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Determination of radioactivity levels and hazards of sediment and rock samples in İkizdere and Kaptanpaşa Valley, Turkey

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► ***Original article***

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ABSTRACT

Background: The radioactivity levels in sediment and rock samples of İkizdere and Kaptanpaşa Valley have been determined. To our knowledge, there seems to be no information about radioactivity level in the İkizdere and Kaptanpaşa Valley sediments and rocks so far. ***Materials and Methods:*** The average radium equivalent activity (Ra_{eq}), the total absorbed dose rate (D), the external risk index (H_{ex}) and the annual effective dose equivalent (AEDE) which will be defined later have been calculated and compared with the results in literature. Rock characterization was also investigated using thin section and X-ray diffraction (XRD) analysis. Also gross α and β activity concentrations were calculated for some rock samples. ***Results:*** Their radiological implications were also calculated and compared with the international recommended values. The gross α activity is generally lower than the corresponding gross β activity for some rock samples. Rock characterization was also investigated using thin section and XRD analysis. ***Conclusion:*** This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping. It seems necessary to determine the radioactivity concentrations in sediments and rock of other parts of Turkey. The results may also be used as a reference data for monitoring possible radioactivity pollutions in future.

Keywords: *Sediment, rock, natural radioactivity, İkizdere and Kaptanpaşa Valley, XRD.*

INTRODUCTION

The knowledge of radionuclides distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and extraterrestrial sources. Natural background radiation is of terrestrial and extraterrestrial origin. Terrestrial radiation is due to radioactive nuclides present in varying amounts in soils, building materials, water, rocks and atmosphere. Some of these radionuclides from these sources are transferred to man through food chain or inhalations, while the extraterrestrial radiation originates from outer space as primary cosmic rays [1-4].

In terms of natural radioactivity, granites exhibit an enhanced elemental concentration of uranium (U) and thorium (Th) compared to the very low abundance of these elements observed in the mantle and the crust of the Earth. Geologists provide an explanation of this behavior in the course of partial melting and fractional crystallization of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into the

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more silica-rich products. 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 to the Earth's crust (average 1.8 ppm for U and 7.2 ppm for Th) [1], the upper continental crust (average 2.7 ppm for U and 10.5 ppm for Th) [2] and rocks of basaltic or ultramafic composition (0.1 ppm of U and 0.2 ppm of Th) [1, 3-5].

İkizdere Valley is on the river bank, 56 km from the city of Rize on the road to Erzurum. It is discharged to the Eastern Black Sea near İyidere town in the west of Rize. İkizdere Valley stand between 40° 42′ 37″ north latitudes, 40° 36′ 55″ east longitudes. The Kaptanpaşa Valley originates from the slopes of Kackar Mountains and is discharged to the Eastern Black Sea, near Çayeli town in the east of Rize. Kaptanpaşa Valley stands between 40° 45′ 0″ north latitudes, 41° 4′ 0″ east longitudes. In the Valley, rainfall is mostly observed during winter and fall. These places, especially the parts having 1500 m height are covered with fog in almost every season of the year. The highest and lowest temperatures were recorded to be 38 and -7 °C, respectively, with the mean value of 15 °C. There are different rock structures having different ages and lithological features. As shown in figure 1, the study area are consists of mainly volcanic rocks such as basalts, andesite and granites [6].

The aim of this study is to determine natural (^{238}U, ^{232}Th, ^{40}K) and artificial (^{137}Cs) radioactivity levels in sediments and rocks collected from different points in İkizdere and Kaptanpaşa Valley in Rize province of Turkey. Also, Ra_{eq}, D, H_{ex}, and AEDE which will be defined later have been calculated and compared with the results in literature. Rock characterization was also investigated using thin section and XRD analysis. Also gross α and β activity concentrations were calculated for some rock samples. The results of this study will provide background data on the natural and artificial radioactive isotopes and environmental pollution.

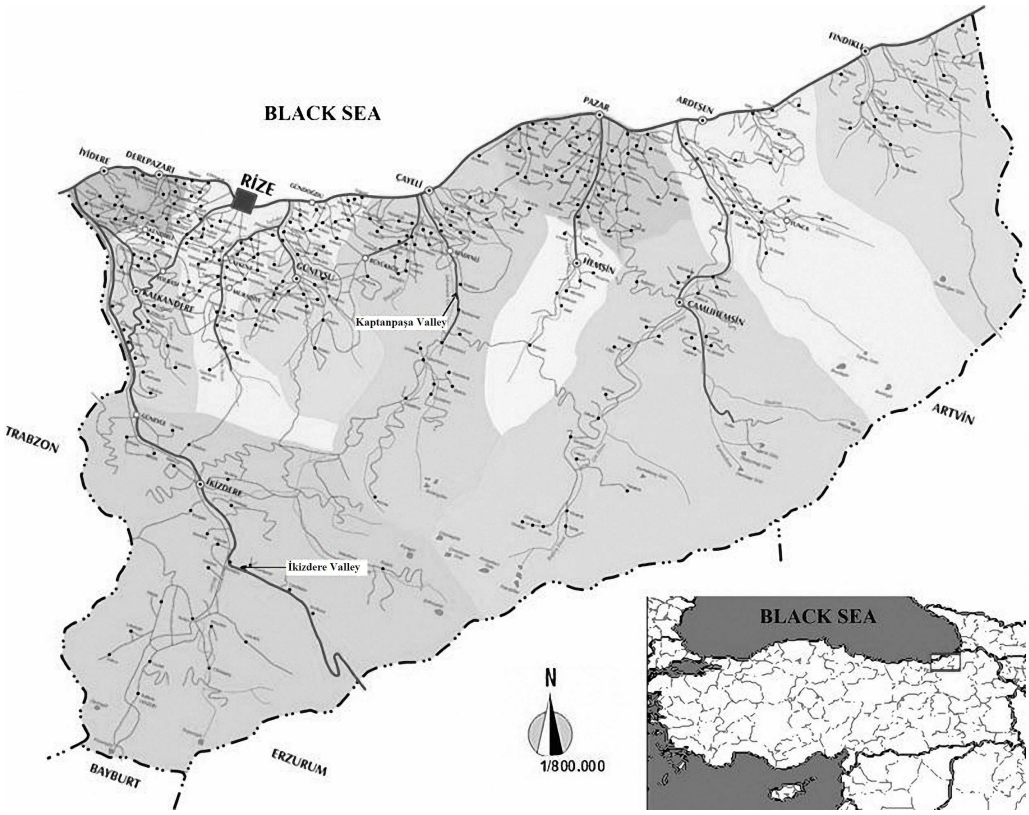


Figure 1. Location of sampling sites the Rize Province, Turkey.

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MATERIALS AND METHODS

Sample collection and preparation

In this study research area was divided into two regions İkizdere and Kaptanpaşa. The sampling stations are given in figure 1. The sediment (at 0-15 cm depth level) and rock samples were collected along the İkizdere and Kaptanpaşa Valley area in the summer of 2007. The sediment samples were taken just near the river bank of about 10–100 cm. The sediment samples were homogenized in situ, and this sand mixture, weighing approximately 20 kg, was considered representative of the sampling site. The samples were cleaned with warm water, dried in the laboratory. In the case of grain size study, samples were sieved after drying into five classes of particle size, namely <250, 250-300, 300-600, 600-850 and 850-1000 mm. The rock samples were taken from sampling locations as blocks cores. Eighteen rock samples were collected from a quarry factory available in İkizdere (Rock1-Rock6) and Kaptanpaşa (Rock 7 -Rock18) areas. All the rock samples are of Granite type. The samples, each about 1 kg in weight, were crushed, homogenized and sieved to about 100 mesh by a crushing machine. The samples were then placed for drying at 105 ^{0}C for 24 h to ensure that moisture is completely removed. About 250 g of sample material were used for the measurements of each granite sample.

The sediment and rock samples were then weighed and transferred to Marinelli beakers of 1000 ml volume. The samples were kept for 4 weeks before analysis under airtight conditions to allow secular equilibrium between thorium and radium and their decay products.

Rock characterization was also investigated using thin section and XRD analysis by X-ray diffraction study using XRD, RIGAKU D-MAX difractometer, CoKα.

Experimental method for γ- spectroscopy

The radiation levels of samples were analyzed using gamma spectrometry, which was equipped with a 55% efficiency high purity germanium (HPGe) detector and a multi-channel analyzer. The gamma spectra were analyzed by using the ORTEC Maestro 32 data acquisition and analysis system. The detector had coaxial closed–facing geometry with the following specifications: resolution full width half maximum (FWHM) at 122 keV ^{57}Co was 1.00 keV and at 1.33MeV ^{60}Co was 1.90 keV. The detector was shielded by a cylindrical lead shield, which had average thickness of 10 cm to reduce the background due to the cosmic rays and the radiation nearby the system. Efficiency of the detector was determined with a ^{152}Eu source of known activity. ^{152}Eu source has been widely used for calibration and efficiency determination due to their large range of energies (122, 244, 344, 411, 443, 779, 964, 1112 and 1408 keV) with emission probabilities of 3-29 % [7, 8]. An ideal measuring geometry of cylindrical source (homogeneously distributed activity with constant volume and distance) was placed coaxially with the detector for the efficiency determination and the same procedure applied for the sample measurements.

The samples were sealed and stored in a tight container to prevent the escape of radiogenic gases ^{222}Rn and ^{220}Rn, and to allow the attainment of radioactive equilibrium in the decay chain. After attainment of secular equilibrium between ^{232}Th, ^{238}U and their daughter products, the samples were subjected to gamma -ray spectrometric analysis. Natural radionuclides of relevance for this work are mainly γ-ray emitting nuclei in the decay series of ^{232}Th and ^{238}U, and single occurring ^{40}K. While ^{40}K can be measured directly by its own γ-rays, ^{232}Th and ^{238}U are not directly γ-ray emitters, but it is possible to measure γ-rays of their decay products. Decay products for ^{238}U (^{214}Pb: 295 and 352 keV; and ^{214}Bi: 609 keV) and ^{232}Th (^{228}Ac: 338, and 911 keV; ^{212}Pb: 239 keV; ^{212}Bi: 727 keV; and ^{208}Tl: 583 keV) were used by assuming the decay series to be in secular equilibrium [7]. Weighted averages of several decay products were used to estimate activity concentrations of ^{238}U and ^{232}Th. The natural abundance of ^{235}U is only 0.72% of the total uranium content and hence was not considered in the present study. The activity concentrations of 40K were measured directly by its own gamma rays (1460.8 keV). The 661.6 keV gamma transition was used to determine the

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^{137}Cs concentration. The minimum detectable activity (MDA) of the system for each radionuclide was calculated as 0.16 Bq kg^{-1} for ^{137}Cs, 3.25 Bq kg^{-1} for ^{40}K, 0.10 Bq kg^{-1} for ^{214}Pb, 0.13 Bq kg^{-1} for ^{214}Bi and 0.11 Bq kg^{-1} for ^{228}Ac.

Measurements of gross α and β activities

Measurements of radioactivity level in all rock samples were performed the gross α and gross β counting system. The device used to count the α and β activities, was an α/β counter of the low background multiple detector type (Berthold LB770). The sample detectors are gas flow window-type counters which are approximately 5 cm in diameter. The counting gas was a mixture of 90% argon and 10% methane. All samples were placed in a 5 cm diameter stainless-steel planchet for counting. Lead shielding was used to attenuate external radiation. The operating voltage on the detector was selected to be 1,650 V.

The system was calibrated for α and β energies by preparing standard samples which contain equal concentrations. ^{241}Am (913 Bq) and ^{90}Sr (931 Bq) were used to calibrate the system for σ and β energies, respectively. The counting efficiencies for the system are 19–21% for α and 71–73% for β.

Background and efficiency data for the detector were collected, stored and used for corrections. The background of the detector was determined with measurements for the length of time that routine samples were counted and were measured using a clean, empty planchet in detector. The repetitive determination of backgrounds served as a check on the operation of the system, with average values of 0.0015 and 0.02 cpm background counting rate for α and β, respectively.

RESULTS

The activity concentrations of ^{232}Th, ^{238}U, ^{40}K and ^{137}Cs

An attempt was made to study the variation in radioactivity associated with grain size of the İkizdere and Kaptanpaşa sediment samples and the results summarized in tables 1 and 2, respectively. It was found that higher activity was associated with the finest size fraction viz. <250 μm at all depths in the case of ^{232}Th. The activity concentration of ^{238}U was nearly independent of the grain size with a somewhat higher activity concentration in the 850-600 μm grain size fraction for İkizdere sediments and in the <250 μm grain size fraction for Kaptanpaşa sediments. The lowest activity of ^{40}K was found in the 300-250 μm particle size group. There is a tendency of ^{137}Cs radionuclide to decrease in concentration with decreasing particle size. The activity is proportional to particle size.

The activity in İkizdere sediment samples range from 28.98±2.92 to 35.01±3.52 Bqkg^{-1} for ^{232}Th, 120.64±6.89 to 144.29±7.6 Bqkg^{-1} for ^{238}U, 705.76±31.23 to 910.56±42.79 Bqkg^{-1} for ^{40}K and 3.83±0.09 to 6.45±0.16 for ^{137}Cs. The overall mean activity levels for ^{232}Th, ^{238}U, ^{40}K and ^{137}Cs are 32.71±3.27, 134.12±7.34, 811.68±34.53 and 5.25±0.13 Bqkg^{-1} (dry weight), respectively. The activity in Kaptanpaşa sediment samples range from 20.42±2.02 to 28.31±2.81 Bqkg^{-1} for ^{232}Th, 15.31±1.51 to 22.38±2.18 Bqkg^{-1} for ^{238}U, 484.3±23.61 to 714.39±35.16 Bqkg^{-1} for ^{40}K and 3.8±0.10 to 6.9±0.18 for ^{137}Cs. The overall mean activity levels for ^{232}Th, ^{238}U, ^{40}K and ^{137}Cs are 24.48±2.43, 19.46±1.91, 609.66±27.90 and 5.13±0.13 Bqkg^{-1} (dry weight) in Kaptanpaşa sediment samples, respectively.

The mean activity concentrations in sediment samples 5.31±0.4 Bqkg^{-1}, 34.04±1.4 Bqkg^{-1} and 401.11±24.3 Bqkg^{-1} for ^{238}U, ^{232}Th and ^{40}K, respectively [9]. The worldwide concentrations of the radionuclides ^{232}Th, ^{238}U and ^{40}K have averages in sediment samples of 40, 40, and 580 Bqkg^{-1}, respectively [10]. ^{137}Cs does not exist in sediment naturally. It is a product of fallout radioactivity. The ^{137}Cs might have been deposited in sediment of İkizdere and Kaptanpaşa Valley, presumably as a result of the nuclear power plant accident at Chernobyl on 26 April 1986. Moreover, measured ^{137}Cs activity concentrations can be attributed to the atmospheric nuclear weapon tests conducted by several countries.

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Table 1. Radioactivity concentrations of ^{232}Th, ^{238U}, ^{40}K and ^{137}Cs, as well as the calculated absorbed dose rates (D), Ra equivalent (Ra_{eq}), external hazard index (H_{ex}) and annual effective dose rates (AEDE) in sediment samples at İkizdere Valley.

Region	Grain Size	^{232}Th (Bqkg^{-1})	^{238}U (Bqkg^{-1})	^{40}K (Bqkg^{-1})	^{137}Cs (Bqkg^{-1})	D (nGyh^{-1})	Ra_{eq} (Bqkg^{-1})	H_{ex}	AEDE (μSvy^{-1})
İkizdere1	1000-850	23.74±2.30	93.60±5.29	560.21±25.18	6.79±0.18	79.88	558.91	0.46	97.97
İkizdere2	1000-850	56.45±5.81	215.72±13.38	1280.98±63.87	6.34±0.15	184.82	1282.80	1.07	226.66
İkizdere3	1000-850	23.82±2.46	197.19±10.19	1355.32±61.80	5.90±0.14	158.52	1274.85	0.91	194.41
İkizdere4	1000-850	11.91±1.12	68.39±3.62	445.74±20.32	6.80±0.17	56.34	428.64	0.32	69.10
	Mean	28.98±2.92	143.73±8.12	910.56±42.79	6.45±0.16	119.89	886.30	0.69	147.04
İkizdere1	850-600	25.71±2.48	92.98±5.79	543.13±28.00	6.21±0.16	80.19	547.96	0.46	98.34
İkizdere2	850-600	62.34±5.96	205.93±12.18	1167.93±54.27	5.86±0.16	179.66	1194.38	1.04	220.33
İkizdere3	850-600	26.45±2.72	192.12±8.05	1299.44±43.76	5.36±0.15	155.68	1230.51	0.89	190.93
İkizdere4	850-600	14.04±1.44	86.13±4.37	567.98±24.03	6.61±0.15	70.61	543.55	0.41	86.59
	Mean	32.13±3.15	144.29±7.6	894.62±37.51	6.01±0.16	121.53	879.09	0.70	149.04
İkizdere1	600-300	28.10±2.85	88.02±4.72	489.45±17.47	5.45±0.13	77.33	505.08	0.45	94.84
İkizdere2	600-300	68.53±7.06	216.24±12.78	1200.51±50.70	5.34±0.12	189.56	1238.63	1.10	232.48
İkizdere3	600-300	26.94±2.70	174.25±7.86	1158.55±42.36	4.92±0.12	142.29	1104.86	0.82	174.50
İkizdere4	600-300	14.01±1.40	72.35±4.22	461.67±22.97	5.32±0.12	60.11	447.87	0.35	73.72
	Mean	34.40±3.50	137.72±7.40	827.55±33.38	5.26±0.12	117.33	824.13	0.68	143.89
İkizdere1	300-250	27.12±2.74	76.01±4.26	401.73±14.37	4.91±0.12	67.76	424.12	0.39	83.11
İkizdere2	300-250	65.14±6.18	183.02±10.08	971.32±36.41	4.60±0.11	163.23	1024.09	0.95	200.19
İkizdere3	300-250	25.25±2.61	163.49±10.15	1086.44±60.55	4.70±0.13	133.46	1036.16	0.77	163.68
İkizdere4	300-250	14.63±1.49	60.03±3.05	363.56±13.58	4.59±0.11	51.02	360.89	0.29	62.58
	Mean	33.04±3.25	120.64±6.89	705.76±31.23	4.70±0.12	103.87	711.32	0.60	127.39
İkizdere1	<250	29.66±3.04	76.13±4.06	385.80±10.86	4.11±0.09	68.81	415.61	0.40	84.39
İkizdere2	<250	66.51±6.51	212.06±12.01	1183.80±48.65	3.77±0.09	185.72	1218.70	1.08	227.77
İkizdere3	<250	28.25±2.96	153.29±7.30	988.24±36.19	3.44±0.08	126.85	954.63	0.73	155.57
İkizdere4	<250	15.62±1.57	55.37±3.34	321.82±15.17	4.01±0.10	47.89	325.51	0.28	58.73
	Mean	35.01±3.52	124.21±6.68	719.91±27.72	3.83±0.09	107.31	728.61	0.62	131.61
All Mean		32.71±3.27	134.12±7.34	811.68±34.53	5.25±0.13	113.99	805.89	0.66	139.79

The activity of the ^{232}Th, ^{238}U and ^{40}K in the rock samples collected from a quarry factory available in İkizdere and Kaptanpaşa area is shown in table 3. The activity concentrations of rock samples range from 26.12±3.24 to 44.39±4.41 Bqkg^{-1} for ^{232}Th, 13.80±1.91 to 29.25±5.04 Bqkg^{-1} for ^{238}U and 307.54±36.10 to 539.15±22.54 Bqkg^{-1} for ^{40}K. The overall mean activity levels for ^{232}Th, ^{238}U and ^{40}K are 35.06±3.85, 19.71±2.15 and 386.16±27.38 Bqkg^{-1} (dry weight) in rock samples, respectively.

The variations of natural radioactivity levels at different sampling sites are due to the variation of concentrations of these elements in the geological formations. The concentrations of the above mentioned radionuclides are compared with that of other author's for sediment and rock in tables 4 and 5, respectively. As shown in table 4, among the max ^{238}U concentrations for İkizdere, our value is higher than the others except Spain and for Kaptanpaşa, our value is similar to Algeria. Among the min ^{232}Th concentrations, our value is similar to the others except Portugal. Among the max ^{40}K concentrations, while our value is similar to Greece, it is higher than the others.

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Table 2. Radioactivity concentrations of ^{232}Th, ^{238}U, ^{40}K and ^{137}Cs, as well as the calculated absorbed dose rates (D), Ra equivalent (Ra_{eq}), external hazard index (H_{ex}) and annual effective dose rates (AEDE)in sediment samples at Kaptanpaşa Valley.

Region	Grain Size	^{232}Th (Bqkg^{-1})	^{238}U (Bqkg^{-1})	^{40}K (Bqkg^{-1})	^{137}Cs (Bqkg^{-1})	D (nGyh^{-1})	Ra_{eq} (Bqkg^{-1})	H_{ex}	AEDE (μSvy^{-1})
Kaptanpaşa1	1000-850	12.8±1.22	8.95±0.95	561.04±23.24	6.54±0.16	36.53	459.25	0.19	44.80
Kaptanpaşa2	1000-850	27.7±2.74	18.25±1.69	567.78±28.19	6.05±0.14	50.66	495.05	0.27	62.13
Kaptanpaşa3	1000-850	31.77±3.23	22.17±2.17	1000.3±40.02	7.14±0.16	73.71	837.82	0.39	90.40
Kaptanpaşa4	1000-850	14.92±1.47	14.41±1.48	714.61±25.42	7.09±0.20	46.90	586.00	0.25	57.52
Kaptanpaşa5	1000-850	14.94±1.45	12.78±1.26	588.58±25.96	6.9±0.18	40.77	487.35	0.21	50.01
	Mean	20.42±2.02	15.31±1.51	686.46±28.57	6.74±0.17	49.71	573.08	0.26	60.96
Kaptanpaşa1	850-600	14.89±1.45	15.61±1.56	530.35±23.90	5.59±0.15	39.43	445.27	0.21	48.36
Kaptanpaşa2	850-600	33.53±3.42	22.11±2.10	664.17±37.05	5.51±0.14	60.33	581.47	0.33	73.99
Kaptanpaşa3	850-600	35.44±3.58	27.15±2.64	1036.3±52.54	5.38±0.13	79.82	875.81	0.43	97.90
Kaptanpaşa4	850-600	14.61±1.43	15.43±1.53	703.75±28.44	5.37±0.13	46.66	578.21	0.24	57.23
Kaptanpaşa5	850-600	18.2±1.81	16.6±1.62	637.36±33.88	5.39±0.15	46.67	533.39	0.25	57.24
	Mean	23.34±2.34	19.38±1.89	714.39±35.16	5.45±0.14	54.59	602.84	0.29	66.95
Kaptanpaşa1	600-300	16.49±1.61	16.93±1.71	488.58±27.87	5.26±0.14	39.25	416.72	0.21	48.14
Kaptanpaşa2	600-300	38.17±3.73	23.63±2.35	534.97±23.58	5.17±0.15	58.47	490.14	0.32	71.71
Kaptanpaşa3	600-300	38.06±3.84	26.98±2.55	1003.4±52.25	4.92±0.14	80.06	853.99	0.43	98.19
Kaptanpaşa4	600-300	17.34±1.76	16.35±1.63	585.98±27.84	4.94±0.13	43.77	492.35	0.23	53.69
Kaptanpaşa5	600-300	18.38±1.75	18.46±1.67	609.48±25.68	5.19±0.11	46.38	514.04	0.25	56.88
	Mean	25.69±2.54	20.47±1.98	644.47±31.44	5.09±0.13	53.59	553.45	0.29	65.72
Kaptanpaşa1	300-250	20.51±2.05	14.88±1.56	361.46±15.64	5.06±0.12	35.55	322.53	0.19	43.59
Kaptanpaşa2	300-250	35.02±3.42	22.9±2.26	515.93±25.73	4.29±0.11	55.25	470.24	0.30	67.76
Kaptanpaşa3	300-250	34.05±3.31	28.14±2.97	781.21±31.36	4.6±0.11	68.31	678.36	0.37	83.77
Kaptanpaşa4	300-250	16.59±1.73	15.55±1.58	375.61±19.20	4.21±0.11	33.85	328.49	0.18	41.51
Kaptanpaşa5	300-250	17.13±1.71	17.2±1.66	387.26±26.11	4.58±0.11	35.41	339.89	0.19	43.43
	Mean	24.66±2.44	19.73±2.01	484.3±23.61	4.55±0.11	45.67	427.90	0.25	56.01
Kaptanpaşa1	<250	23.6±2.30	19.67±1.95	378.04±13.10	3.78±0.09	40.35	344.51	0.22	49.49
Kaptanpaşa2	<250	40.8±4.07	26.86±2.63	576.09±17.96	3.85±0.09	63.37	528.79	0.35	77.71
Kaptanpaşa3	<250	39.89±3.95	29.69±2.75	830.25±37.44	3.94±0.11	74.95	726.03	0.41	91.92
Kaptanpaşa4	<250	17.94±1.81	18.79±1.89	360.9±15.80	3.78±0.11	35.49	322.34	0.20	43.53
Kaptanpaşa5	<250	19.32±1.93	16.9±1.67	448.09±19.22	3.68±0.10	39.36	389.56	0.21	48.28
	Mean	28.31±2.81	22.38±2.18	518.67±20.70	3.8±0.10	50.70	462.24	0.28	62.18
All Mean		24.48±2.43	19.46±1.91	609.66±27.90	5.13±0.13	50.85	523.90	0.27	62.37

Table 5 shows a comparison of radioactivity levels in rocks with other areas of the world. As shown in Table 5, among the ^{238}U concentrations, while our average value is higher than ^{238}U concentrations of India, is lower than average ^{238}U concentrations of Egypt, Brazil, Cyprus, China and Turkey (Eskişehir). Among the ^{232}Th concentrations, while our value is lower than Egypt, Brazil, Cyprus and Turkey (Eskişehir), it is higher than China and India. For average ^{40}K concentrations, while our value is higher than that of India, it is lower than the others.

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Table 3. Radioactivity concentrations of ^{232}Th, ^{238U} and ^{40}K, as well as the calculated absorbed dose rates (D), Ra equivalent (Ra_{eq}), external hazard index (H_{ex}) and annual effective dose rates (AEDE) in rock samples at İkizdere and Kaptanpaşa Valley.

Sample ID	^{232}Th (Bqkg^{-1})	^{238}U (Bqkg^{-1})	^{40}K (Bqkg^{-1})	D (nGyh^{-1})	Ra_{eq} (Bqkg^{-1})	H_{ex}	AEDE (µSvy^{-1})
Rock 1	40.25±3.51	20.20±2.20	539.15±22.54	58.56	119.27	0.32	71.82
Rock 2	42.72±3.71	23.91±2.96	312.03±39.69	51.97	109.03	0.29	63.74
Rock 3	40.56±4.36	27.50±3.91	336.24±44.31	53.12	111.39	0.30	65.14
Rock 4	44.39±4.41	29.25±5.04	313.80±25.82	55.43	116.89	0.32	67.98
Rock 5	33.68±4.25	17.33±2.04	518.40±71.58	52.09	105.41	0.28	63.88
Rock 6	36.00±4.17	18.69±1.65	337.88±47.68	46.41	96.19	0.26	56.92
Rock 7	35.98±4.16	18.00±2.02	315.99±31.93	45.16	93.78	0.25	55.38
Rock 8	38.18±4.70	20.18±2.41	414.23±35.62	51.79	106.67	0.29	63.51
Rock 9	37.95±3.72	20.35±2.45	433.68±32.14	52.55	108.01	0.29	64.44
Rock 10	41.54±3.95	21.43±2.58	491.24±35.20	57.87	118.66	0.32	70.97
Rock 11	33.06±2.92	15.78±1.90	369.07±15.17	44.57	91.47	0.25	54.66
Rock 12	35.32±3.12	18.28±2.20	483.78±42.69	52.09	106.04	0.29	63.88
Rock 13	26.68±3.92	18.61±2.57	362.13±30.18	41.25	84.65	0.23	50.59
Rock 14	28.88±4.27	20.48±2.83	441.40±47.45	46.93	95.77	0.26	57.56
Rock 15	26.12±3.24	13.80±1.91	307.54±36.10	36.47	74.83	0.20	44.73
Rock 16	27.93±3.48	15.66±2.16	310.51±38.47	38.59	79.51	0.21	47.33
Rock 17	28.71±3.58	16.86±2.33	325.90±42.95	40.28	83.01	0.22	49.40
Rock 18	33.11±3.76	18.40±2.54	337.88±47.68	44.37	91.76	0.25	54.42
Mean	35.06±3.85	19.71±2.15	386.16±27.38	48.31	99.57	0.27	59.24

Table 4. Comparison of radioactivity of sediment samples with other areas of the world.

Country	^{238}U (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	References
India	1.2-21.4	6.3-224.7	178.1-1698	Musugesan *et al.* [9]
Albania	8–27	13–40	266–675	Tsabaris *et al.* [24]
Spain	77–6401	12–63	-	Lozano *et al.* [25]
Spain	-	11–16	220–460	Ligero *et al.* [26]
Algeria	11–25	6–32	56–607	Benamar *et al.* [27]
Egypt	-	8–50	16–487	Ibrahiem *et al.* [28]
Portugal, River Tejo	-	54–76	-	Carreira and Sequeira [29]
Greece	29–110	19–88	152–1593	Florou and Kriditis [30]
Italy	42–70	31–37	410–475	Doretti *et al.* [31]
Turkey (Fırtına River)	16-113	17-87	51-1605	Kurnaz *et al.* [32]
Turkey (İkizdere Valley)	124.21	32.71	811.68	Present study
Turkey (Kaptanpaşa Valley)	19.46	24.48	609.66	Present study

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Table 5. Comparison of radioactivity of rock samples with other areas of the world.

Country	^{238}U (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	References
Egypt(Wadi Karim)	14-227	10.5-183	2299-7356	El Arabi [16]
Egypt(Um Taghir)	2.9-9087	1.4-3834	132-10230	El Arabi [16]
Brazil	31	73	1648	Anjos *et al.* [31]
Cyprus	1-588	1-906	50-1606	Tzortzis *et al.* [32]
China	20.4	30.1	1009.5	Lu and Zhang [15]
India (Kakrapar)	9.1-24.1	3.7-12.7	101.8-264.1	Patra *et al.* [20]
Turkey (Eskişehir)	43-651	51-351	418-1618	Orgun *et al.* [13]
Turkey (İkizdere-Kaptanpaşa)	19.71	35.06	386.16	Present study

Absorbed dose rate in air (D)

The contribution of natural radionuclides to the absorbed dose rate in air (D) depends on the natural specific activity concentration of ^{238}U, ^{232}Th and ^{40}K. The greatest part of the gamma radiation comes from terrestrial radionuclides. There is a direct connection between terrestrial gamma radiation and radionuclide concentrations. If a radionuclide activity is known then its exposure dose rate in air at 1m above the ground can be calculated using the formula proposed by Beck [11] and UNSCEAR [12]:

$$D\ (nGy/h) = 0.427A_U + 0.662A_{Th} + 0.0432A_K \quad (1)$$

where D is the dose rate at 1m above the ground, A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively, in the samples. The absorbed dose rates in air for the areas under investigation are listed in Tables 1and 2 for sediments and in Table 3 for rocks samples. The absorbed dose rates in air for sediment samples the average dose rate was 113.99 nGyh^{-1} and 50.85 nGyh^{-1} at İkizdere and Kaptanpaşa Valley, respectively.. The average D value for sediments was calculated as 96.10,124, 167, 67.2 nGyh^{-1} in Cauvery River, Tamilnadu, India [9], in the Xiazhuang Granite area [13], in Southeast part of Eskisehir[14], in Fırtına valley [15], respectively. The population weighted values give an average absorbed dose rate in air outdoors from terrestrial gamma radiation of 55 nGyh^{-1}. This reveals that the mean absorbed dose rate in air outdoors from İkizdere areas is almost two times higher than that of the world-wide average value. For rock samples, the average absorbed dose rate was 48.31 nGyh^{-1} which is lower than China [16], in Um Taghir and Wadi Karim of Egypt [17] and the international recommended value (55 nGyh^{-1}) [12].

Radium equivalent activity (Ra_{eq}) and external hazard index (H_{ex})

The results were evaluated in terms of the radiation risk by means of the Ra_{eq} and H_{ex}. Radium equivalent activity is a widely used D risk index and it is calculated through the relation given by Beretka and Mathew [18]. It is assumed that 370 Bqkg^{-1} of ^{226}Ra, 259 Bqkg^{-1} of ^{232}Th and ^{4810} Bqkg^{-1} of ^{40}K produce the same gamma-ray dose rate

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively. Ra_{eq} is estimated for the collected samples and are given in Tables 1 and 2 for sediments and in Table 3 for rocks. The average values of Ra_{eq} were found to be 805.89 and 535.90 Bqkg^{-1} in İkizdere and Kaptanpaşa sediment samples, respectively. The average Ra_{eq} value for sediments was calculated as 166.3 Bqkg^{-1} in the Fırtına valley [15]. This value is lower than our study.

For rock samples, the average absorbed dose rate was 99.57 Bqkg^{-1}. The estimated average values of Ra_{eq} in the present work are lower than the measured mean values from some of the other countries and the recommended maximum value of 370 Bqkg^{-1}[18]. It is observed that value of this work is lower than the

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measured values of 493.8 Bqkg^{-1} in Eastern Desert of Egypt [19], 366.9 Bqkg^{-1} in southeast part of Eskisehir (Turkey) [14] and 266 Bqkg^{-1} in Xiazhuang Granite area (China) [13].

The external risk index was calculated for the investigated samples using the model proposed by Krieger [20] assuming thick walls without windows and doors, where the external risk index is given by

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively. The results of H_{ex} based on the criterion formula (Eq. (3)) are given in tables 1 and 2 for sediments and in Table 3 for rocks. The average values of H_{ex} were found to be 0.66 and 0.27 in İkizdere and Kaptanpaşa sediment samples, respectively. Only four value estimated of H_{ex} in the present work are higher than 1 for İkizdere Valley. The results range from 0.20 to 0.32 and average value was found to be 0.27 for the rock samples. The average values of H_{ex} were found to be 0.22 for India [9], 0.12 for Kakrapar [21], 0.45 for Fırtına valley of Turkey [15], 0.99 for Southeast part of Eskisehir (Turkey) [14] and 0.84 for Xiazhuang Granite area (China) [13].

Annual effective dose equivalent (AEDE)

In order to estimate the annual effective doses, one has to take into account to conversion coefficient from absorbed dose in air to effective and the outdoor occupancy factor. In the UNSCEAR[23] reports, a value of 0.7 SvGy^{-1} was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.2 for the outdoor occupancy factor. The annual effective dose equivalent was calculated from following equation:

$$\text{AEDE } (\mu\text{Svy}^{-1}) = D\ (\text{nGyh}^{-1}) \times 8760\ \text{hy}^{-1} \times 0.2 \times 0.7\ \text{SvGy}^{-1} \times 10^{-3} \quad (5)$$

The results of the calculation are given in Tables 1 and 2 for sediments and in Table 3 for rocks. The average values of AEDE were found to be 139.79 μSvy^{-1} and 62.37 μSvy^{-1} in İkizdere and Kaptanpaşa sediment samples, respectively. The results range from 44.73 to 71.82 and average value was found to be 59.24 μSvy^{-1} for the rock samples. The world average annual effective dose equivalent (AEDE) from outdoor terrestrial gamma radiation is 70 μSvy^{-1} [12]. So, the obtained value for İkizdere sediment samples is higher than the world average value. The average AEDE value was calculated to be 152 μSvy^{-1} in the Xiazhuang Granite area (China) [13], 314.1 μSvy^{-1} in the Southeast part of Eskisehir (Turkey) [14]. These average values are generally higher than our result.

Gross α and β activities in Rize rock

Total gross α and gross β activity concentrations in some rock samples are given in table 6. The gross α activity is generally lower than the corresponding gross β activity. The gross α activities range between a minimum of 100.72 ± 7.32 Bqkg^{-1} and a maximum of 932.57 ±65.72 Bqkg^{-1}. The gross β activities range between 171.35 ± 11.38 and 1269.01 ± 78.33 Bqkg^{-1}. An enhancement of gross α activity was observed in some region, especially in samples from Rock10 and Rock18. This can be explained by the radioactive material from granite rocks in these regions. Similarly, gross beta activity also increased in Rock18 of the region.

Thin section analysis

The thin section analysis of the granite samples, collected from the study area, shows that they are granodiorite and the following minerals were observed. The Asniyor leucogranite has equigranular to porphyritic textures consisting of quartz, orthoclase, plagioclase and chloritized biotite (which are scarce in the modal mineralogical composition). All these minerals include some zircon grains. There is widespread hydrothermal alteration in the Asniyor leucogranite which resulted in alteration of

Table 6. Gross α and β activity concentration of some rock samples.

Sample ID	Gross α (Bqkg^{-1})	Gross β(Bqkg^{-1})
Rock1	394.12±27.13	243.76±15.43
Rock3	276.25±19.21	419.24±26.37
Rock6	131.65±9.09	302.18±19.87
Rock7	245.38±17.18	415.22±26.68
Rock10	432.45±30.22	502.18±31.49
Rock13	100.72±7.32	252.79±16.55
Rock15	306.16±21.56	171.35±11.38
Rock18	932.57±65.72	1269.01±78.33

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feldspars to kaolinite and secirite. Although most of the samples are granite in composition, some of them are tonalite. Figure 2 shows the thin section analysis of the granite sample.

XRD Analysis

The XRD analysis was carried out on all of the rock samples. Figure 3 shows XRD analysis of the Rock 11. Although there is not much Zr in the sample, some of the zircon peaks were observed under the background, and some of them overlap with albite and quartz peaks. Thorite and baddeleyite compounds and zircon element were observed with fewer amounts in the rock sample. Also, the thin section investigation of the granite samples supports the presence of zircons in the sample (figure 2).

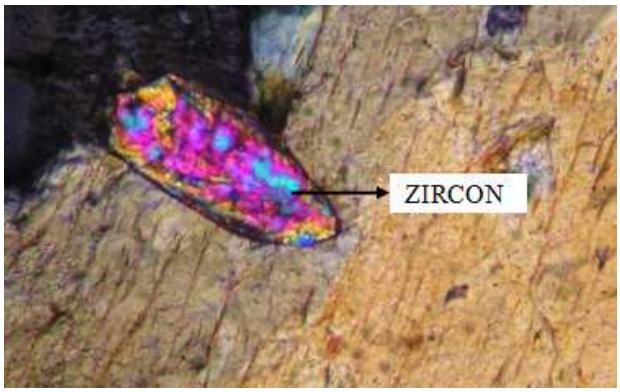


Figure 2. A zircon mineral in the granite sample collected from studying area.

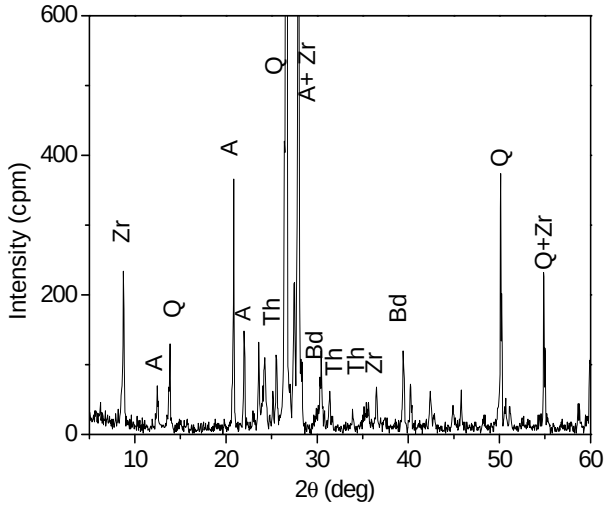


Figure 3. XRD analysis of the Rock 11 sample: (Q quartz, A albite, Zr zircon, Th thorite, Bd baddeleyite).

CONCLUSION

The mean concentrations of the radionuclides 238U, 232Th, 137Cs, and 40K in sediment and rock samples collected from different points in İkizdere and Kaptanpaşa Valley in Rize province of Turkey determined in this study compare suitably with literature values. But the ^{137}Cs activity concentrations in some places are higher than the other results. This can be attributed to the Chernobyl nuclear power plant accident and the atmospheric nuclear weapon tests conducted by several countries. Health effect due to natural radiation from sediment and rock of the İkizdere and Kaptanpaşa Valley are low and thus, health hazards are insignificant.

Total gross α and gross β activity concentrations in some rock samples were determined. The gross α activity is generally lower than the corresponding gross β activity. Rock characterization was also investigated using thin section and XRD analysis.

This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping. It seems necessary to determine the radioactivity concentrations in sediments and rock of other parts of Turkey. The results may also be used as a reference data for monitoring possible radioactivity pollutions in future.

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