated as (Korkulu and Özkan 2013 ; Beretka and Matthew [1985;](#page-8-11) Uosif et al. [2008;](#page-10-9) Ravisankar et al. [2012\)](#page-10-10)

$$
Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K
$$
\n(4)

The last two hazard indexes that measure the health of environment are internal (H_{in}) and external (H_{ex}) hazard indexes (Korkulu and Özkan [2013;](#page-9-19) Beretka and Matthew [1985;](#page-8-11) Uosif et al. [2008;](#page-10-9) Ravisankar et al. [2012\)](#page-10-10). These values are expected to be less than "1" for a safe environment for human health.

$$
H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}
$$
 (5)

$$
H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}
$$
 (6)

Results and discussion

Activity concentrations of interested radionuclides were calculated and these values are given in Table [2.](#page-3-2) According to Table [2,](#page-3-2) the activities range from 7.51 ± 0.74 *Bq/kg* to 53.04 \pm 2.43 *Bq/kg* for ²³⁸*U*, 10.98 \pm 5.06 *Bq/kg* to 75.54 ± 4.95 *Bq/kg* for ^{232}Th , 205.00 ± 40.10 *Bq/kg* to

Fig. 3 The location of the Sample11. It is between three quarries

Fig. 4 Distribution of ²³⁸*U* activity concentration with altitude

 $1713.94 \pm 82.72 Bq/kg$ for ^{40}K , and $1.55 \pm 0.56 Bq/kg$ to 138.97 \pm 6.55 *Bq/kg* for ¹³⁷*Cs*. As seen in Table [2,](#page-3-2) some of the concentrations of ¹³⁷*Cs* and ²³²*T h* are below *MDL*.

¹³⁷*Cs* is an artificial radionuclide and this kind of radionuclides come as residue from nuclear power plants, nuclear weapon tests and accidents like Chernobyl in 1986 and Fukushima in 2011. Despite the activity concentrations of ¹³⁷*Cs* seem very low in soil samples from Bakırlıtepe, they are higher than activities for samples from sea level of Antalya given in Ref. Eke and Boztosun [\(2015\)](#page-9-3). It is also noticed that there is an anomaly over 2000 *m* for ¹³⁷*Cs*. Figure [2](#page-4-0) shows the distribution of activity concentration with altitude for $137Cs$. In Fig. [2,](#page-4-0) the anomaly point (the maximum activity concentration) is removed from data because its value exceeds all other data enormously and prevent drawing a suitable graph. The maximum activity concentration is 138.97 *Bq/kg*, while the highest activity concentration in Fig. [2](#page-4-0) is 9.88 *Bq/kg*. The sample point of this anomaly is placed near a water drainage way. Clearly, this is a cumulative point for $137Cs$ because of these waters from rains and snow. The winds also carry the radionuclides to slope of hills and the mountains and they may cause some cumulative radionuclide points (Ritcihie and McHenry [1990\)](#page-10-11).

In Table [2,](#page-3-2) it is noticed that Sample 11 has the maximum values except for $137Cs$. However, minimum values are seen in different samples; for ${}^{40}K$ in sample 10, for ${}^{137}Cs$ in sample 3, for $23^{2}Th$ in sample 17 and for $23^{8}U$ in sample 5. For $137Cs$, it is interesting to get very different values for very close sample points. Especially, samples between

²³²Th Activity Concentration World Averag 80 70 Activity Concentration[Bq/kq] Activity Concentration[Bq/kg] 60 50 40 $3⁰$ \mathfrak{p} ł 10 0_c 0 200 400 600 800 1000 1200 1400 1600 1800 2000 2200 Altitude[m]

Fig. 5 Distribution of ²³²*T h* activity concentration with altitude

Fig. 6 Distribution of $40K$ activity concentration with altitude

14 − 21 which are from top of the mountain vary too much for ¹³⁷*Cs*. This is an interesting result because even the altitudes of locations are different, they are very close to each other.

Figure [3](#page-4-1) shows the location where we collected the Sample 11 (Google Earth [2020\)](#page-9-28). This location is placed between three marble quarries. The quarries are shown with red circles in Fig. [3.](#page-4-1) Furthermore, Figs. [4,](#page-5-0) [5](#page-5-1) and [6](#page-6-0) show the

comparison between world averages and our results. It is clear that the marble quarries affect the natural radioactivity around their vicinity as seen on Sample 11. It can be seen that there is another anomaly for Sample 4 in Fig. [4](#page-5-0) and Fig. [5.](#page-5-1) Activities of ²³⁸U and ²³²Th are very high in this region. Since there are no quarries around this location, we concluded that it must be an effect of the structure of the rocks.

*Not detected

The activity concentrations of the ${}^{40}K$ are higher than world's average values almost in all samples (see Fig. [6\)](#page-6-0). The quarries are placed on higher altitudes and we have not observed any other variable that can increase these values. So, this must be related with only the structure of the rocks and soil of the mountain. The other possible reasons are the water drainage ways or winds like in ¹³⁷*Cs* .

Table [3](#page-6-1) shows the most commonly used hazard indexes for environmental human health. Because of Thorium levels in sample 15 and 18 are below *MDL*, these indexes are marked as *n.d.* (not detected) in Table [3](#page-6-1).

The world's average values of activity concentrations and hazard indexes are given in U.N.S.C.E.A.R [\(2000\)](#page-10-0). For the activity concentrations, these values are 33 Bq/kg for ^{238}U , [4](#page-7-0)5 Bq/kg for ^{232}Th and 420 Bq/kg for ^{40}K . In Table 4, it is showed all the minimum, maximum and mean values of the soil samples. The world's average concentrations and hazard indexes are also given in Table [4.](#page-7-0) The mean values of these indexes were calculated as 66.516 *nGy/h*, 138.992 *Bq/kg*, 81.575 *μSv/y*, 0.451 *Bq/kg* and 0.375 *Bq/kg* for *D*, Ra_{eq} , $AEDE$, H_{in} and H_{ex} respectively. In Table [4,](#page-7-0) only the mean value of ${}^{40}K$'s concentration is higher than world's average. For the hazard indexes; absorbed dose rate in air *D* and annual effective dose equivalent *AEDE* are also higher than the values in world. Even if the values are slightly higher than the world's average, long term exposures may lead to series health risks including cancer. The effect of ionizing radiation shows itself at cellular level. Although the death of cells is the main effect of this kind of radiation, degeneration of nucleus or more specifically degeneration of the DNA is what causes the most critical harm. The imperfections, which are occurred during the process of repairing the DNA, may lead to various kind of cancer (U.N.S.C.E.A.R [2000\)](#page-10-0). Both internal and external hazard indexes $(H_{in}$ and $H_{ex})$ are below "1". Also, it must be mentioned that the activity concentrations of ¹³⁷*Cs* for sample 15 and 18 cannot calculated even if they are above *MDL*. Because the uncertainties of activity concentrations are greater than the activity itself.

Eke and Boztosun [\(2015\)](#page-9-3) and Eke et al. [\(2015\)](#page-9-29) studied different regions of Antalya for sand and soil samples. These works show the mean values are lower than Saklıkent region. Mean values for ^{238}U , ^{232}Th and ^{40}K from different mountains around the world and results of this study are given in Table [5.](#page-8-12) As shown in Table [5](#page-8-12), 238 U values are lower than other studies, ^{232}Th concentrations are only higher than Mountain Lebanon (Lebanon) and Marrah Mountain Range (Sudan). For the ^{40}K , activity concentrations of Bakırlıtepe are in the fourth place after Mountain Lebanon (Lebanon), Mount Karadağ (Turkey) and Marrah Mountain Range (Sudan).

** Due to low activity concentrations in Samples 15 and 18, they are not included to statistics

Due to low activity concentrations in Samples 15 and 18, they are not included to statistics

Study	$^{238}U(Bq/kg)$	$^{232}Th(Bq/kg)$	40K(Bq/kg)	Region
This Study	26.70	38.36	682.86	Bakırlıtepe, Beydağları Mountain Range, Turkey
Korkmaz et al. (2017)	71.6	83.9	451.1	Mount Karadağ, Turkey
Abdalhamid et al. (2017)	41	24	540	Marrah Mountain Range, Sudan
El Samad et al. (2018)	37	24	206	Mountain Lebanon, Lebanon
Otwoma et al. (2013)	195.3	409.5	915.6	Mount Homa, Kenya
Mitrović et al. (2016)	62	63	722	Gobelja Site, Mount Kopaonik, Serbia
Mitrović et al. (2016)	160	151	1291	Pancicey vrh Site, Mount Kopaonik, Serbia
Dzaluk et al. (2018)	31	41	900	Opava Mountains, Poland

Table 5 Comparison of mean values from different mountain sites in world

Conclusion

In this paper, we investigated the activity concentrations and hazard indexes of soil samples from Saklıkent region in Antalya, Turkey. "21" soil samples were investigated in total. These samples were collected from ∼ 100 *m* to ∼ 2000 *m* through Saklıkent, which is one of the highest mountain in Antalya. As a result, we have seen that there is not a uniform change in activity levels with altitude. It is also observed that the activity concentration of $40K$ is higher than world's average for this region. At this point, it must be indicated that the mountain is hosting many quarries. During and after the mining process, it is possible that many contaminant may spread to environment. The anomaly observed at higher altitudes may be the result of the stone mining (Shehzad et al. [2019\)](#page-10-6).

The absorbed dose rate in air and annual effective dose equivalent are also higher than world's mean values. When comparing the previous works of Eke and Boztosun [\(2015\)](#page-9-3) and Eke et al. [\(2015\)](#page-9-29), this study shows that the coast of Antalya is safer for human health than the inner parts of the city.

In U.N.S.C.E.A.R [\(2000\)](#page-10-0), it is reported that the igneous rocks are more enriched with ^{238}U and ^{232}Th , while sedimentary rocks have lower levels of radiation. Poisson et al. (2003) , Kosun (2012) , and Uner et al. (2018) showed that upper levels of Bakırlıtepe are formed mostly with sedimentary rocks (limestone etc.). So, our results are consistent with UNSCEAR report. In conclusion, this work will be a reference for upcoming years when the nuclear power plants are operational in Turkey.

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