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Seasonal Variations of Radioactivity Concentrations in Soil and Sediment of Meriç River, Turkey

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Abstract

In this study, the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs radionuclides were determined for spring, summer, autumn and winter in soil and sediment samples collected from different sites in the Meriç River using a gamma spectrometer with an HPGe detector. The results showed that the mean activity concentrations in the soil samples were 45.26±1.80 Bq kg^{-1}, 56.24±1.87 Bq kg^{-1}, 704.32±10.82 Bq kg^{-1}, and 2.41±0.54 Bq kg^{-1} for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, respectively. The mean ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs activities in the sediment samples were 25.99±0.73 Bq kg^{-1}, 31.05±0.79 Bq kg^{-1}, 658.03±6.27 Bq kg^{-1}, and 2.71±0.24 Bq kg^{-1}, respectively. The mean radioactivity levels in this study were compared with other activity concentrations in various regions' soils and sediments and also with the world mean values. The mean activity concentrations of ^{40}K were found to be higher than the world mean value in both soil and sediment samples. To estimate the potential health risk in samples, radiological hazard parameters such as the radium equivalent activity, the absorbed dose rate, the annual effective dose equivalent, and the external hazard index were calculated for samples and compared with the recommended values.

1. Introduction

All living organisms are continually exposed to background radiation that comes from radioactive sources [1]. Background radiation is natural ionizing radiation from different sources in the environment. There are various sources of background radiation, including food and drinks, rocks in the ground, radon gas and cosmic rays. The composition of the earth's crust is a major source of natural radiation. The natural sources are mainly due to the primordial radionuclides such as ^{238}U and ^{232}Th and their decay products, as well as ^{40}K [2]. Rock, sand, sediments, soil, water, quarry products and other materials used in building constructions contain various concentrations of radioactivity. Natural radioactivity concentrations differ for each region depending on the geological structure. Since natural radionuclides are not homogeneously distributed across regions, their concentration and analysis of their distribution in materials play an important role in radiation protection [3]. Major routes of exposure to radionuclides include external exposure from radionuclide deposits in river and marine sediments and consumption of foodstuffs [4].

In addition to natural radionuclides, artificial radionuclides are released into the atmosphere due to anthropogenic activities such as nuclear power plant accidents, nuclear weapon tests, medical applications, and industrial activities [5]. One of the most important artificial radionuclides is the anthropogenic radionuclide ^{137}Cs (half-life 30.2 years) which is a product of radioactive fallout. ^{137}Cs radionuclide reached Turkey with radioactive clouds as a result of the Chernobyl disaster in 1986. The Thrace and Black Sea regions of Turkey were highly contaminated after the Chernobyl nuclear power plant accident. ^{137}Cs radionuclide is strongly absorbed and retained in the soil and is soluble in water [5]. Knowledge of the level of natural and artificial radioactivity in river and

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coastal ecosystems is important because rivers transport particulate materials and dissolved species from land to sea. Natural and artificial radionuclides migrate from the lithosphere to wider natural ecosystems in a variety of ways, including erosion of terrestrial rocks and subsequent transport by water, wind, and gravity [4], [6]. Long-term exposure to radionuclides has several health problems such as lung diseases, acute leucopenia, anemia, and cancers [7].

The objective of this study is to obtain seasonal radioactivity levels of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in soil samples collected from different sites across the Meriç River and in the sediment samples collected from various sites within the Meriç River. Radiological parameters were calculated to evaluate potential health hazards. The data in this study will provide background data on environmental pollution.

2. Material and Method

2.1. Study Area

The samples were collected seasonally from different sites in the Meriç River. The Meriç River is the longest river in the Balkans (about 480 km long) and also has an important aquatic ecosystem in the Thrace region in Turkey [8], [9]. The Meriç River Basin covers an area of approximately 14.600 km^2 in Turkey [8]. The Meriç River comes from Bulgaria and flows along the border between Greece and Turkey. 187 km of the Meriç River is located in Turkey [10]. The samples were collected in the areas of the Meriç River in Edirne, Turkey. The location of Edirne province in Turkey and also the Meriç River is shown in Figure 1.

2.2. Sample Preparation and Activity Analysis

20 measurement sites were selected along the Meriç River, where measurements were carried out for one year. The survey of terrestrial gamma radiation was performed once during each season: autumn, winter, spring and summer. One soil and sediment sample was collected from each point in one season. The samples were collected from the surface layer (8-10 cm). At each sampling point, approximately 1 kg of sediment and 500 g of soil were collected. The soil and sediment samples were dried at 105 °C for more than a day. Impurities such as stones, grass, and weeds in the samples were removed and passed through a 2 mm mesh. The samples were finally placed in 250 ml Marinelli containers for more than 4 weeks to achieve secular equilibrium between ^{226}Ra and its decay products [12]. Sample preparation pictures are given in Figure 2. The net masses of the soil and sediment samples in the Marinelli containers were about 200-250 g.

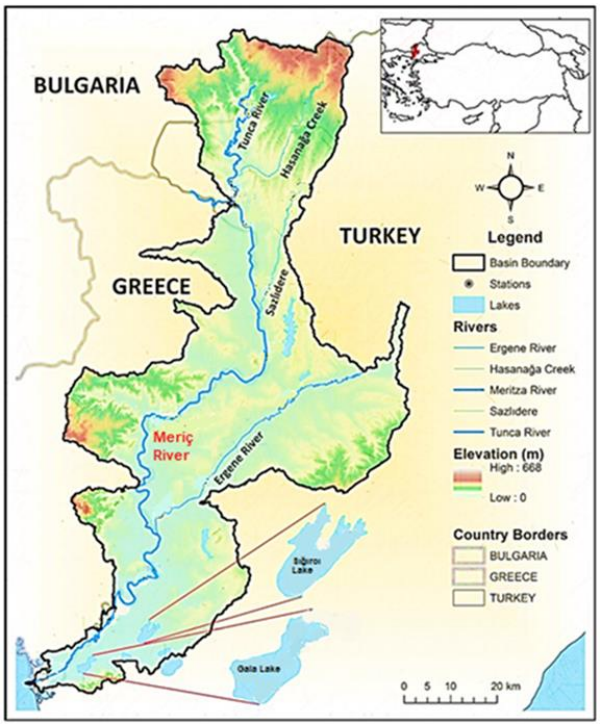

Figure 1. Meriç Basin Map [11]

Radionuclide analysis was obtained using a gamma-ray spectrometer equipped with a high-purity germanium (HPGe) detector (Ortec GEM70P4-95, 70% relative efficiency) in Kırklareli University Central Research Laboratory. A solid nuclide mixture of gamma reference calibration source from Isotope Product Laboratories (Eckert&Ziegler, Berlin, Germany) containing the radionuclides ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co was used for the energy and efficiency calibrations. The spectra collection and the analysis of the spectra were obtained by GammaVision-32 software and Maestro software, respectively. The activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K were determined using the gamma-ray peaks of the 911.2 keV (^{228}Ac) and 583.1 keV (^{208}Tl), 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi), and 1460.8 keV, respectively. The activity concentration of ^{137}Cs was evaluated from the gamma-ray peak at 661.66 keV.

The activity concentrations in the samples were calculated by Eq. 1.

$$A = \frac{C}{\varepsilon x I_{\gamma} x m} \qquad (1)$$

In Eq. 1, A is the activity in Bq kg^{-1}, C is counts (per second), ε is the detector efficiency, Iγ is the gamma-ray emission probability, and m is the mass of the soil and sediment sample in kg [13], [14].

Figure 2. Sample preparation pictures

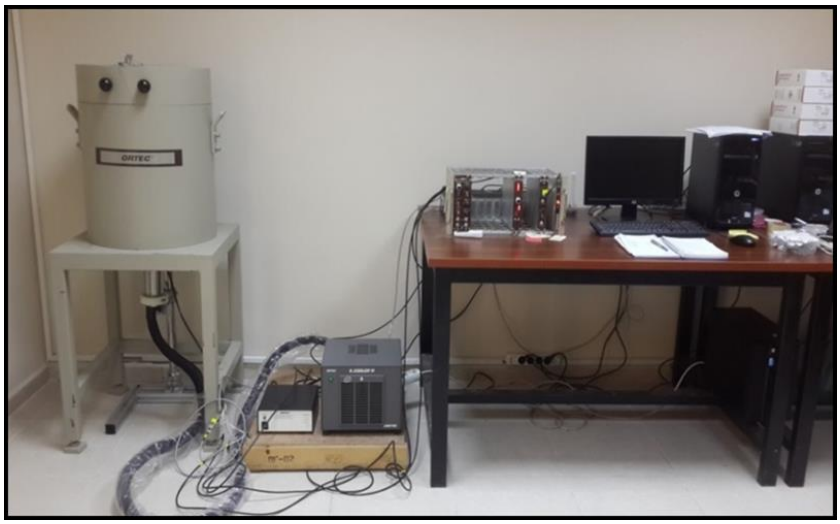

Figure 3. Gamma spectrometer with High Purity Germanium (HPGe) detector in Kırklareli University Central Research Laboratory

2.3. Calculation of Radiological Hazards

Radiological hazard parameters were determined using Eq. 2, 3, 4, and 5. In equations, Ra_{eq} is radium equivalent activity, C_{Ra} is activity concentration of ^{226}Ra, C_{Th} is activity concentration of ^{232}Th, C_K is activity concentration of ^{40}K, C_{Cs} is activity concentration of ^{137}Cs, D is the total absorbed dose rate in air at l m above ground level, AEDE is the annual effective dose equivalent, and H_{ex} is the external hazard index [15], [16].

$$Ra_{eq}(Bq\,kg^{-1}) = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

$$D\,(nGy\,h^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K + 0.03C_{Cs} \quad (3)$$

$$AEDE\,(\mu Sv\,y^{-1}) = D\,(nGy\,h^{-1}) \times 8760\,(h\,y^{-1}) \times 0.2 \times 0.7\,(SvGy^{-1}) \times 10^{-3} \quad (4)$$

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (5)$$

3. Results and Discussion

Activity concentrations in soil and sediments of the Meriç River were measured for four seasons and the mean activity concentrations of radionuclides are summarized in Table 1.

The seasonal variations of the mean activity concentrations of ^{226}Ra and ^{232}Th in samples are given in Figure 4. The mean activity concentrations of ^{226}Ra in soil samples were determined as 43.64±0.70, 35.87±0.77, 41.78±0.62 and 45.26±1.80 Bq kg^{-1} in spring, summer, autumn and winter, respectively. The mean values of ^{232}Th in soil were found as 40.76±1.01 Bq kg^{-1} (in spring), 47.34±0.46 Bq kg^{-1} (in summer), 57.83±2.37 Bq kg^{-1} (in autumn) and 56.24±1.87 Bq kg^{-1} (in winter).

The mean values of ^{226}Ra and ^{232}Th activity concentrations in soil samples were found to be higher than those of the worldwide mean values (35 Bq kg^{-1} for ^{226}Ra and 30 Bq kg^{-1} for ^{232}Th) as reported by UNSCEAR [15]. When we examine the seasonal analysis results of the sediment samples, the mean values of ^{226}Ra activity concentrations were 19.85±0.44, 22.36±0.41, 33.51±1.27 and 28.26±0.79 Bq kg^{-1} in spring, summer, autumn and winter, respectively.

The mean activity concentrations of ^{232}Th in sediment samples were determined as 25.48±0.25 Bq kg^{-1} (in spring), 18.59±0.76 Bq kg^{-1} (in summer), 37.18±1.19 Bq kg^{-1} (in autumn) and 42.95±0.94 Bq kg^{-1} (in winter). The mean activity concentrations of ^{226}Ra in sediment samples were lower than the world average value in all seasons. The mean values of ^{232}Th activity concentrations in sediment samples calculated for the autumn and winter seasons were found to be higher than worldwide mean values [15].

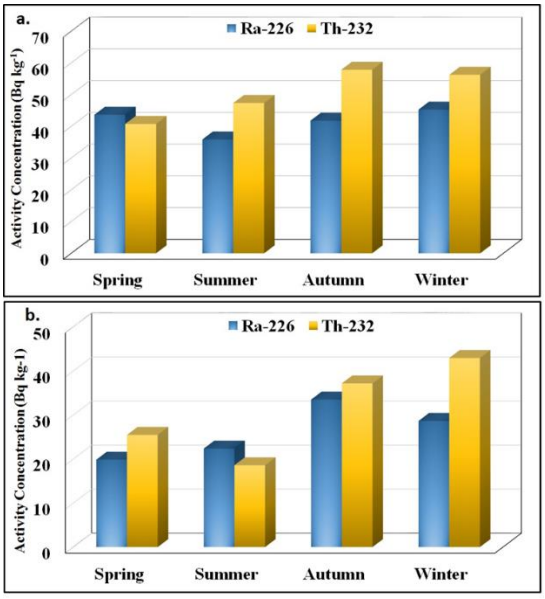

Figure 4. The mean activity concentrations of ^{226}Ra and ^{232}Th in a. soil samples and b. sediment samples

The seasonal variations of the mean activity concentrations of ^{40}K and ^{137}Cs emitted from the samples are presented in Figure 5 and Figure 6, respectively. The highest mean of ^{40}K was found in spring (719.93±5.44 Bq kg^{-1}) in the soil sample. The lowest mean concentration of ^{40}K was obtained in spring (647.23±4.32 Bq kg^{-1}) in the sediment sample. The mean activity concentrations of ^{40}K were found to be higher than the world mean value of 400 Bq kg^{-1} in both soil and sediment samples [15]. Artificial fertilization processes, especially potassium-containing fertilizers, are known to increase radioactivity. In addition, the transport of fertilized soil from one place to another by rivers also increases radioactivity [17-19]. The high activity concentrations of ^{40}K and ^{226}Ra in these areas of the Meriç River Basin may have resulted from over-fertilization.

The highest mean of artificial ^{137}Cs was obtained in autumn (4.34±0.35 Bq kg^{-1}) in the sediment sample. The lowest mean value of ^{137}Cs was found as 1.38±0.25 Bq kg^{-1} (winter) in the sediment sample. Since the Chernobyl accident affected the Thrace region in Turkey, ^{137}Cs radionuclide have been found in both soil and sediment samples.

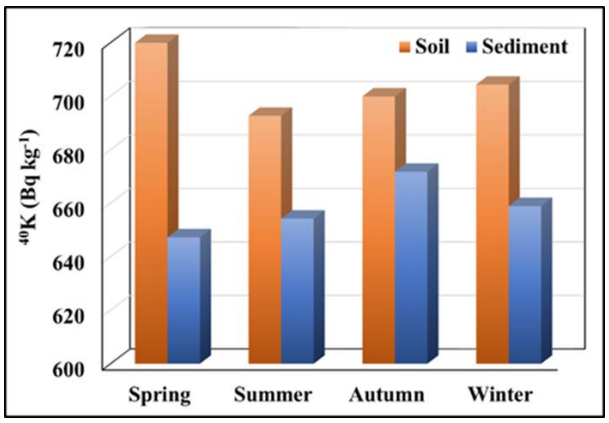

Figure 5. Seasonal variations of the mean activity concentration of ^{40}K

Although soil and sediment gamma activity at some sampling points shows similar activity concentration changes, there are significant variations between winter and summer measurements. As shown in Figures 4a and 5, maximum activity concentrations in soils were generally determined in spring and winter. The frequency and magnitude of floods in the region have increased in recent years. Therefore, the main reason for seasonal variation of radionuclide concentrations in the study area is floods and rainfall regimes. Floods caused the displacement of soils in the study area and the concentrations of radionuclides varied significantly from season to season. It is also thought that potassium fertilizer was used for the soils in the study area and this may have increased the phosphate content of the soil and as a result, disproportionately ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K levels may have accumulated in the soil [20].

Higher values for ^{137}Cs were determined in summer and autumn as a result of dry deposition [21]. Water erosion contributes to the redistribution of washed soil particles in river basins with ^{137}C fixed on them. Therefore, areas of secondary pollution are

formed in river basins where the total inventory of ^{137}Cs exceeds the initial accumulation levels [22]. Humid climates cause the leaching of alkaline metals and soil acidification and can also affect U and Th mobility. Depending on the environmental characteristics (geological factors, climate, human activities, etc.), coprecipitation and sorption in organic matter, clays and oxides may hold radionuclides in soils [23].

Table 1. The mean activity concentrations (Bq kg^{-1}) of radionuclides in soil and sediments of the Meriç River

Sample Type	Season	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Soil	Spring	43.64±0.70	40.76±1.01	719.93±5.44	1.24±0.12
	Summer	35.87±0.77	47.34±0.46	692.67±7.55	3.98±0.35
	Autumn	41.78±0.62	57.83±2.37	699.90±5.86	2.50±0.31
	Winter	45.26±1.80	56.24±1.87	704.32±10.82	2.41±0.54
	Mean	41.64±0.97	50.54±1.43	704.21±7.42	2.53±0.33
Sediment	Spring	19.85±0.44	25.48±0.25	647.23±4.32	2.25±0.11
	Summer	22.36±0.41	18.59±0.76	654.25±4.38	2.86±0.26
	Autumn	33.51±1.27	37.18±1.19	671.73±8.84	4.34±0.35
	Winter	28.26±0.79	42.95±0.94	658.91±7.54	1.38±0.25
	Mean	25.99±0.73	31.05±0.79	658.03±6.27	2.71±0.24

Table 2. Mean activity concentrations of the radionuclides (Bq kg^{-1}) in soil samples from various studies in the literature compared with those of this study

Region	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Reference
Jordan	42	42	23	3.7	[24]
Russia	35	30	400	48.9	[25]
Pakistan	32.9	53.6	647.4	1.5	[26]
Botswana	34.8	41.8	432.7	-	[27]
China	75.1	101.0	535.8	-	[28]
India	60.3	64.5	481.0	-	[29]
North Cyprus	83.7	53.6	593.9	7.1	[30]
Bolu (Turkey)	18.2	17.3	258.3	7.5	[31]
Bartın (Turkey)	8	7	136	2	[32]
Nevşehir(Turkey)	49.45	54.08	698.4	8.26	[33]

Table 3. Mean activity concentrations of the radionuclides (Bq kg^{-1}) in sediment samples from various studies in the literature compared with those of this study

Region	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Reference
Mediterranean Sea coast	20.1	18.4	467.3	-	[35]
Egypt	23.8	19.6	374.9	-	[36]
Iraq	15.48	-	418.47	2.88	[37]
Boka Kotorska Bay	20	35	580	6.1	[38]
Barents Sea	14.2	21.1	439.1	3.2	[39]
Nigeria	-	35.2	501.0	-	[40]
Calabria	21.3	30.3	849	-	[41]
Borçka Black Lake (Turkey)	-	13.85	473.67	35.06	[42]
Izmit Bay (Turkey)	18	-	568	21	[43]
Aegean Sea (Turkey)	21.50	23.13	541.88	-	[44]

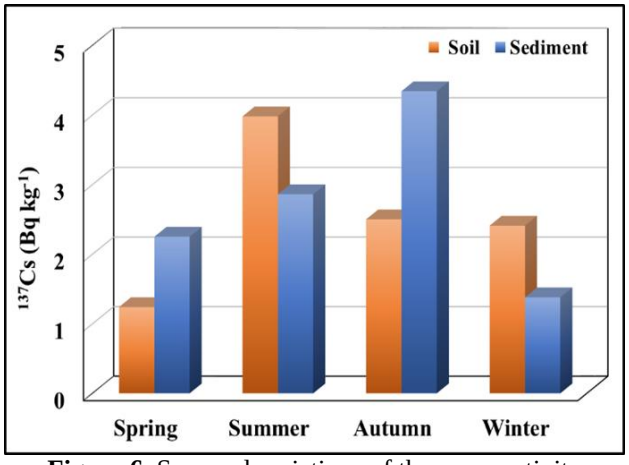

Figure 6. Seasonal variations of the mean activity concentration of artificial ^{137}Cs

The mean activity concentrations calculated in this study were compared with the results obtained in other studies in the literature (Tables 2 and 3). The mean of the activity concentrations for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in soil samples were found as 41.64, 50.54, 704.21, and 2.53 Bq kg^{-1} in this study, respectively. The mean value of the activity concentration of ^{226}Ra in soil samples is less than the values obtained in China, India, North Cyprus, and Nevşehir (Turkey), as illustrated in Table 2. The mean of the activity concentration of ^{232}Th in soil samples is less than Pakistan, China, India, North Cyprus, and Nevşehir (Turkey) and higher than Jordan, Russia, Botswana, Bolu (Turkey) and Bartın (Turkey). Soil and rock characteristics in a region are the most important factors affecting radioactivity.

In Nevşehir (Turkey), there are metamorphic, sedimentary and volcanic rocks. Higher activity concentrations were observed due to the dominance of volcanic rocks in Nevşehir province. The mean of the activity concentrations of ^{40}K in the soil and sediment samples are higher than in the other regions. The mean activity concentration for ^{137}Cs in soil samples in this study is higher than in Pakistan and Bartın (Turkey). The mean concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in sediment samples were in Meriç River (Turkey) obtained as 25.99, 31.05, 685.03, and 2.71 Bq kg^{-1}, respectively. The mean of the activity concentrations of ^{226}Ra and ^{232}Th in sediment samples are higher than Mediterranean Sea coast, Egypt, Iraq, Boka Kotorska Bay, Barents Sea, Izmit Bay (Turkey), and Aegean Sea (Turkey). The mean activity concentration for ^{137}Cs in sediment samples in the present study is less than Iraq, Boka Kotorska Bay, Barents Sea, Borçka Black Lake (Turkey), and Izmit Bay (Turkey). Izmit Bay is located in the most industrialized region of the Marmara region. In Izmit Bay (Turkey), the highest activity concentrations of ^{40}K and ^{226}Ra were found near phosphate, fertilizer and petrochemical industrial facilities and are higher than the values in this study. Borçka Lake (Turkey) has been highly contaminated due to the Chernobyl accident and ^{137}Cs concentrations are very high compared to other lakes in Turkey [42]. The variation of obtained values is due to some factors such as the geology of the region, the area (agricultural or industrial), soil and rock type or other characteristic factors [34].

The Meriç River is a transboundary river and industrial wastes, sewage wastes, and radioactive pollution resulting from agricultural activities are transported by water along this long river. The Bulgarian part of the Meriç River Basin has been used for agricultural activities and uranium mining [45]. These radioactive pollutants, especially those coming to Turkey via the river from Bulgaria, cause an increase in the natural and artificial radionuclides in the soil and sediment along the river in Turkey. Therefore, activity concentrations in soil and sediment samples collected along the Meriç River were observed to be higher than in some regions in Turkey.

Figure 7 shows the variation of radiological hazard parameters in samples. The mean values of Ra_{eq} in sediment samples were found to be 101.59, 94.74, 133.69, and 135.80 Bq kg^{-1} in spring, summer, autumn and winter, respectively. In soil samples, the mean values of Ra_{eq} were 152.32, 152.05, 173.47, and 174.99 Bq kg^{-1} in spring, summer, autumn and winter, respectively. All the values of Ra_{eq} in samp1es were lower than the recommended maximum value of 370 Bq kg^{-1} [16]. The mean values of D in sediment samples were 51.62, 48.93, 66.08, and 66.52 nGy h^{-1} in spring, summer, autumn and winter, respectively. In soil samples, the mean values of D were calculated as 74.84, 74.17, 83.49, and 84.32 nGy h^{-1} in spring, summer, autumn and winter, respectively. The mean values of D in autumn and winter for sediment samples were higher than the world average value (57 nGy h^{-1}) [15]. All the values of D in soil samples were found to be higher than the world average. The obtained values of AEDE due to natural and artificial radioactivity in soil samples ranged from 90.96 to 103.41 μSv y^{-1} with a mean value of 97.14 μSv y^{-1}, which is higher than the world average of 70 μSv y^{-1} [15]. In addition, all the calculated values of H_{ex} for soil and sediment samples were found to be lower than unity which does not cause any harm to the human beings in the studied area.

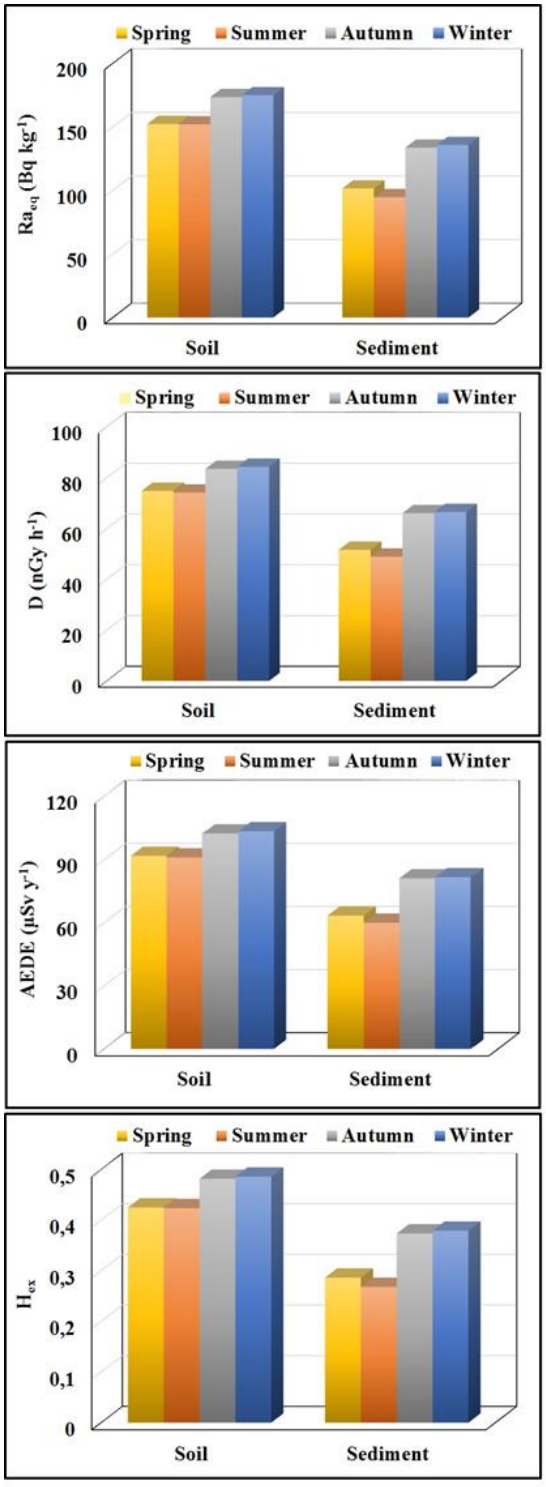

Figure 7. Radiological hazard parameters in soil and sediment samples

4. Conclusion

This study provides a baseline for radioactivity levels in both soil and sediment samples in the Meriç River Basin for spring, summer, autumn and winter. The mean activity concentrations of ^{226}Ra in sediment samples were lower than the world average value for all seasons. The radioactivity concentrations of ^{232}Th in sediment samples for the autumn and winter seasons were higher than in summer and spring. The differences between activity concentrations may be a consequence of variations in meteorological conditions and soil moisture. In addition, the mean activity concentrations of ^{40}K were higher than the world mean value in both soil and sediment samples. The use of artificial fertilizers and their transport via rivers increases radioactivity. ^{137}Cs radionuclide detected in the study area. Since the Chernobyl disaster in 1986 affected the Thrace region of Turkey, ^{137}Cs radionuclide continues to exist in both soil and sediment. To estimate the potential health risk, radiological hazard parameters were calculated. ELCR and D were found higher than the recommended values for some samples. This implies that the population near these areas is likely to receive some radiation dose. Radiological hazard parameters for soil samples were higher than sediment samples. The high-level radiological hazard parameters were due to the high level of radioactivity concentrations of radionuclides in both soil and sediment samples. However, all the calculated values of H_{ex} for samples were found to be lower than unity which does not cause any harm to the human beings in the studied area.

Contributions of the authors

The authors' contributions to the paper are equal

Conflict of Interest Statement

There is no conflict of interest between the authors.

Statement of Research and Publication Ethics

The study is complied with research and publication ethics

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