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Natural radioactivity analysis in soil samples of Ardahan province, Turkey for the assessment of the average effective dose

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ABSTRACT

In this study, the distribution of natural radionuclides in surface soils in Ardahan province has been determined. The activity concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in 35 soil samples collected from the Ardahan city and its surrounded districts were measured through NaI (Tl) gamma spectrometry. The average activities of the samples were determined to be 29.9 ± 6.2 , 36.7 ± 6.8 and 435.1 ± 23.9 Bq kg^{-1} for the natural radionuclides ^{238}U , ^{232}Th and ^{40}K , respectively. For the fission product ^{137}Cs , the average activity of the samples was assigned to be 15.5 ± 0.8 Bq kg^{-1} . The average outdoor gamma dose rate (terrestrial and cosmic) and also the average annual effective dose rate in air due to the presence of radionuclides in 35 soil samples were obtained as 56.3 ± 8.7 nGy h^{-1} and 69.0 ± 6.7 $\mu\text{Sv y}^{-1}$, respectively. This research showed a clear correlation between the altitude and the natural radionuclides. The results obtained for Ardahan city centrum and its districts were presented graphically and compared with the world average values and those of Turkey.

Keywords: Ardahan province, natural radioactivity, soil, gamma-ray spectrometry, altitude.

Ardahan ilinin (Türkiye) toprak örneklerinde ortalama etkin dozun değerlendirilmesi için doğal radyoaktivite analizi

ÖZ

Bu çalışmada Ardahan ili yüzey topraklarında doğal radyonüklidlerin dağılımı belirlenmiştir. Ardahan ili ve çevre ilçelerinden toplanan 35 adet toprak örneğinin ^{238}U , ^{232}Th , ^{40}K ve ^{137}Cs aktivite konsantrasyonları, NaI(Tl) gama spektrometresi ile ölçülmüştür. Toprak numunelerindeki ^{238}U , ^{232}Th ve ^{40}K doğal radyonüklitlerinin ortalama aktiviteleri sırasıyla, 29.9 ± 6.2 , 36.7 ± 6.8 ve 435.1 ± 23.9 Bq kg^{-1} olarak tespit edildi. Fisyon ürünü ^{137}Cs için, numunelerin ortalama aktivitesi 15.5 ± 0.8 Bq kg^{-1} olarak belirlenmiştir. 35 adet toprak numunesinde radyonüklidlerin varlığına bağlı olarak ortalama açık hava gama doz oranı (karasal ve kozmik) ve havadaki ortalama yıllık etkili doz oranı sırasıyla, 56.3 ± 8.7 nGy h^{-1} ve 69.0 ± 6.7 $\mu\text{Sv y}^{-1}$ olarak bulunmuştur. Bu araştırma, rakım ile doğal radyonüklidler arasında net bir korelasyon olduğunu gösterdi. Ardahan şehir merkezi ve ilçeleri için elde edilen sonuçlar, grafiksel olarak sunulmuş ve Türkiye ortalamaları ve dünya ortalamaları ile karşılaştırılmıştır.

Anahtar Kelimeler: Ardahan ili, doğal radyoaktivite, toprak, gama spektrometresi, rakım.

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1. INTRODUCTION

Human beings are subjected to radiations stemming from both the natural and artificial sources in their living environments. It is of great importance to know the natural radioactivity level in any region not only for letting people recognize the natural radioactivity levels of their living area, but also for detecting any possible variations in the radioactivity levels. Major contributions to environmental radiation are caused by radioactive elements such as ^{238}U , ^{232}Th and ^{40}K . The concentrations of uranium and thorium in soils are high in localized areas and cosmic rays are more intense at higher altitudes [1]. Natural environmental radioactivity and the associated outside exposure on account of gamma radiation depend largely on the geological and geographical circumstances, and seem at different levels in the soils of any domain in the world [2]. Therefore, many scientists around the world work on determining the terrestrial radiation from the radionuclides in the soil and investigating the effects of radiation on biological systems. The intention of this study was to ascertain the concentration of natural radionuclides, such as ^{238}U , ^{232}Th , ^{40}K and artificial radionuclides ^{137}Cs radioactivity in 35 soil samples collected from Ardahan Province for health risk assessment.

2. MATERIALS AND METHODS

2.1 Survey Area

In this study, the measurement fields as a center of Ardahan, Çıldır, Damal, Göle, Hanak, and Posof and also the surroundings have been selected. Locations of sampling points are determined with GPS (global position system) and locations are recorded. Ardahan province located in the Northeastern Anatolia is at an altitude of 1829 m between $41^{\circ}36'13''$ north, $40^{\circ}45'24''$ south latitudes and $42^{\circ}25'43''$ west, $43^{\circ}29'17''$ east longitudes. Ardahan Plateau is located in the central part of the province and this plateau is composed of the Neogene volcanic lavas. Animal husbandry has become a major source of livelihood, as landforms and climate conditions largely limit agricultural activities in this basin. Therefore, baseline data on a systematic evaluation of the background radiation is felt that it was necessary to define. The concentrations of natural radioactivity in soil

samples taken from these stations are determined (Figure 1).

2.2 Sampling

A total of 35 surface samples were collected from the undisturbed and uncultivated locations. Every soil sample was collected from 6 different spot in an area of about 120 m^2 and up to a deepness of 15-20 cm. After cleaning the foreign objects off the ground, the sub soil samples were blended properly and were placed in labeled plastic bags. The homogenized soil samples were dried in air and then pulverized with blender to pass through a 100-mesh sieve. Each sample was filled up in airproof cylindrical plastic container and dry-weighed. The dried samples were kept for about 45 days period to let for secular equilibrium between ^{238}U and ^{232}Th and their short lived decay products.

2.3 Gamma-ray detection system

The concentrations of natural radioactivity in soil samples taken from these stations were determined by the Ortec manufactured a $7.62\text{ cm} \times 7.62\text{ cm}$ NaI(Tl) detector. Multi-channel analyzer emulation software (MAESTRO-32) is used in a conjunction with a personal computer to analyze the obtained spectral data. The detector was surrounded by a 5 cm thick lead shield to decrease background gamma-radiation [3]. Each sample was then counted for 86000 s using a NaI(Tl) detector. The background gamma-ray spectrum of the detection system was detected with an empty PVC container under same conditions, and was deducted from the spectra of each sample. Standard reference material (IAEA-375) was used to found energy calibration and relative efficiency calibration of the gamma spectrometer. For calculating the net count rates under the most considerable photo peaks of the radium and thorium daughter peaks, the corresponding count rate from the background spectrum acquired for the same counting time was deducted. Afterwards the background extracted area prominent gamma ray energies was used to calculate the activity of the radionuclide [4]. The activity concentrations per unit mass of the above radionuclides were ascertained for each soil samples in units of Bq kg^{-1} . The activity ^{238}U concentration was determined from the average concentrations of ^{214}Bi (609, 1120 and 1765 keV) in the samples and that of ^{232}Th was determined from the average concentrations of ^{208}Tl (583 keV) and ^{228}Ac (911 keV).

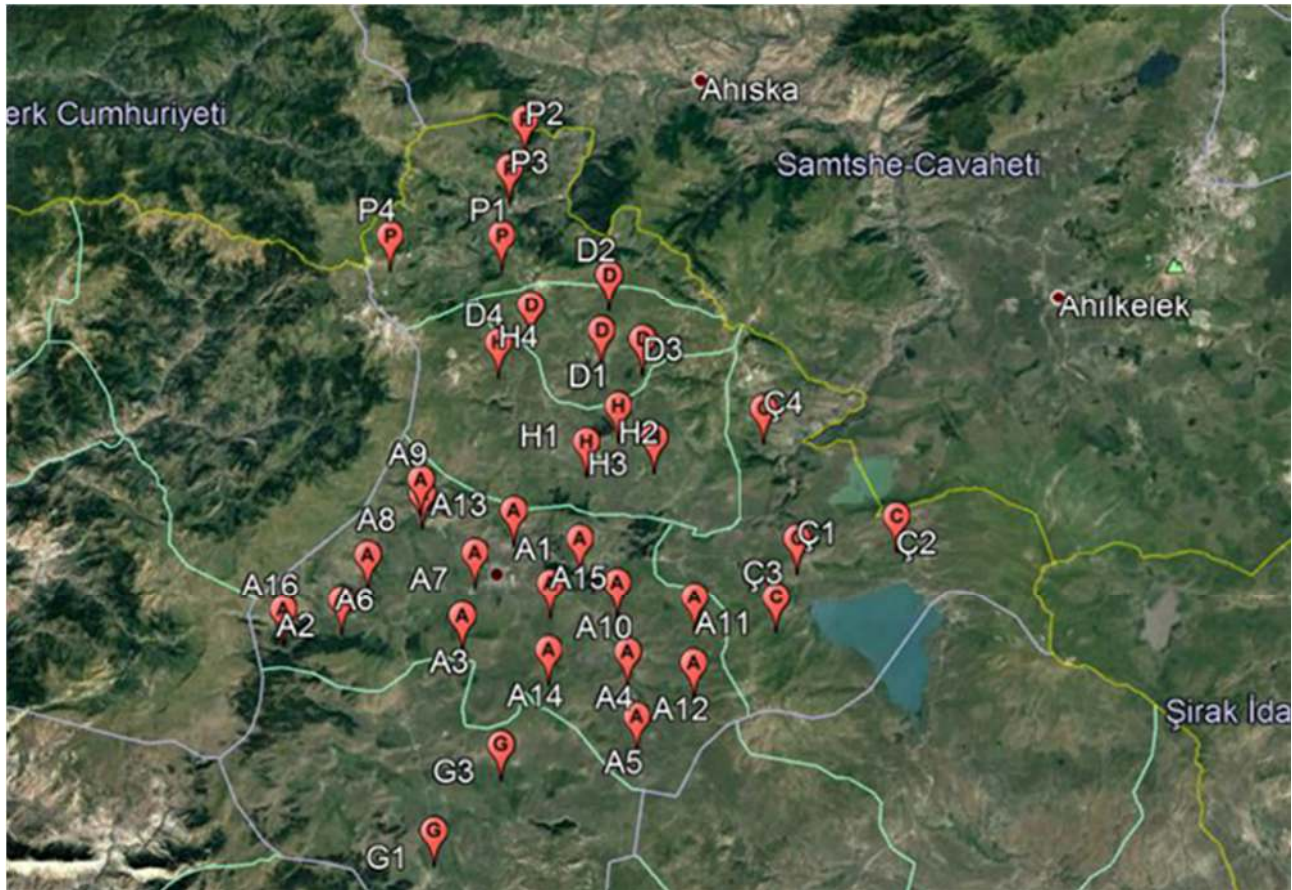


Figure 1. Satellite view of the sampling stations.

For the activity concentrations of ^{40}K and ^{137}Cs , the 1461 keV and 662 keV gamma lines analyzed [5].

2.4 Calculation of absorbed dose rate and annual effective dose rate

The release of gamma radiation from natural radionuclides must be determined and monitored for reducing the risk of cancer of people. The external terrestrial and cosmic origin gamma-radiation absorbed dose rate in air (ADR) in nGy h^{-1} at a height of about 1 meter about the ground are calculated by using the following formula [6]

$$ADR(\text{nGy h}^{-1}) = 0.462C_U + 0.623C_{Th} + 0.0417C_K + 0.1243C_{Cs} \quad (1)$$

where C_U , C_{Th} , C_K and C_{Cs} are the activity concentrations of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs , respectively, in collected soil samples. The conversion factors of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs are 0.462, 0.623, 0.04147 and 0.1243 nGy h^{-1} per Bq kg^{-1} , respectively [7].

For estimating the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective and the outdoor occupancy factor was taken into account. The annual effective dose rate

(AED) was calculated by using the following relation

$$AED (\mu\text{Svy}^{-1}) = (ADR) n\text{Gy h}^{-1} \times 8760\text{h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-3} \quad (2)$$

where ADR is absorbed dose rate in air (nGy h^{-1}), the conversion coefficient from absorbed dose in air to effective dose received by adults (0.7 Sv Gy^{-1}), the outdoor occupancy factor (0.2) and time (8760 h) [6].

3. RESULTS AND DISCUSSION

The results of the measurements of natural radionuclide (^{238}U , ^{232}Th and ^{40}K) concentrations and also ^{137}Cs concentrations in the 35 soil samples collected from the studied areas are summarized in Table 1. The activity concentrations of ^{228}U ranged from $7.9 \pm 2.5 \text{ Bq kg}^{-1}$ (Posof) to $48.6 \pm 7.4 \text{ Bq kg}^{-1}$ (Damal), of ^{232}Th from $11.8 \pm 2.9 \text{ Bq kg}^{-1}$ (Posof) to $55.5 \pm 9.0 \text{ Bq kg}^{-1}$ (Damal), of ^{40}K from $256.2 \pm 26.4 \text{ Bq kg}^{-1}$ (Ardahan Centrum) to $667.6 \pm 27.4 \text{ Bq kg}^{-1}$ (Damal) and ^{137}Cs from $1.3 \pm 0.6 \text{ Bq kg}^{-1}$ (Ardahan) to $39.50 \pm 1.2 \text{ Bq kg}^{-1}$ (Hanak). The world average concentrations are 35 Bq kg^{-1} , 30 Bq kg^{-1} and 400 Bq kg^{-1} for ^{228}U ,

^{232}Th and ^{40}K , respectively. Table 2 represents the average concentrations of ^{228}U in soil of these areas is lower than the world of average value and also the Turkey average value but slightly higher than the East Anatolia average value [6, 8, 9].

Table 1. Radioactivity concentrations ^{228}U , ^{232}Th , ^{40}K and ^{137}Cs measured in the soil samples, absorbed dose rate (ADR) and annual effective dose rate (AED) at present study.

Districts of Sample	Number of Sample	^{238}U	^{232}Th	^{40}K	^{137}Cs	ADR (nGy h ⁻¹)		AED (μSv y ⁻¹)	
		(Bqkg ⁻¹)	(Bqkg ⁻¹)	(Bqkg ⁻¹)	(Bqkg ⁻¹)	Terrestrial+Cosmic		Terrestrial+Cosmic	
Ardahan									
Centrum	16	32.6±6.4	33.7±5.6	438.2±22.0	14.1±0.7	55.5±6.4		68.1±5.6	
Çıldır	4	27.6±6.2	44.8±7.7	410.7±21.8	10.5±0.7	59.4±6.2		72.9±7.7	
Damal	4	36.6±7.3	48.3±8.9	441.9±28.4	21.5±1.0	67.3±7.3		82.5±8.9	
Göle	3	23.9±6.2	42.1±7.5	397.2±21.8	22.2±0.9	55.8±6.2		68.5±7.5	
Hanak	4	30.7±6.7	38.0±8.3	421.9±25.2	18.1±0.9	57.0±6.7		69.9±8.3	
Posof	4	14.0±4.7	21.9±6.6	490.0±20.0	11.0±1.1	41.9±6.1		51.4±8.2	
Minimum		7.9±2.5	11.8±2.9	256.2±26.4	1.3±0.6	28.5±6.2		35.0±6.9	
Maximum		48.6±7.4	55.5±9.0	667.6±27.4	39.50±1.2	76.9±8.9		94.3±9.0	
Mean	35	29.9±6.2	36.7±6.8	435.1±23.9	15.5±0.8	56.3±8.7		69.0±6.7	

The average activity concentrations of ^{232}Th and ^{40}K for Ardahan province are higher than the average activity concentrations of the world and lower than some other published results mentioned in Table 2. The relatively high values of ^{40}K may be a result of its abundance in the earth crust. In addition the heavy utilization of potassium having manure in the area close to the sampling areas may conduce to the excessive values of ^{40}K activity [6]. ^{137}Cs is disseminated to atmosphere through nuclear power plant accidents, worldwide nuclear explosion and other preceding test of nuclear devices throughout the world. This study aimed to obtain an estimate of fallout in all of the locations. ^{137}Cs activity concentration results in soil samples are given in Table 1. While some areas show significantly low activity concentration results, the

other areas show high results. The less fallout values of ^{137}Cs can be explained by rain, winds, cattle grazing, harvest etc. or some other reason [10]. Figure 2 shows a map of the ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs activity concentrations which is obtained from 35 soil samples in Ardahan Province.

The absorbed dose rates in air due to the terrestrial radiations and cosmic radiations were calculated using formula (1). The values ranged from 28.5 to 76.9 nGy h⁻¹ with a mean of 56.3 nGy h⁻¹. These values are comparable to the world average of 60 nGy h⁻¹ [6]. The annual effective outdoor dose rates estimated according to formula (2) for soil samples in Ardahan province ranged between 35.0 and 94.3 μSv y⁻¹.

The mean annual effective dose rate equivalents from the calculated outdoor gamma radiation (the terrestrial radiations and cosmic radiations) were found to be 69.0 μSv y⁻¹. The minimum value was obtained in Posof district and maximum value in Damal district. Our results for average annual effective dose rate are in the range of world average value (70 μSv y⁻¹) [6]. In Table 2, the obtained mean outdoor gamma absorbed dose rates in air and the obtained mean annual effective dose rates for studied area were compared with the reported mean values of Turkey and worldwide.

Altitude of region has a significant effect on the level of background radiation. Despite their high altitude, low background radiation is seen in some areas due to low concentrations of radionuclides in their soils [26]. According to our measurements in this study indicate that there is a good correlation between the altitude and the concentration activity of the soil samples. Figure 3 shows that the concentrations of ^{228}U and ^{232}Th are increased with altitude but ^{40}K is decreased linearly with altitude (Figure 4).

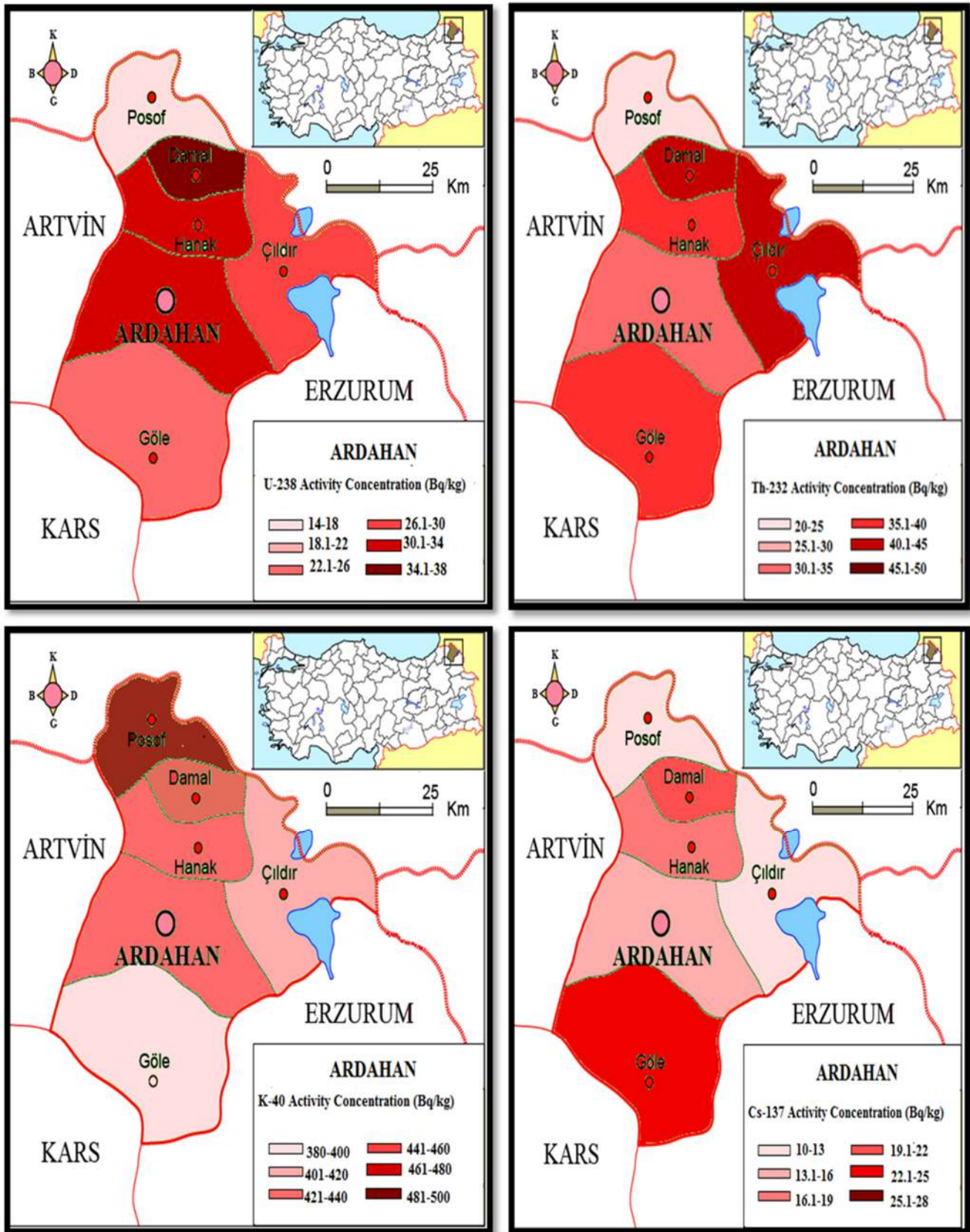


Figure 2. ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs activity concentrations map of the studied area

Table 2: Comparison of natural radioactivity levels in soil samples, absorbed dose rate (ADR) and annual effective dose rate (AED) at present study stations with values reported in literature.

Region (References)	^{238}U (Bqkg ⁻¹)	^{232}Th (Bqkg ⁻¹)	^{40}K (Bqkg ⁻¹)	^{137}Cs (Bqkg ⁻¹)	$\frac{\text{ADR (nGyh}^{-1})}{1}$ Terrestrial	$\frac{\text{AED (\muSvy}^{-1})}{1}$ Terrestrial
Present Study						
Ardahan Province	29.9±6.5	36.70±6.9	435.10±23.9	15.5±0.8	56.3	69.0
Istanbul [11]	21	37	342	1.8-81	49	65
Manisa [12]	29	27	340		54	66
Rize [13]	11-188	10-105	105-1235	19-232	77.4	
Kastamonu [14]	32.93	27.17	431.43	8.02	48.03	60
Sanliurfa [15]	20.8	24.95	298.6		38.24	46.9
Adana [16]	17.6	21.1	297.5	6.8	67	82
Giresun [17]	33±13	43±14	733±86	318±46		92
Kirklareli [18]	28±13	40±18	667±282	8±5	71	87
Artvin and Ardahan [19]	22±2	19±2	358±4	54±2	38	47
Trabzon [20]	36±1	31±0.6	341±3	14±0.4	50	62
Kars Centre [21]	41	35	437	21	59	72
Kars Province [22]	47.8	31.2	536	18	44.76	54.89
East Anatolia [9]	31.95±18.5	27.70±16.5	458.47±18.6	14.9±0.4	52.3	64.17
India [23]	29.1	31.7	437.3	13.8	50.9	62
China [24]	37.7	75.3	195.2		74.3	
Nigeria [25]	38.5	54.6	584		122	150
United States [6]	41.0±5.0	29.7 ± 4	412.5 ±20.0		54.6	200
Greece [6]	40	35	370		47	
Bulgaria [6]	25	21	360		56	
Turkey [8]	45	30	400		45	
Worldwide [6]	34.7±1.7	35.4±0.8	450±17.9	11.6±0.5	54.6	70
	35	30	400		60	70

The natural radioactivity levels in soil are dependent to the sort of rock from which the soils arise. As seen from Figure 3, the increasing of the concentrations of ^{238}U and ^{232}Th as a function of altitude may be explained by the presence of radioactive-rich granite, phosphate, quartzite and sedimentary rocks. According to our calculations the measured activity concentrations of ^{40}K decrease at the higher attitudes of the studied area due to inability of agriculture and animal husbandry.

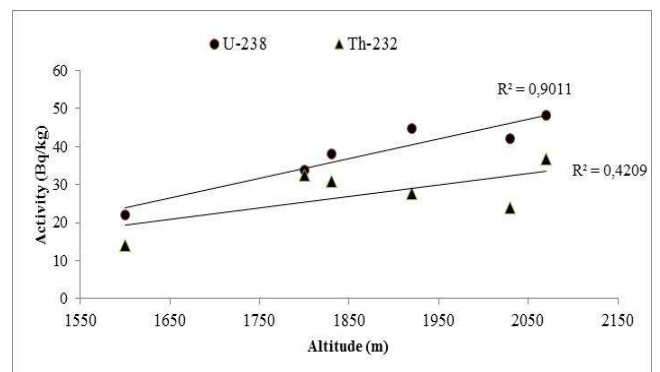


Figure 3. The determined activity concentrations of ^{238}U and ^{232}Th radionuclides from studied areas in soil at the various altitudes.

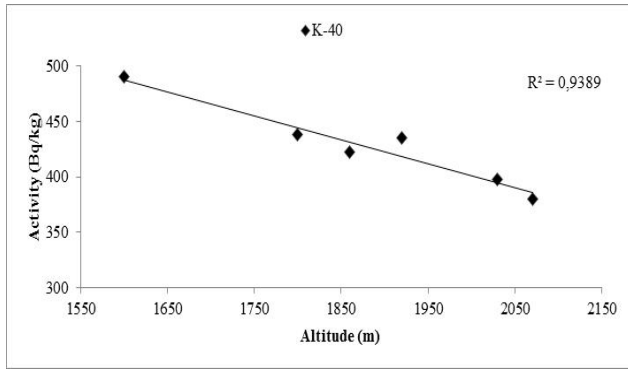


Figure 4. The activity concentrations of ^{40}K radionuclide in soil at the different altitudes.

4. CONCLUSION

The average activity concentrations of soil samples for the Ardahan province were found to be 29.9 ± 6.5 , 36.7 ± 6.9 , 435 ± 239 and 15.5 ± 0.8 Bq kg^{-1} for ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs , respectively. The concentrations of ^{232}Th and ^{40}K in soil samples of Ardahan province are higher than the world wide mean values but the concentration of ^{238}U is slightly lower than the world wide mean values. As can be seen from our analysis there is a good correlation between the altitude and the activity concentrations of soil samples for the studied area. The results obtained have shown that the annual effective dose rate due to natural radioactivity of samples is very much comparable with recommended world value and also with values reported in literature. The obtained data from this study can be used as baseline data for producing a radiological map of the studied area and determining the effects of radioactive contamination in the future.

REFERENCES

- [1] Anagnostakis M. J; E.P. Hinis, S.E. Simopoulos, et al. *Natural Radioactivity Mapping of Greek Surface Environment International*, 1996, Vol. **22**, Suppl. 1, S3-S8, Editor: J.P. McLaughlin, E.S. Simopoulos, F. Steinhäusler.
- [2] Ravisankar, R., Chandrasekaran, A., Vijayagopal, P., et al. Natural radioactivity in soil samples of Yelagiri Hills, Tamil Nadu, India and the associated radiation hazards, *Radiation Physics and Chemistry*, 2012, **81**, 1789-1795.
- [3] Chiozzi P., P. De Felice, A. Fazio, Pasquale, V.; Verdoya, M. Laboratory application of NaI (Tl) γ -ray spectrometry to studies of natural radioactivity in geophysics, *Applied Radiation and Isotopes*, 2000, **53**, 127-132.
- [4] Mehra, R., Singh, S. and Singh K. Analysis of ^{226}Ra , ^{232}Th and ^{40}K in soil samples for the assessment of the average effective dose, *Indian J. Phys.*, 2009, **83** (7), 1031-1037.
- [5] Hamby, D. M., Tynybekov, A.K. Uranium, thorium and potassium in soils along the shore of lake Issyk-Kyol in the Kyrghyz Republic, *Environ. Monitoring Assessment*, 2000, **73**, 101-108.
- [6] UNSCEAR, Sources, Effects and Risks of Ionizing Radiation Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly. United Nations, New York, 2000.
- [7] Beck, H. L. Physics of Environmental Gamma Radiation Fields. The Natural Radiation Environment 2, USERDA Report CON-720805-P2, 101-134, 1972.
- [8] TAEK, Türkiye'deki Çevresel Radyoaktivitenin İzlenmesi 2009, Technique Report, Ankara 9-14, 2010.
- [9] Turhan Ş., Köse A., Varinlioğlu A., et al. Distribution of terrestrial and anthropogenic radionuclides in Turkish surface soil samples, *Geoderma*, 2012, **187-188**, 117-124.
- [10] Akhtar N., M. Ashraf, M. Tifail, et al. Radiometric and Chemical Analysis of Saline Soil Samples of Pacca Anna, Faisalabad, *Journal of Research*, 2003, **14**(1), 49-59.
- [11] Karahan, G., Bayülken, A. Assessment of Gamma Dose Rates Around İstanbul (Turkey), *Journal of Environmental Radioactivity*, 2000, **47**, 213-221.
- [12] Erees, F.S., Akozcan, S., Parlak, Y., et al. Assessment of dose rates around Manisa (Turkey), *Radiation Measurements*, 2006, **41**, 598-601.
- [13] Kurnaz, A., Küçükömeroğlu, B., Keser, et al. Determination of radioactivity levels and hazards of soil and sediment samples in Fırtına Valley (Rize, Turkey), *Applied*

- Radiation and Isotopes*, 2007, **65**, 1281–1289.
- [14] Kam, E., Bozkurt, A. Environmental radioactivity measurements in Kastamonu Region of northern Turkey, *Applied Radiation and Isotopes*, 2007, **65**, 440–444.
- [15] Bozkurt, A. Yorulmaz, N., Kam, E., et al. Assessment of environmental radioactivity for Sanliurfa region of Southeastern Turkey, *Radiation Measurements*, 2007, **42**, 1387–1391.
- [16] Değerlier, M, Karahan, G., Özger, G. Radioactivity concentrations and dose assessment for soil samples around Adana, Turkey, *Journal of Environmental Radioactivity*, 2008, **99(7)**, 1018–1025.
- [17] Çelik, N., Çevik, U., Çelik, A., Küçükömeroğlu, B. Determination of indoor radon and soil radioactivity levels in Giresun, Turkey, *Journal of Environmental Radioactivity*, 2008, **99**, 1349–1354.
- [18] Taskin H., Karavus, M., Ay, P., et al. Radionuclide concentrations in soil and lifetime cancer risk due to gamma radioactivity in Kirklareli, Turkey, *Journal of Environmental Radioactivity*, 2009, **100**, 49-53.
- [19] Küçükömeroğlu, B., Yeşilbağ, Y.O., Kurnaz, et al. Radiological characterization of Artvin and Ardahan provinces of Turkey, *Radiation Protection Dosimetry*, 2011, **145(4)**, 389–394.
- [20] Kurnaz, A., Küçükömeroğlu, B., Damla, N., et al. Radiological maps for Trabzon, Turkey, *Journal of Environmental Radioactivity*, 2011, **102**, 393-399.
- [21] Cengiz, G. B., Reşitoğlu, S. Determination of natural radioactivity levels in Kars City center, Turkey, *Journal of Nuclear Sciences*, 2014, **1**, 32-37.
- [22] Cengiz, G. B., İ. Çağlar. Determination of the Health Hazards and Life time Cancer Risk Due to Natural Radioactivity in Soil of Akyaka, Arpaçay and Susuz Areas of Kars, Turkey, *International Journal of Scientific & Engineering Research*, March 2016, **7(3)**, 619-626.
- [23] Selvasekarapandian S., R Sivakumar, NM Manikandan, et al. Natural radionuclide distribution in soils of Gudalore, India, *Applied Radiation and Isotopes* 2000, **52**, 299-306.
- [24] Y. Yang, X. Wu, Z. Jiang, et al. Radioactivity concentrations in soils of the Xiazhuang granite area China, *Appl. Radiat. Isot.*, 2005, **63**, 255–259.
- [25] Agbalagba, E. O, Avwiri, G.O, Chad-Umoreh, Y.E. γ -Spectroscopy measurement of natural radioactivity and assessment of radiation hazard indices in soil samples from oil fields environment of Delta State, Nigeria, *Journal of Environmental Radioactivity*, 2012, **109**, 64-70.
- [26] Daryoush Shahbazi-Gahrouei. Natural Background Radiation Dosimetry in the Highest Altitude Region of Iran, *Journal of Radiation Research*, 2003, **44**, 285-287.