Journal of Hazardous Materials 162 (2009) 146–153

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Natural and artificial radioactivity measurements in Eastern Black Sea region of Turkey

N. Celik[a,*], U. Cevik[a,d], A. Celik[b], B. Koz[c]

[a] Department of Physics, Karadeniz Technical University, Trabzon, Turkey
[b] Department of Physics, Giresun University, Giresun, Turkey
[c] Department of Biology, Giresun University, Giresun, Turkey
[d] Department of Chemistry, University of Antwerp, Antwerp, Belgium

ARTICLE INFO

Article history:
Received 29 January 2008
Accepted 5 May 2008
Available online 9 May 2008

Keywords:
Environmental radioactivity
Artificial radioactivity
Eastern Black Sea
Chernobyl
Cs-137

ABSTRACT

In the present work, naturally occurring radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were measured in soil samples collected from the Eastern Black Sea region of Turkey. It was found that the activity concentrations ranged from 12 to 120 Bq kg^{-1} for ^{226}Ra, from 13 to 121 Bq kg^{-1} for ^{232}Th and from 204 to 1295 Bq kg^{-1} for ^{40}K. Besides naturally occurring radionuclides, ^{137}Cs activity concentration was measured in soil, lichen and moss samples and it was found that ^{137}Cs activity concentration ranged from 27 to 775 Bq kg^{-1} with for soil, from 29 to 879 Bq kg^{-1} for lichen and from 67 to 1396 Bq kg^{-1} for moss samples. Annual effective doses due to the naturally occurring radionuclides and ^{137}Cs were estimated. Ecological half-lives of ^{137}Cs in lichen and moss species were estimated. The decrease of the activity concentrations in the present measurements (2007) relative to those in 1993 indicated ecological half-lives between 1.36 and 2.96 years for lichen and between 1.35 and 2.85 years for moss species.

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1. Introduction

Natural and artificial radionuclides are two sources of radiation in the environment which are the main sources of radiation exposure to human beings [1]. The natural sources are largely due to the primordial radionuclides, mainly ^{238}U and ^{232}Th and their decay products, as well as ^{40}K. These radionuclides are present in various degrees in all media in the environment, including the human body itself. Besides naturally occurring radionuclides, many radionuclides of artificial origin have been released into the environment by different processes. The isotope ^{137}Cs is one of them and is produced anthropogenically by several types of nuclear activities including past testing of nuclear weapons, accidents in nuclear facilities, reprocessing of spent nuclear fuel and nuclear power reactors. Until the Chernobyl event on 26 April 1986, the activity of ^{137}Cs occurred mostly as a result of global fallout from atmospheric nuclear weapons tests. Around 9.6×10^{15} Bq of ^{137}Cs was injected in the stratosphere [2] from past tests. After the Chernobyl, ^{137}Cs atmospheric activity in central and northern Europe is expected to be strongly controlled by the Chernobyl influence [3].

Turkey, especially the northern part of it, was one of the countries which were contaminated by the Chernobyl accident [4]. The radioactive plume from the accident reached Turkey by 5 May 1986, substantially contaminating various regions and ecosystems of the country. During the emergency, Cekmece Nuclear Research and Training Center (ÇANEM) performed an analysis of various substances. It their report, it has been noted that the surface soil ^{137}Cs activity concentration of the eastern part of the Black Sea mountains was around 4000–4500 Bq kg^{-1} at the 0.5 cm soil in the year 1988 [5]. And also the level of ^{137}Cs activity concentration in Turkish tea plant was found to be maximum value of 44,000 Bq kg^{-1} for the 1986 products by Gedikoglu and Sipahi [6]. 373 Bq kg^{-1} activity-level was reported elsewhere [7] for ^{137}Cs in lichen samples collected from the Giresun province in 1997. Since then almost any comprehensive studies have been conducted in the mentioned study area.

In the present work, natural (^{226}Ra, ^{232}Th and ^{40}K) and artificial (^{137}Cs) radionuclides have been measured in various environmental samples and the dose received due to the exposure of these radionuclides have been estimated and compared to the world average dose rates. Ecological half-lives of ^{137}Cs in lichen and moss species collected from the studied area were estimated.

* Corresponding author. Tel.: +90 462 377 38 22; fax: +90 462 325 31 95.
E-mail address: necaticelik1@yahoo.com (N. Celik).

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doi:10.1016/j.jhazmat.2008.05.017

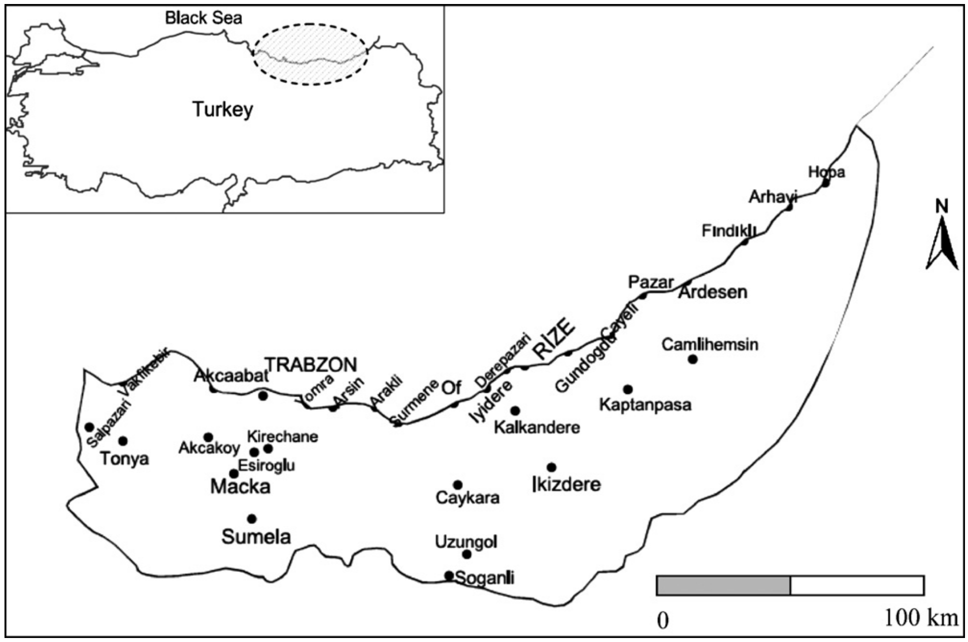

Fig. 1. Studied area.

2. Materials and methods

2.1. Study area, sampling and analysis

The Eastern Black Sea region (Fig. 1) has a steep, rocky coast with rivers that cascade through the gorges of the coastal ranges. The region covers approximately 18 percent of the land in Turkey, with a surface area of 141,000 km^2. The great majority of the people in the region earn their living from the land. The Black Sea coast receives the greatest amount of rainfall. The eastern part of that coast averages 1400 mm annually and is the only region of Turkey that receives that rainfall throughout the year.

Soil, lichen and moss samples were collected from locations in the Eastern Black Sea region of Turkey in 2006. In all a total of 180 (60 soil, 60 lichen, 60 moss) samples were analyzed. The plant samples were collected using simple tools such as large knife, an iron bar etc. These plant samples were cleaned of soil mud and stones and temporarily stored in nylon bags. A special attention was paid to recollect the same species which were collected before by other researchers [8,9] at the same sampling sites. For identification of moss and lichen species, macroscopic and microscopic characters were examined with stereo and light microscope and by reference to literature [10–14].

Then the samples were dried in an oven at a temperature of 70 °C, sieved to remove the stones and pebbles, and crushed in the laboratory to homogenize them. Then they were weighed, sealed and stored at least for 3 weeks to allow secular equilibrium between radium and thorium and their decay products. Then each sample was measured and the values are given in Bq kg^{-1} dry weight.

The gamma-ray activities were measured using HPGe computer controlled detector having the resolution of 2 keV for the 1332 keV energy line of ^{60}Co with conventional electronics and 20% relative efficiency and Genie 2000 as the software. The detector was shielded with 10 cm thick lead layer to reduce the background due to the cosmic rays and the radiation nearby the system.

Full energy peak efficiencies were determined using Standard Reference Material [15] for soil samples and Standard Reference Material [16] for lichen and moss samples prepared by International Atomic Energy Agency. Decay corrections were performed to the sampling date.

The gamma-ray lines of 238 keV from ^{212}Pb, 352 keV from ^{212}Pb and 609 keV from ^{214}Bi were used to evaluate the ^{226}Ra activity concentration; 583 keV gamma-rays from ^{208}TI and 911 keV from ^{228}Ac were used to determine to the ^{232}Th activity concentration. The activity concentrations of ^{40}K and ^{137}Cs were determined using their 1460 keV and 661 keV gamma-ray lines, respectively. The specific activity of each sample was then calculated by using the following formula:

$$A = \frac{C_{net}}{\varepsilon I_{\gamma} t m} \quad (1)$$

where C_{net} is the net area of the total absorption line, A is the activity of the isotope in Bq kg^{-1}, I_γ is the absolute intensity of the transition, t is the sample measurement time, ε is the full energy peak efficiency, and m is the mass of the sample.

3. Results and discussions

3.1. Gamma-activity results

3.1.1. Activity in soil

The results of activity concentrations in the soil samples from sixty different sites are gives in Table 1 for the natural radionuclides of ^{226}Ra, ^{232}Th and ^{40}K and the artificial radionuclide of ^{137}Cs. The concentrations found in the present study ranged from 12 to 120 Bq kg^{-1} (the average value is around 55 Bq kg^{-1}) for ^{226}Ra, from 13 to 121 Bq kg^{-1} (the average value is 41 Bq kg^{-1}) for ^{232}Th, from 204 to 1295 Bq kg^{-1} (the average value is around 622 Bq kg^{-1}) for ^{40}K and from 27 to 775 Bq kg^{-1} (average value is around 170 Bq kg^{-1}) for ^{137}Cs. The world's mean values of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations are 32, 45 and 420 Bq kg^{-1}, respectively [1]. The mean values of ^{226}Ra and ^{40}K are higher than the world's average values, whereas the mean value of ^{232}Th observed in the present study is lower than the world's average value.

Table 1
Activity concentrations in soil samples, dose rates and effective dose rates

Sampling site	Specific activity concentrations (Bq kg^{-1})				Dose rate (nGy h^{-1})		Annual effective dose (μSv $year^{-1}$)	
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Due to ^{226}Ra, ^{232}Th and ^{40}K	Due to ^{137}Cs	Due to ^{226}Ra, ^{232}Th and ^{40}K	Due to ^{137}Cs
Hopa (Sarp)	51.2 ± 6.3	35.3 ± 4.4	572.5 ± 34.2	75.3 ± 6.4	69.54	2.25	84.07	2.72
Hopa (Kemalpasa)	46.1 ± 5.4	24.0 ± 3.7	617.9 ± 36.4	82.5 ± 6.9	62.10	2.46	75.08	2.97
Hopa (Gunesli)	36.5 ± 4.6	28.3 ± 4.2	327.0 ± 22.1	95.3 ± 7.2	47.97	2.85	57.99	3.45
Arhavi (Merkez)	65.3 ± 7.6	46.9 ± 6.5	1005.7 ± 43.4	67.1 ± 5.6	102.13	2.01	123.46	2.43
Findikli (Aslandere)	44.4 ± 5.0	32.3 ± 4.7	553.2 ± 36.6	73.2 ± 6.0	63.75	2.19	77.07	2.65
Findikli (Cinarli)	79.0 ± 7.7	25.2 ± 3.1	498.5 ± 32.6	60.3 ± 5.1	71.74	1.8	86.73	2.18
Findikli (Beydere)	72.7 ± 6.3	26.0 ± 3.1	369.1 ± 29.7	77.9 ± 6.9	63.82	2.34	77.15	2.83
Findikli (Tatlisu)	93.1 ± 8.3	64.3 ± 6.7	204.5 ± 20.4	127.5 ± 11.2	90.85	3.84	109.83	4.64
Findikli (Caglayan)	47.2 ± 6.9	32.7 ± 4.5	668.7 ± 37.4	79.2 ± 6.3	70.02	2.37	84.65	2.87
Ardesen (Seslikaya)	58.3 ± 7.3	50.5 ± 4.6	618.3 ± 35.0	78.2 ± 5.8	84.44	2.34	102.08	2.83
Ardesen (Dogankoy)	49.2 ± 5.3	33.2 ± 4.7	532.1 ± 32.6	89.3 ± 7.1	65.65	2.67	79.36	3.23
Pazar (Merkez)	59.8 ± 6.6	57.0 ± 5.7	1253.6 ± 48.9	749.3 ± 35.4	116.81	22.47	141.20	27.16
Pazar (Hemsin)	46.4 ± 5.0	30.4 ± 4.6	432.3 ± 31.0	774.7 ± 35.4	58.08	23.25	70.21	28.11
Pazar (Hilalkoy)	36.3 ± 5.7	25.5 ± 4.0	368.8 ± 24.5	608.5 ± 32.3	47.79	18.24	57.77	22.05
Camlihemsin (Merkez)	43.8 ± 4.6	50.7 ± 7.6	398.2 ± 24.7	62.9 ± 7.2	69.24	1.89	83.70	2.28
Cemlihamsin (S. Oc. Yani)	40.7 ± 5.1	27.3 ± 3.4	523.5 ± 32.5	216.5 ± 13.5	57.44	6.48	69.44	7.83
Cayeli (Kaptanpasa)	67.3 ± 7.5	46.8 ± 6.8	770.1 ± 39.0	127.2 ± 9.3	92.83	3.81	112.22	4.61
Cayeli (Buyukkoy)	86.5 ± 7.9	48.7 ± 5.7	1103.5 ± 44.7	209.1 ± 9.5	116.59	6.27	140.94	7.58
Cayeli (Madenli)	57.6 ± 5.9	35.5 ± 4.8	659.2 ± 37.4	94.9 ± 6.4	75.85	2.85	91.69	3.45
Gundogdu (Akpinar)	36.4 ± 5.1	28.9 ± 4.1	496.7 ± 31.9	46.3 ± 6.7	56.03	1.38	67.73	1.67
Gundogdu (Valikoy)	95.3 ± 7.9	55.0 ± 6.8	556.4 ± 37.1	26.8 ± 4.2	100.88	0.81	121.96	0.98
Rize (Salarha)	75.5 ± 6.3	34.4 ± 5.6	805.4 ± 40.3	106.5 ± 7.8	89.15	3.18	107.77	3.84
Rize (Derepazari)	27.2 ± 3.2	36.3 ± 4.7	675.3 ± 38.2	148.4 ± 8.5	64.39	4.44	77.83	5.37
Iyidere (Ciftlik)	45.4 ± 5.7	56.4 ± 7.8	1132.5 ± 41.0	60.3 ± 7.6	104.96	1.8	126.89	2.18
Kendirli	80.5 ± 7.5	52.6 ± 6.5	481.3 ± 25.6	153.4 ± 8.7	89.93	4.59	108.71	5.55
Kalkandere (Yolbasikoy)	71.0 ± 5.7	48.6 ± 5.4	851.2 ± 41.5	176.7 ± 7.7	98.69	5.34	119.30	6.46
Ikizdere	88.3 ± 8.4	35.0 ± 4.3	369.9 ± 23.5	234.3 ± 13.9	76.66	7.02	92.67	8.49
Rize (Kucukkoy)	12.5 ± 3.2	53.6 ± 5.9	656.7 ± 36.6	209.5 ± 9.2	68.50	6.27	82.81	7.58
Of (Klavuz)	17.3 ± 3.8	40.3 ± 4.6	459.1 ± 29.7	131.1 ± 9.0	53.48	3.93	64.65	4.75
Of (Bolumlu)	32.4 ± 4.1	13.2 ± 3.0	1038.3 ± 40.6	67.4 ± 5.8	66.90	2.01	80.88	2.43
Of (Kadinlar Yaylasi)	31.4 ± 4.3	27.4 ± 3.9	653.0 ± 35.1	159.9 ± 8.7	59.19	4.8	71.55	5.80
Caykara (Uzungol)	43.3 ± 5.1	31.1 ± 4.5	439.2 ± 25.8	156.1 ± 7.5	57.76	4.68	69.82	5.66
Caykara (Uzuntarla)	29.9 ± 4.3	38.5 ± 4.8	567.7 ± 33.4	132.3 ± 8.1	61.96	3.96	74.91	4.79
Caykara (Soganli)	46.1 ± 5.3	32.1 ± 5.8	593.5 ± 38.1	162.4 ± 9.4	66.33	4.86	80.18	5.88
Caykara (Camlibel)	75.4 ± 6.3	47.4 ± 4.7	872.3 ± 42.7	134.3 ± 8.3	100.64	4.02	121.66	4.86
Surmene (Ormansever)	57.7 ± 7.4	28.4 ± 4.8	596.3 ± 37.5	147.6 ± 7.9	68.50	4.41	82.81	5.33
Surmene (Buyukdogan)	29.9 ± 5.3	30.0 ± 5.2	969.2 ± 45.6	464.5 ± 24.3	73.91	13.98	89.35	16.90
Surmene (Tasliyayla)	85.3 ± 8.7	47.5 ± 4.5	547.6 ± 32.5	396.2 ± 22.7	90.93	11.88	109.92	14.36
Arakli (Dagbasi)	61.2 ± 5.4	37.3 ± 5.1	348.9 ± 23.8	124.5 ± 9.2	65.46	3.72	79.14	4.50
Arakli (Tastepe)	25.2 ± 3.6	19.5 ± 3.2	431.3 ± 24.4	40.4 ± 3.8	41.79	1.2	50.51	1.45
Arakli (Pazarcik)	32.2 ± 5.2	18.3 ± 4.1	547.8 ± 31.2	158.4 ± 9.4	49.19	4.74	59.46	5.73
Arsin (Cubuklu)	17.9 ± 4.3	26.9 ± 3.7	582.2 ± 36.5	102.3 ± 8.4	50.16	3.06	60.64	3.70
Yomra (Gulyurdu)	49.2 ± 6.0	121.4 ± 9.8	651.3 ± 38.4	171.2 ± 9.8	129.02	5.13	155.97	6.20
Yomra (Demirciler)	120.3 ± 10.6	36.3 ± 5.1	705.4 ± 36.2	182.4 ± 10.1	105.39	5.46	127.40	6.60
Trabzon (Yesilyurt)	93.3 ± 8.0	56.3 ± 5.5	695.7 ± 32.9	125.4 ± 8.7	106.71	3.75	129.00	4.53
Trabzon (Kirechane)	82.3 ± 6.5	65.3 ± 5.7	778.1 ± 38.3	343.6 ± 21.5	111.50	10.32	134.79	12.48
Trabzon (Akyazi)	45.3 ± 4.9	32.1 ± 4.2	632.3 ± 34.4	107.6 ± 8.0	67.58	3.21	81.69	3.88
Macka (Ormanici)	80.6 ± 7.6	48.2 ± 5.0	592.1 ± 33.2	65.3 ± 5.5	91.39	1.95	110.48	2.36
Macka (Esiroglu)	72.0 ± 5.6	36.3 ± 4.8	761.4 ± 36.5	76.4 ± 5.2	87.30	2.28	105.53	2.76
Macka (Zirasa)	36.4 ± 5.2	14.4 ± 2.5	435.6 ± 24.7	79.8 ± 6.9	43.39	2.4	52.45	2.90
Macka (Sumela)	95.7 ± 8.7	57.3 ± 5.2	748.9 ± 37.1	157.3 ± 7.5	110.51	4.71	133.59	5.69
Akcaabat (Karacayir)	73.0 ± 5.9	30.2 ± 4.6	679.2 ± 37.9	127.9 ± 7.9	80.23	3.84	96.99	4.64
Akcaabat (Karasu)	61.4 ± 5.5	45.7 ± 3.9	835.8 ± 40.2	196.5 ± 9.2	92.45	5.88	111.76	7.11
Akcaaat (Akcakoy)	37.3 ± 4.8	27.1 ± 4.2	452.1 ± 25.3	185.3 ± 9.6	53.11	5.55	64.20	6.71
Vakfikebir (Kizilkaya)	42.5 ± 3.9	98.9 ± 6.6	1294.5 ± 47.6	66.5 ± 4.7	139.16	1.98	168.22	2.39
Vakfikebir (Kirazlik)	47.3 ± 4.3	32.1 ± 3.7	540.5 ± 31.2	177.2 ± 9.6	64.47	5.31	77.94	6.42
Tonya (Cayirici)	54.3 ± 5.4	57.8 ± 4.9	457.8 ± 30.4	186.5 ± 9.4	81.23	5.61	98.20	6.78
Tonya (Yakcukur)	37.2 ± 4.3	38.6 ± 5.2	312.5 ± 22.9	132.4 ± 7.8	55.03	3.96	66.53	4.79
Tonya (Rosada)	20.5 ± 3.1	16.9 ± 3.6	435.7 ± 23.3	144.9 ± 6.7	38.54	4.35	46.59	5.26
Salpazari (Karakaya)	66.3 ± 6.4	81.7 ± 7.3	213.5 ± 21.4	367.4 ± 23.5	91.63	11.01	110.76	13.31
Mean	55.2 ± ±6.1	40.9 ± 5.1	622.8 ± 34.4	169.7 ± 13.6	77.18	5.09	93.30	6.15

3.1.1.1. Dose estimation due to both natural and artificial radionuclides. The total absorbed dose rate D (nGy h^{-1}) in air at 1 m above ground level due to the presence of natural and artificial radionuclides in the samples was estimated using the following formula [1]:

$$D = aC_{Ra} + bC_{Th} + cC_K + dC_{Cs} \quad (2)$$

where a, b, c, and d are the dose rates per unit activity concentrations of Ra, Th, K, and Cs (Gy h^{-1}/Bq kg^{-1}) and C_{Ra}, C_{Th}, C_K, and C_{Cs} are the activity concentrations of Ra, Th, K, and Cs (Bq kg^{-1}), respectively. The values of a, b, c, and d are taken to be 4.27×10^{-10}, 6.62×10^{-10}, 0.43×10^{-10}, and 0.30×10^{-10} (Gy h^{-1}/Bq kg^{-1}), respectively [1,17]. In the present work the total absorbed dose rates due to naturally occurring radionuclides of ^{226}Ra, ^{232}Th and ^{40}K and the artificial

Table 2
Activity concentrations and ecological half-lives of ^{137}Cs in lichen and moss species

Sampling site	Scientific name		^{137}Cs activity concentration ($Bq\ kg^{-1}$ in dry weight)				Ecological half-life, T_e (year)	
	Lichen	Moss	Lichen		Moss		Lichen	Moss
			1993[a]	Present work	1993[b]	Present work		
Hopa (Sarp)	*Parmelia caperata* (L.) Hale	*Hypnum cupressiforme* Hedw.	4482	255.9 ± 20.9	6507	105.2 ± 7.8	1.66	1.57
Hopa (Kemalpaşa)	*Parmelia caperata* (L.) Hale	*Hypnum sp.* Hedw.	458	256.4 ± 21.1	3984	377.4 ± 22.6	2.41	1.68
Hopa (Güneşli)	*Parmelia caperata* (L.) Hale	*Pleurochaete squarrosa* (Brid.) Lindb.	8135	365.2 ± 24.3	5668	401.9 ± 26.7	1.53	1.60
Arhavi	*Parmelia caperata* (L.) Hale	*Thuidium tamariscinum* (Hedw.) Schimp.	10674	784.8 ± 38.9	6418	309.1 ± 24.0	1.48	1.58
Fındıklı (Aslandere)	*Parmelia caperata* (L.) Hale	*Pseudoscleropodium* sp. M. Fleisch.	8129	613.1 ± 36.5	21471	764.6 ± 34.3	1.53	1.35
Fındıklı (Çınarlı)	*Parmelia sulcata*	*Homalothecium lutescens* (Hedw.) H. Rob.	2290	455.6 ± 25.4	3356	844.3 ± 42.5	1.83	1.73
Fındıklı	*Parmelia sulcata*	*Pseudoscleropodium* sp. M. Fleisch.	7626	540.2 ± 30.9	9667	604.8 ± 35.5	1.54	1.49
Fındıklı	*Parmelia sulcata*	*Hypnum sp.* Hedw.	17035	879.2 ± 43.5	10035	918.2 ± 44.7	1.39	1.49
Fındıklı (Tatlısu)	*Parmelia sulcata*	*Hypnum sp.* Hedw.	3211	254.8 ± 21.1	6084	627.2 ± 36.4	1.74	1.59
Fındıklı (Çağlayan)	*Parmelia caperata* (L.) Hale	*Brachythecium starkei* (Brid.) Schimp.	1524	867.3 ± 40.7	14468	1395.5 ± 46.1	1.95	1.42
Ardeşen (Doğanköy)	*Parmelia sulcata*	*Brachythecium starkei* (Brid.) Schimp.	3553	286.5 ± 22.4	14007	714.4 ± 30.5	1.71	1.43
Ardeşen (Seslikaya)	*Parmelia caperata* (L.) Hale	*Thuidium tamariscinum* (Hedw.) Schimp.	4975	76.3 ± 5.6	1384	140.1 ± 10.8	1.63	1.98
Ardeşen (Ortaalan)	*Parmelia caperata* (L.) Hale	*Brachythecium* sp. Schimp.	1422	175.4 ± 10.2	6981	398.7 ± 26.7	1.97	1.56
Çamlıhemşin	*Parmelia caperata* (L.) Hale	*Brachythecium* sp. Schimp.	1142	234.2 ± 19.6	3590	426.2 ± 23.6	2.04	1.71
Pazar (Hemşin)	*Parmelia caperata* (L.) Hale	*Isothecium striatulum* (Spruce) Kindb.	3337	341.6 ± 22.6	14530	716.4 ± 32.4	1.73	1.42
Pazar	*Parmelia caperata* (L.) Hale	*Isothecium striatulum* (Spruce) Kindb.	5077	371.2 ± 23.6	3170	231.1 ± 20.0	1.63	1.74
Pazar (Hilalköy)	*Parmelia caperata* (L.) Hale	*Rhynchostegiella* sp. (Schimp.) Limpr.	1043	89.1 ± 6.1	3369	138.3 ± 11.2	2.07	1.72
Çayeli (Kaptanpaşa)	*Parmelia caperata* (L.) Hale	*Rhynchostegiella* sp. (Schimp.) Limpr.	710	100.8 ± 8.7	1013	265.4 ± 20.5	2.22	2.09
Çayeli (Büyükköy)	*Parmelia caperata* (L.) Hale	*Rhynchostegiella* sp. (Schimp.) Limpr.	6153	760.2 ± 36.4	2151	128.6 ± 6.7	1.59	1.84
Çayeli (Madenli)	*Parmelia caperata* (L.) Hale	*Rhynchostegiella* sp. (Schimp.) Limpr.	8565	349.4 ± 23.6	9398	625.5 ± 35.4	1.52	1.50
Rize (Akpınar)	*Parmelia caperata* (L.) Hale	*Thuidium delicatulum* (Hedw.) Schimp.	20665	654.5 ± 38.6	3381	335.1 ± 22.3	1.36	1.72
Rize (Veliköy)	*Parmelia caperata* (L.) Hale	*Homalothecium* sp. Schimp.	9912	543.3 ± 31.0	1521	116.2 ± 8.6	1.49	1.95
Rize (Salarha)	*Parmelia caperata* (L.) Hale	*Brachythecium* sp. Schimp.	3930	236.4 ± 20.1	7022	230.5 ± 21.3	1.69	1.56
Rize (Küçükköy)	*Parmelia caperata* (L.) Hale	*Hypnum cupressiforme* Hedw.	1455	355.2 ± 22.6	7146	264.6 ± 21.3	1.96	1.55
Derepazarı (Korona)	*Parmelia caperata* (L.) Hale	*Thuidium tamariscinum* (Hedw.) Schimp.	2049	315.7 ± 23.4	4756	197.3 ± 11.5	1.86	1.64
Kendirli	*Parmelia caperata* (L.) Hale	*Rhynchostegiella* sp. (Schimp.) Limpr.	1026	763.2 ± 37.6	10224	1353.2 ± 52.4	2.08	1.48
İyidere	*Parmelia caperata* (L.) Hale	*Homalothecium lutescens* (Hedw.) H. Rob.	480	131.6 ± 9.3	1299	244.8 ± 19.5	2.39	2.00
Kalkandere (Y. Başıköyü)	*Parmelia caperata* (L.) Hale	*Plagiomnium undulatum* (Hedw.) T.J.Kop.	2152	47.4 ± 4.8	1020	106.2 ± 7.6	1.84	2.08
İkizdere (Çağrankaya)	*Usnea flipendula*	*Hypnum sp.* Hedw.	648	320.3 ± 21.6	3330	266.1 ± 19.8	2.26	1.73
Of (Kadınlaryaylası)	*Usnea flipendula*	*Dicranum scoparium* Hedw.	3515	214.1 ± 22.3	1062	190.5 ± 11.2	1.71	2.07
Of (Bölümlü)	*Usnea flipendula*	*Sphagnum cuspidatum* Ehrh. ex Hoffm.	2984	87.4 ± 7.9	1343	96.4 ± 6.3	1.76	1.99
Of (Klavuz)	*Usnea flipendula*	*Isothecium myurum* Brid.	715	65.2 ± 7.5	1088	121.0 ± 9.2	2.22	2.06
Çaykara (Uzuntarla)	*Parmelia caperata* (L.) Hale	*Hypnum sp.* Hedw.	172	267.3 ± 20.4	2749	392.1 ± 25.0	2.96	1.78
Çaykara (Çamlıbel)	*Laboria pulmonaria*	*Isothecium myurum* Brid.	246	101.2 ± 7.8	1766	107.2 ± 7.6	2.74	1.90
Çaykara (Uzungöl)	*Usnea florida*	*Isothecium myurum* Brid.	234	290.1 ± 23.5	6693	537.5 ± 26.4	2.77	1.57
Sürmene (Taşlıyayla)	*Usnea florida*	*Brachythecium starkei* (Brid.) Schimp.	3066	340.5 ± 22.7	996	219.1 ± 19.8	1.75	2.09
Sürmene (Büyükdoğanlı)	*Usnea rigida*	*Eurhynchium pulchellum* (Hedw.) Jenn.	3108	219.2 ± 21.8	1469	264.7 ± 23.5	1.75	1.96
Sürmene (Ormanseven)	*Parmelia sulcata*	*Dicranum scoparium* Hedw.	516	306.6 ± 22.3	5376	167.3 ± 11.2	2.36	1.61
Araklı (Pazarcık)	*Parmelia sulcata*	*Isothecium myurum* Brid.	406	270.4 ± 23.7	1105	132.8 ± 9.7	2.47	2.05
Araklı (Taştepe)	*Usnea florida*	*Isothecium myurum* Brid.	3150	319.2 ± 19.4	1377	125.1 ± 7.6	1.74	1.98

Table 2 *(Continued)*

Sampling site	Scientific name		^{137}Cs activity concentration (Bq kg^{-1} in dry weight)				Ecological half-life, T_e (year)	
	Lichen	Moss	Lichen		Moss		Lichen	Moss
			1993[a]	Present work	1993[b]	Present work		
Araklı	*Usnea flipendula*	*Homalothecium lutescens* (Hedw.) H. Rob.	1450	31.5 ± 4.2	1421	99.3 ± 6.5	1.96	1.97
Araklı (Dağbaşı)	*Xanthoria parietina* (L.) Th. Fr.	*Dicranum scoparium* Hedw.	446	125.2 ± 9.8	512	93.2 ± 5.8	2.42	2.36
Araklı (Oğulkaya)	*Parmelia caperata* (L.) Hale	*Homalothecium lutescens* (Hedw.) H. Rob.	3067	312.3 ± 24.3	1762	170.4 ± 19.6	1.75	1.90
Arsin (Çubuklu)	*Parmelia caperata* (L.) Hale	*Eurhynchium pulchellum* (Hedw.) Jenn.	959	194.8 ± 20.1	2297	262.2 ± 24.0	2.10	1.83
Yomra (Gülyurdu)	*Parmelia caperata* (L.) Hale	*Eurhynchium pulchellum* (Hedw.) Jenn.	747	29.3 ± 5.3	681	86.2 ± 5.2	2.20	2.24
Maçka (Zarişa)	*Usnea flipendula*	*Eurhynchium pulchellum* (Hedw.) Jenn.	2752	370.1 ± 24.7	1850	119.1 ± 7.3	1.78	1.89
Maçka (Ormaniçi)	*Usnea flipendula*	*Brachythecium starkei* (Brid.) Schimp.	611	94.3 ± 6.6	1181	94.5 ± 5.1	2.28	2.03
Maçka (Esiroğlu)	*Usnea flipendula*	*Isothecium myurum* Brid.	857	128.4 ± 8.7	2130	230.3 ± 22.7	2.15	1.85
Maçka (Sümela)	*Usnea flipendula*	*Plagiomnium undulatum* (Hedw.) T.J.Kop.	725	214.5 ± 21.3	1236	235.8 ± 23.5	2.21	2.02
Trabzon (Yeşilyurt)	*Parmelia sulcata*	*Brachythecium starkei* (Brid.) Schimp.	1235	108.1 ± 7.2	1981	183.4 ± 18.6	2.02	1.87
Trabzon (Akyazı)	*Usnea flipendula*	*Rhynchostegiella* sp. (Schimp.) Limpr.	1477	218.7 ± 21.2	1004	162.1 ± 10.6	1.96	2.09
Trabzon (Kireçhane)	*Parmelia caperata* (L.) Hale	*Isothecium myurum* Brid.	228	56.2 ± 5.3	1027	66.5 ± 6.7	2.78	2.08
Akçaaabat (Akçaköy)	*Parmelia caperata* (L.) Hale	*Eurhynchium pulchellum* (Hedw.) Jenn.	1980	607.5 ± 32.2	1029	203.4 ± 18.1	1.87	2.08
Akçaabat (Karasu)	*Parmelia caperata* (L.) Hale	*Pleurozium schreberi* (Willd. ex Brid.) Mitt.	799	186.1 ± 19.3	765	106.2 ± 7.8	2.17	2.19
Akçaabat (Çalköy)	*Usnea rigida*	*Eurhynchium pulchellum* (Hedw.) Jenn.	3418	570.6 ± 34.6	3703	661.3 ± 35.0	1.72	1.70
Akçaabat (Karaçayır)	*Usnea rigida*	*Brachythecium starkei* (Brid.) Schimp.	322	210.2 ± 20.5	1610	317.2 ± 34.6	2.59	1.93
Tonya (Yakçukur)	*Usnea flipendula*	*Rhynchostegiella* sp. (Schimp.) Limpr.	2838	790.7 ± 38.6	2879	964.8 ± 45.8	1.77	1.77
Tonya (Rosoda)	*Parmelia caperata* (L.) Hale	*Hypnum sp.* Hedw.	350	125.4 ± 9.8	203	101.6 ± 6.5	2.54	2.85
Tonya (Çayıriçi)	*Parmelia caperata* (L.) Hale	*Pleurozium schreberi* (Willd. ex Brid.) Mitt.	1902	660.3 ± 34.7	2937	1003.2 ± 48.3	1.88	1.76
Şalpazarı (Karakaya)	*Parmelia caperata* (L.) Hale	*Hypnum sp.* Hedw.	3418	831.5 ± 40.3	4064	982.4 ± 46.3	1.72	1.68
Mean	–	–	3159.18	329.5 ± 23.9	4104.10	362.4 ± 24.9	1.97	1.82

[a] Data from Orhan Uzun (Doğu Karadeniz likenlerinin gama aktifliğinin incelenmesi, Master Thesis).
[b] Data from Ali Azar (Doğu Karadeniz karayosunularının gama aktifliğinin incelenmesi, Master Thesis).

Table 3
Ecological half-lives of ^{137}Cs in lichen and moss species

Scientific names		Ecological half-life T_e (years)	
Moss	Lichen	Moss	Lichen
Hypnum cupressiforme Hedw.	*Parmelia caperata* (L.) Hale	1.56	1.93
Hypnum sp. Hedw.	*Parmelia sulcata*	1.83	1.88
Pleurochaete squarrosa (Brid.) Lindb.	*Usnea flipendula*	1.6	2.02
Thuidium tamariscinum (Hedw.) Schimp.	*Usnea rigida*	1.73	2.15
Pseudoscleropodium sp. M. Fleisch.	*Laboria pulmonaria*	1.42	2.74
Homalothecium lutescens (Hedw.) H. Rob.	*Usnea rigida*	1.9	1.75
Brachythecium starkei (Brid.) Schimp.	*Usnea florida*	1.79	2.08
Brachythecium sp. Schimp.	*Xanthoria parietina* (L.) Th. Fr.	1.58	2.42
Isothecium striatulum (Spruce) Kindb.		1.58	
Isothecium myurum Brid.	–	1.93	–
Rhynchostegiella sp. (Schimp.) Limpr.	–	1.83	–
Thuidium delicatulum (Hedw.) Schimp.	–	1.72	–
Homalothecium sp. Schimp.	–	1.95	–
Plagiomnium undulatum (Hedw.) T.J. Kop.	–	2.05	–
Dicranum scoparium Hedw.	–	2.07	–
Sphagnum cuspidatum Ehrh. ex Hoffm.	–	1.99	–
Eurhynchium pulchellum (Hedw.) Jenn.	–	1.95	–
Pleurozium schreberi (Willd. ex Brid.) Mitt.	–	1.48	–

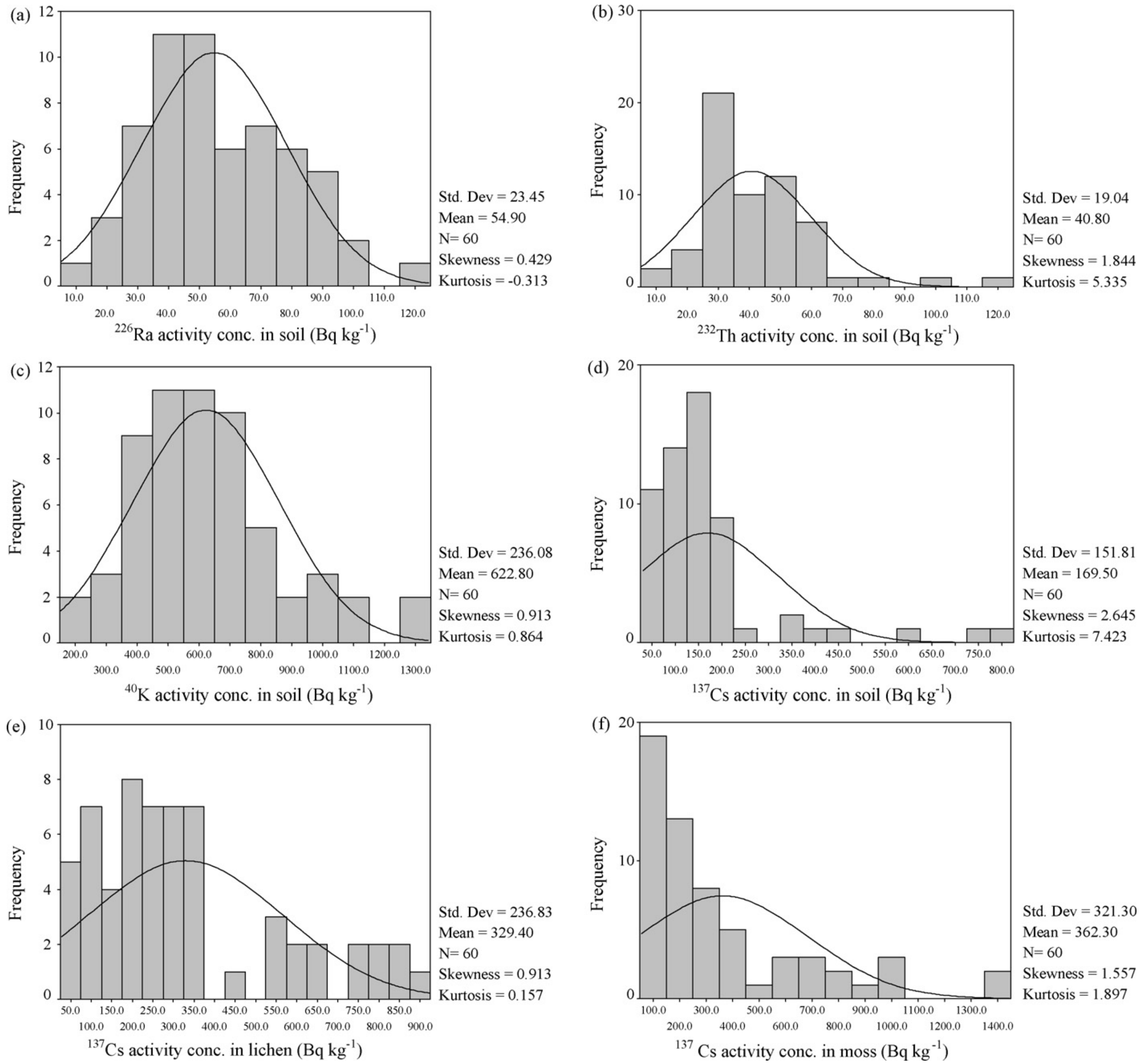

Fig. 2. Frequency distributions of radionuclides for (a) ^{226}Ra; (b) ^{232}Th; (c) ^{40}K; (d) ^{137}Cs in soil; (e) ^{137}Cs in lichen; (f) ^{137}Cs in moss samples.

radionuclide of ^{137}Cs ranged from 38.54 to 139.16 with the average value of 77.18 nGy h^{-1} and from 0.81 to 23.25 with the mean value of 5.09 nGy h^{-1}, respectively (Table 1).

To determine the biological hazard to which an individual is exposed, Gy was converted to Sv taking into account the conversion factor for the biological effectiveness of the dose in causing damage in human tissue recommended by The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and the occupancy factor of 0.2 [1,18] that specifies the proportion of the total time spent outdoors. In the present work, while the annual effective dose due to the natural radionuclides ranged between 46.59 and 168.22, with 93.30 μSv year^{-1} as average value; the annual effective dose due to the ^{137}Cs ranged between 0.98 and 28.11 with the mean value of 6.15 μSv year^{-1} (Table 1). As can be seen from the table, the annual effective dose received due to the exposure to natural radionuclides is relatively higher compared to the annual effective dose due to ^{137}Cs. The mean value of 93.30 μSv year^{-1} (due to natural radionuclides) is higher than the worldwide annual effective dose value of 70 μSv year^{-1} reported by UNSCEAR [1].

3.1.2. Activity in moss and lichen species and ecological half-life of ^{137}Cs

Sixty lichen (8 different species) and sixty moss (19 different species) samples, their scientific names, ^{137}Cs activity concentrations and ecological half-lives of ^{137}Cs in lichen and moss samples are presented in Table 2. It was observed that ^{137}Cs activity concentration ranged from 29 to 879 Bq kg^{-1} (average value is 329 Bq kg^{-1}) for lichen and from 67 to 1396 Bq kg^{-1} (average value is 326 Bq kg^{-1}) for moss samples.

Ecological half-life (T_e) is the time required for a given contaminant concentration to decrease by 50% as a result of physical,

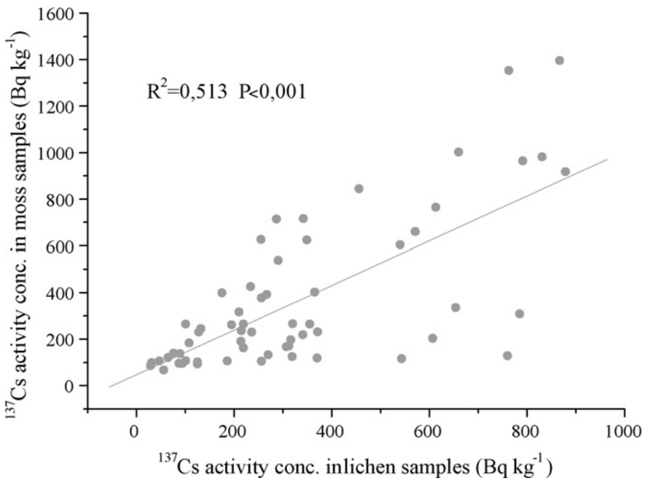

Fig. 3. ^{137}Cs activity concentration (Bq kg^{-1}) in moss and lichen samples.

chemical, and/or biological processes that remove it from an ecosystem or render it biologically unavailable. It is a useful measure for studying the long-term decline of contaminants, such as radionuclides, in natural system and studied by many researchers [19–22]. Ecological half-life of ^{137}Cs was estimated for each species as $T_e = \ln 2/\lambda_e$, where λ_e is the ecological loss-rate constant for ^{137}Cs in that species. Estimates of λ_e were obtained from the slope of the natural-log regression of ^{137}Cs versus time. ^{137}Cs values from the current investigation and historical data reported by previous investigators [8,9] were used in the estimation of T_e's for each species.

The decrease of the activity concentrations in the present measurements (2007) relative to those in 1993 (over a period of 14 years) indicated ecological half-lives between 1.36 and 2.96 years (the mean value is 1.97 years) for lichen and between 1.35 and 2.85 years (the mean value is 1.82 years) for moss species. The reduction of the average activity concentrations over a period of 14 years was similar in all sampling sites, suggesting that ecological clearance mechanisms did not vary substantially over the whole sampling area.

Ecological half-lives of ^{137}Cs in moss and lichen species are summarized in Table 3. As can be seen from the table, while *Dicrranum*

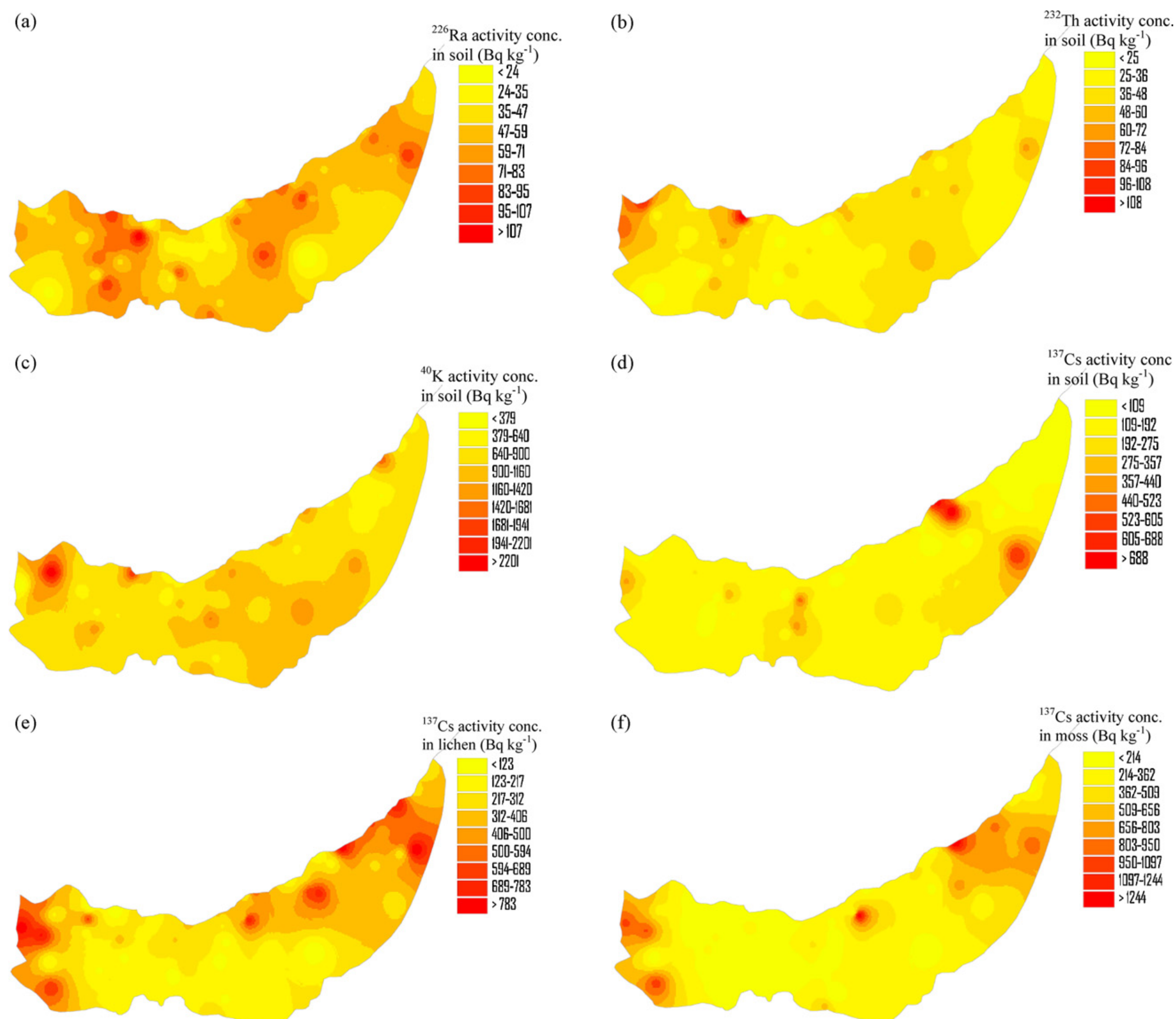

Fig. 4. Interpolated maps of the studied area for (a) ^{226}Ra; (b) ^{232}Th; (c) ^{40}K; (d) ^{137}Cs in soil; (e) ^{137}Cs in lichen; (f) ^{137}Cs in moss samples.

scoparium and *Laboria pulmonaria* showed the longest ecological half-lives; *Preudosceropedium* sp. and *Usnea rigida* showed the shortest ecological half-lives for moss and lichen species, respectively.

However, within a given sampling site, ^{137}Cs activity concentrations vary substantially even among the same lichen and moss species and within a very limited area. This introduces some uncertainty in the estimation of the ecological half-lives.

3.2. Statistical analysis and interpolated maps

SPSS computer software was used to compute ordinary statistics. For the collected samples, the frequency distributions of ^{137}Cs and the naturally occurring radionuclides of ^{226}Ra, ^{232}Th and ^{40}K activities are represented in Fig. 2(a)–(f) and their fits are plotted considering the skewness and the kurtosis coefficients which show the distribution of variables whether it obeys the normal or lognormal law. It was found that ^{137}Cs activity concentration distributions in soil, lichen and moss samples follow approximately log-normal pattern (Fig. 2). On the other hand the activity distributions of naturally occurring radionuclides of ^{226}Ra, ^{232}Th and ^{40}K in soil samples are approximately normal (Fig. 2). The skewness and the kurtosis coefficients for frequency distribution of naturally occurring radionuclides of ^{226}Ra, ^{232}Th,and ^{40}K in soil samples are 0.429, −0.313; 1.844, 5.336 and 0.913, 0.864, respectively. The same coefficients for distributions of ^{137}Cs in soil, lichen and moss samples are 2.645, 7.423; 0.913, 0.157 and 1.557, 1.897, respectively. All distributions have positive skew. The positive values of the skewness coefficients indicate that the distributions are asymmetric with the right tail longer than the left tail. The low kurtosis coefficients suggest that the distribution is close the normal.

The Pearson's correlation analysis showed a strong correlation between ^{137}Cs activity concentrations in lichen and moss samples with the significant level of 0.001 and $R^2 = 0.513$. This correlation is shown in Fig. 3. However no correlation has been found between ^{137}Cs activity concentration in soil, and ^{137}Cs concentration in moss and lichen samples.

The results obtained are shown in Fig. 4(a)–(f) as interpolated maps created by ArcView GIS with the Kriging gridding method. This method determines a value at each grid node based on the *XYZ* data. It first calculates a variogram of the data, which shows the correlation of the data as a function of distance. The mean of the geostatistical analysis in this study is the estimation of the activity concentrations for locations within the area that were not sampled. The interpolated maps shows that while the ^{226}Ra activity concentration distributed over the studied area quite uniformly (Fig. 4(a)), the highest activity concentrations of ^{232}Th and ^{40}K have been recorded in the west of the studied area (Fig. 4(b) and (c)). On the other hand the activity concentrations of ^{137}Cs lichen and moss samples have been observed in the east and the west of the studied area (Fig. 4(e) and (f)). The activity concentrations of ^{137}Cs in soil samples show the highest concentration in the east of the studied area (Fig. 4(d)).

4. Conclusions

Natural and artificial radioactivity measurements have been performed in soil, lichen and moss samples collected from the Eastern Black Sea region of Turkey. The results showed that ^{137}Cs is still eminent in the region of interest. This artificial radioactivity may have different origins: nuclear weapon and bomb tests and the well-known Chernobyl accident. Although the lack of data related to ^{137}Cs activity concentration level in the region of interest before the Chernobyl event makes the evaluation of the above mentioned origin's respective contributions quite difficult, it seems to be quite obvious that the Chernobyl event is the best candidate. We readily deduce that ^{137}Cs activity concentration due to the Chernobyl accident has added up to that global fallout.

The results showed that the activity concentration of ^{137}Cs in soil, moss, and lichen samples follow the descending order $A_{\text{Cs-137moss}} > A_{\text{Cs-137lichen}} > A_{\text{Cs-137soil}}$. The decrease in activity concentration from moss to soil could be explained by the factors such as removal from soil by washing-off, continuing fixation of radionuclides in plants.

Acknowledgement

This work was supported by Karadeniz Technical University under the project number 2005.111.001.1.

References

[1] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Exposures from Natural Radiation Sources, UN, New York, 2000.
[2] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Sources and Biological Effects, UN, New York, 1982.
[3] A. Kulan, Seasonal ^{7}Be and ^{137}Cs activities in surface air before and after the Chernobyl event, J. Environ. Radioactiv. 90 (2006) 140–150.
[4] Turkish Atomic Energy Authority (TAEK), Türkiye'de Çernobil Sonrasi Radyasyon ve Radyasyon Calismalari, TAEK Report, April 1988 (in Turkish).
[5] M.Y. Unlu, S. Topcuoğlu, R. Kucukcezzar, A. Varinlioglu, N. Gungor, A.M. Bulut, E. Gungor, Natural effective half-life of ^{137}Cs in tea plants, Health Phys. 65 (1995) 94–99.
[6] A. Gedikoglu, B.L. Sipahi, Chernobyl radioactivity in Turkish tea, Health Phys. 56 (1989) 11–97.
[7] A.Z. Saka, U. Cevik, E. Bacaksız, A.I. Kopya, E. Tıraşoğlu, Levels of cesium radionuclides in lichens nd mosses from the province of Ordu in the Eastern Black Sea area of Turkey, J. Radioanal. Nucl. Chem. 222 (1997) 87–92.
[8] A. Azar, Doğu Karadeniz karayosunlarının gama aktifliğinin ölçülmesi, Master Thesis (in Turkish).
[9] O. Uzun, Doğu Karadeniz Bölgesi liklerinin gama aktifliğinin ölçülmesi, Mater Thesis (in Turkish).
[10] D.H. Vitt, J.E. Marsh, R.G. Bovey, Mosses, Lichens and Ferns of Northwest North America, University of Washington Press, 1988.
[11] W. Frey, J.P. Fram, E. Fischer, W. Lobin, Die Moos und Farnpflanzen Europas, Stuttgart, Jena, New York, 1995.
[12] A.J.E. Smith, The Moss Flora of Britain and Ireland, Cambridge University Press, London, 2004.
[13] O.W. Purvis, B.J. Coppins, D.L. Hawksworth, P.W. James, D.M. Moore, The Lichen Flora of Great Britain and Ireland, Natural History Museum and British Lichen Society, London, 1993, pp. 710.
[14] C. Scheidegger, A revision of European *Soxicolous* species the genus *Buellia* de not and formerly included genera, Lichenologist 25 (1993) 315–365.
[15] International Atomic Energy Agency, Reference Material, IAEA-375, 2000.
[16] International Atomic Energy Agency, Reference Material, IAEA-156, 2000.
[17] N. Jibiri, I.P. Farai, S.K. Alausa, Estimation of annual effective dose due to natural radioactive elements in ingestion of foodstuffs in tin mining area of Jos-Plateau, Nigeria, J. Environ. Radioactiv. 94 (2007) 31–40.
[18] Turkish Atomic Energy Authority (TAEK), Türkiye için doz değerlendirmeleri, TAEK Report, June 2007.
[19] M.H. Paller, J.M. Littrel, E.L. Peters, Ecological half-lives of ^{137}Cs in fishes from the Savannah River site, Health Phys. 77 (1999) 392–402.
[20] J.D. Peles, M.H. Smith, I.L. Bristin, Eological half-life of ^{137}Cs in plants associated with a contaminated stream, J. Environ. Radioactiv. 59 (2002) 169–178.
[21] G. Prohl, S. Ehlken, I. Fiedler, G. Kirchner, E. Klemt, G. Zibold, Ecological half-lives of ^{90}Sr and ^{137}Cs in terrestrial and aquatic ecosystems, J. Environ. Radioactiv. 91 (2006) 41–72.
[22] P. Machart, W. Hofmann, R. Turk, F. Steger, Ecological half-life of ^{137}Cs in lichens in an alphine region, J. Environ. Radioactiv. 97 (2007) 70–75.