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ScienceDirect
Journal of Hazardous Materials 157 (2008) 328–334
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Vertical distribution of ^{226}Ra and ^{210}Po in agricultural soils in Buyuk Menderes Basin, Turkey

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Received 29 March 2007; received in revised form 2 January 2008; accepted 2 January 2008
Available online 6 January 2008

Abstract

The vertical distribution of ^{226}Ra and ^{210}Po was investigated in the cultivated soils of the Buyuk Menderes Basin in Turkey. Five soil cores down to a depth of about 50 cm were taken from each site and divided into strata of 2–3 cm intervals. The samples were analyzed for their ^{226}Ra and ^{210}Po activity concentrations using radioanalytical methods. Down-core concentration profiles of ^{226}Ra and ^{210}Po in the soil cores from five sites are obtained. The activity concentrations of soil cores range from 80 to 1170 Bq kg^{-1} for ^{226}Ra and from 10 to 870 Bq kg^{-1} for ^{210}Po with the depth. Analysis of the vertical soil profiles indicate that the activity concentrations of ^{226}Ra and ^{210}Po for soil strata at all the sites have not extremely changed with depth.

Keywords: Radium-226; Polonium-210; Gross alpha activity; Vertical distribution; Soil texture

1. Introduction

Soil is the upper layer of the unsaturated zone of the earth, and very diverse in composition and behaviour. The soil phase consists of mineral particles of various sizes, shapes and organic matter in various stages of degradation. Soil is one of the important components in the evaluation of radionuclide migration behaviour and distribution of uranium series radionuclides in a terrestrial ecosystem. Migration of heavy natural radionuclides has been studied in rocks, drainage water, bottom sediment, soils and plants in different regions by determination the natural distributions of radionuclides [1–4].

The analysis of radium isotopes is of great concern in geochemistry since four radium isotopes (^{223}Ra, ^{224}Ra, ^{226}Ra, ^{228}Ra) of the uranium and thorium decay series can be used as natural geochemical tracers to investigate important processes in the bio- and geospheres [5]. Radium rarely occurs alone and the environmental distribution of radium varies depending on its origin [2]. The behaviour of Ra in soils is important because of the hazard of its daughter products. ^{226}Ra has always attracted particular attention from the view point of its health hazard to the public.

The ^{210}Po in soils may originate either as a product of the radioactive decay of radionuclides of ^{238}U series present in the soil or the result of the precipitation of radon decay products from the atmosphere. ^{210}Po occurs widely in nature and gives an important contribution to man's natural radiation background. The ^{210}Po content of soil varies with soil type. The levels of ^{210}Pb and ^{210}Po contained in the top layer of soil can be correlated with the amount of atmospheric precipitation. In soils, ^{210}Po is in equilibrium with ^{210}Pb, suggesting that the ^{210}Pb in the soil is the main source of ^{210}Po. Parfenov has reported that ^{210}Po is very immobile in soil and that it is likely irreversibly sorbed on clay and organic colloids [6]. Polonium is also a particle reactive element, but its geochemical pathways appear to be different from lead. ^{210}Po behaves more like the nutrient elements [7].

The purpose of this study was to determine the ^{226}Ra and ^{210}Po activity concentrations using radiochemical methods for five core soil samples and to present an overview of their vertical distributions in agricultural soil of the Buyuk Menderes Basin where is one of the most important plain in the Aegean Region of Turkey and has agricultural lands and localities, and the largest river in the Aegean Region flows along the study area. The data were evaluated statistically. Special attention was paid to these sites where known uranium anomalies identified in the West part

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doi:10.1016/j.jhazmat.2008.01.001

of the Basin by the researchers and our previous study [8–10]. The average grade of these uranium deposits in the Basin is ranged from 0.02 to 0.08% U_3O_8.

2. Material and methods

2.1. Study area

The sampling sites in Buyuk Menderes Basin have been selected since it is the one of the most important Basins in Aegean Region, which has the widest agricultural lands under cultivation for a long time, at least for 3000 years [11]. The east–west trends of the Buyuk Menderes Basin appear to have been evolving since the late Pliocene, and this area includes the oldest alluvial sediment. The soil types in the Buyuk Menderes Basin are as following: Alluvial, limeless brown soil, colluvial, reddish Mediterranean soil, regosol, brown forest soil, salty, alkaline and alkaline+salty in the eastern part of the Buyuk Menderes Basin and alluvial, colluvial, limeless brown soil, brown forest soil, rendzina, red-brown Mediterranean soil, regosol in the western part of the Basin [12,13].

The Basin is located in the Aegean region (37°25′ N to 38°10′ N and 27°00′ E to 29°10′ E) in Turkey. There are farming activities in the Basin, and the area covers the agricultural lands about 714.000 ha. The crops grown in this land are cotton, clover, wheat, rye, fig, peach, orange and olive. The Basin also has the largest river (Buyuk Menderes River) of the Aegean Region, which is 450 km in length and passes the Basin in the east–west direction. The river collects 6 rivulets from the Basin and drains out to Aegean Sea.

Soil texture includes the proportions of sand, silt and clay particles in the soil. The terms sand, silt and clay refer to different size fractions of the soil's mineral content. The sizes of clay, silt and sand are <0.002 mm; 0.002–0.06 mm and 0.06–2 mm, respectively. The soil texture of the Buyuk Menderes Basin in our previous study was recorded as 1–13% for clay, 1–42% for silt and 58–99% for sand content. In addition, the organic matter and pH value of soil samples were also determined in the range of 1.2–13.0% and 5.65–8.35, respectively [14].

The main agricultural product at the west part of the Buyuk Menderes Basin where the core samples was taken the core samples is cotton. For soil homogeneity before planting cotton in autumn, the soil was cultivated in 20–25 cm depth to grow the crops.

Cotton production estimated by USDA in this area in 2000/2001 was 1.20 tonnes/ha. The cotton area in this region in 2000/2001 was estimated in the range of 210.000–230.000 ha. Percentage of total cotton production in this region in Turkey was 4–9% [15].

2.2. Sample collection and preparation

The core soil samples were collected at the five sites from cultivated lands of the Buyuk Menderes Basin of Turkey shown in Fig. 1 using a box corer of Ø: 5 cm in June 2000 and divided into strata of between 2- and -3 cm intervals. In addition, uncultivated soil sample was collected from the site far from cultivated lands within the same region in order to compare for the activity of radionuclides. The depth intervals of soil samples varied from 0 to 30 cm for Site-1, Site-3 and Control Site, 0–27.5 cm for Site-2, 0–40 cm for Site-4, 0–50 cm for Site-5.

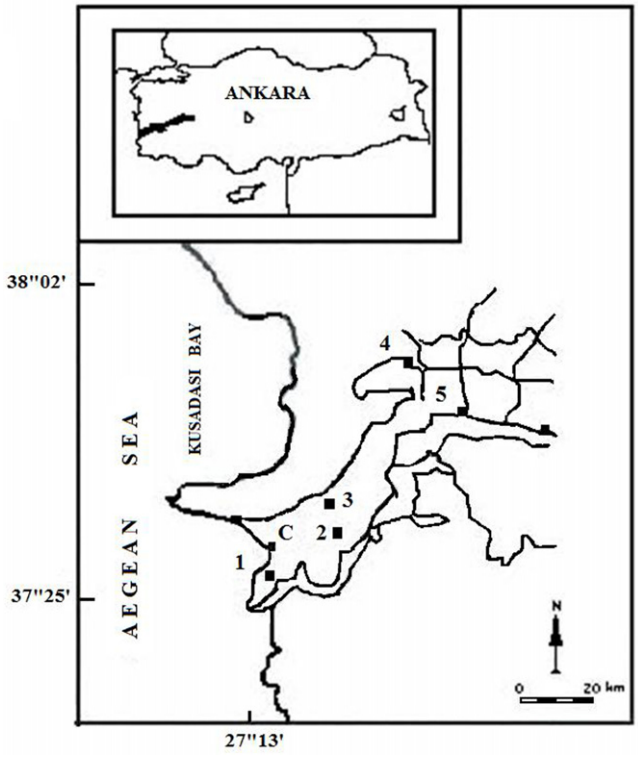

Fig. 1. The sampling sites in the Buyuk Menderes Basin.

The sampling sites coordinates were determined by a Global Positioning System (Garmin, GPS XL-45). The core samples were transferred to the laboratory in pre-cleaned polyethylene vials for further treatment and analyses. The core samples were oven-dried until they achieved a constant dry weight and were prepared in a powdered form of approximately <200 mesh size. The ^{226}Ra and ^{210}Po activities were measured by α-counting technique after the radiochemical processing. The samples were analyzed about 2 years later so that secular equilibrium between ^{210}Pb and ^{210}Po is attained.

All samples were carefully processed following the standard procedures. Soils were well mixed after removing extraneous materials such as roots, pieces of stones and gravel. Each soil sample was first air-dried, and then placed in an oven at 105 °C for 24 h until their weight remains constant [16].

2.3. ^{226}Ra measurements

A conventional radiochemical method was used to determine the activity concentration of ^{226}Ra. All chemicals used for the preparation of solutions were of analytical-reagent grade. After waiting for about 20 days allowing ^{224}Ra to decay, 1 g of soil sample was dissolved in H_2SO_4 and HNO_3 acid mixture. Barium carrier was added to the acid solution. The $Ba(Ra)SO_4$ was precipitated, centrifuged, washed with some H_2SO_4 and distilled water and then dissolved in EDTA. $(NH_4)_2SO_4$ was

used to reprecipitate $Ba(Ra)SO_4$. After filtering and drying, the precipitate on the membrane filter (Whatman 0.45 μm) was counted by a ZnS(Ag) alpha scintillation counter (Eberline SAC-4 Model) [17]. The chemical yield of the radiochemical procedure determined using ^{226}Ra standard radioactive solution in soil matrix was found to be $90 \pm 3\%$. The counting efficiency was determined as $25 \pm 2\%$. A ^{226}Ra standard radioactive stock solution of 370 Bq mL^{-1} was obtained from Turkish Atomic Energy Authority-Cekmece Nuclear Research and Training Centre (CNAEM). Minimum detection limit for ^{226}Ra is 0.08 Bq g^{-1}.

2.4. ^{210}Po measurements

The spontaneous chemical deposition method was employed for the determination of ^{210}Po activity. About 1 g of soil sample was digested in a HNO_3, HCl and HF acid mixture in a Teflon® beaker on the hot plate at 65 °C. For deposition of polonium on copper disc, the following pretreatment procedure was used: the solution having ^{210}Po adjusted to pH 1.2–1.5 was spontaneously deposited onto a copper disc at 75 °C for 6 h after adding ascorbic acid to prevent Fe interference [18,19]. The α-activity of the disc was measured using a ZnS(Ag) scintillation alpha counter (Eberline SAC-4 Model). The counting time was 50 min. Due to the lack of availability of a polonium standard and also because of the fact that $^{210}Po(^{210}Pb)$ occurs in the environmental samples as the decay product of ^{210}Pb, for the efficiency study, a ^{210}Pb standard solution (7.2×10^{-3} Bq mL^{-1}, Baird Atomic), which is in equilibrium with ^{210}Po, was used. The average chemical efficiency of ^{210}Po of this method in soil matrix was found to be as $77 \pm 3\%$. The counting efficiency was determined as $25 \pm 2\%$. Minimum detection limit for ^{210}Po is 0.01 Bq g^{-1}.

3. Results and discussion

The current study was carried out on five selected cultivated sites located in the Buyuk Menderes Basin because the ^{226}Ra and ^{210}Po activity levels were found higher in West of the Basin as compared to inland sites of the Buyuk Menderes Basin reported in our previous study [10,14]. The ^{210}Po activity in uncultivated soil profile showed almost homogeneous distribution downward the soil profile. Ra-226 activity concentrations in each uncultivated soil profile were observed to present a slight increase by depth. The downward core activity concentration profiles of ^{226}Ra and ^{210}Po in five core soils from five sites and one core from uncultivated site were obtained. A representation of the variation in down-core concentrations and distribution of ^{226}Ra and ^{210}Po can be seen from the profiles shown in Fig. 2. The activity concentrations in the soil cores vary between 80 and 1170 Bq kg^{-1} for ^{226}Ra and between 10 and 890 Bq kg^{-1} for ^{210}Po by depth. The activity concentrations in the uncultivated soil core vary between 280 and 400 Bq kg^{-1} for ^{226}Ra and between 50 and 100 Bq kg^{-1} for ^{210}Po.

In general, the activity concentrations of ^{226}Ra and ^{210}Po for soil strata at all the sites have not extremely changed with depth. It is evident that the activity concentrations of ^{226}Ra at all the sites are slightly low in the surface of the soil cores compared to the deeper strata except site-3. ^{226}Ra concentrations increase slightly with increasing depth at site-2, site-4 and site-5. ^{226}Ra and ^{210}Po concentrations are the highest and change very lit-

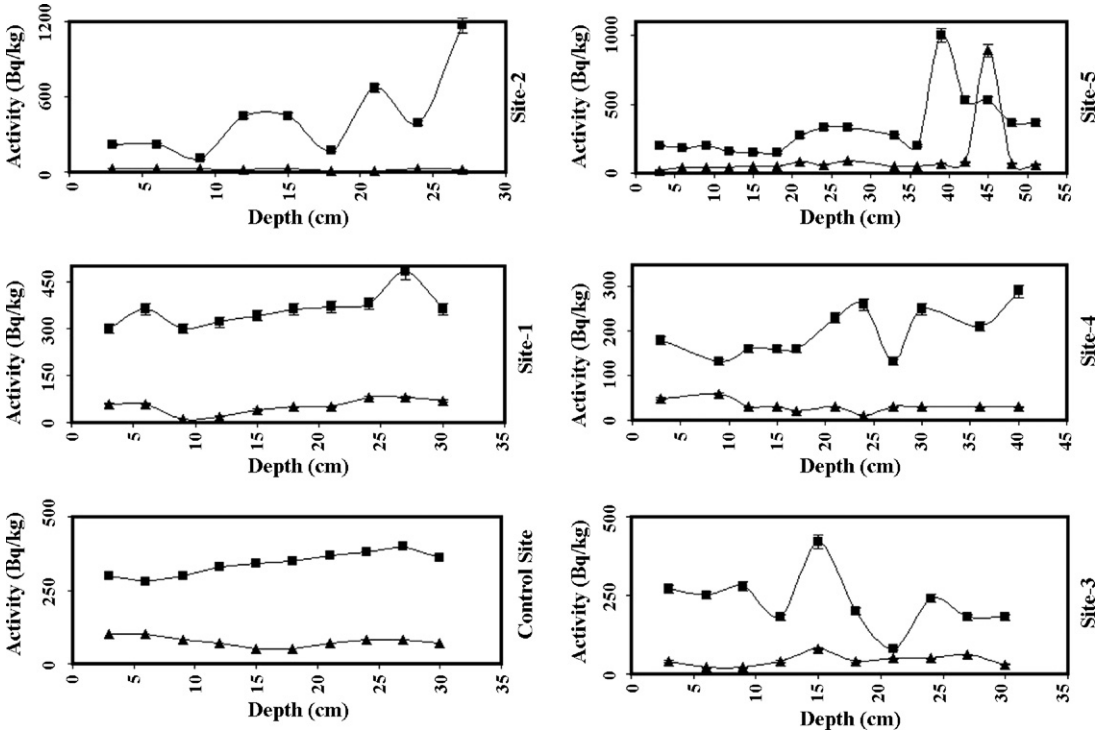

Fig. 2. The vertical profiles of ^{226}Ra and ^{210}Po difference with depth at all sites (■)^{226}Ra, ▲^{210}Po).

tle with depth in soils from site-5, the farthest station from the coastal. The activity concentrations of ^{210}Po are slightly high in the surface of all the soil cores. An exceptional profile occurs at site-2 and site-5. The observed anomalous may be related to known uranium anomalies around this district [8,9]. ^{226}Ra and ^{210}Po concentrations remain fairly constant at site-5. Close to the bottom, ^{226}Ra and ^{210}Po profiles show a sharp increase and a maximum at about 40 and 46 cm, respectively. The profiles of ^{226}Ra and ^{210}Po have a tendency to diverge at site-2, with ^{226}Ra concentrations increasing toward the bottom. The variation of ^{226}Ra and ^{210}Po concentrations has a similarity at site-1 and site-5 and there is a general trend for these sites. At almost all depth, there is a tendency for ^{226}Ra profiles parallel to these of ^{210}Po at these sites. This similarity is to be expected if the principal source of ^{210}Po is the decay of ^{226}Ra.

The migration and distribution of the radionuclides in the environment are determined by their chemical properties, half-lives, the physicochemical properties of the environment, the peculiarities of the sources and environment pathways of the radionuclides. It is evident that the ^{226}Ra is more enriched in the deeper strata of the soils for sites 1, 2, 4 and 5 whereas ^{210}Po is more enriched for only site-5. This enrichment may reflect the soil type and may be due to the distribution of well-known uranium anomalies in the area. The lower activity in the surface soils may be attributed to the mixing of soil from the upper part of the soil cores and also to the washout of ^{226}Ra and ^{210}Po from the surface soil during the cultivation process.

The behaviour of heavy natural radionuclides including ^{232}Th, ^{238}U and its decay products (^{226}Ra, ^{210}Pb and ^{210}Po) in soil and their removal by irrigation waters have been mentioned by Martyushov and Bazylev. It was found that 96% of heavy natural radionuclides were removed by determining the correlation between the removal amounts of radionuclides and volumes of irrigation waters. The greatest removal is characteristic of ^{238}U (1.1%); the least, of ^{232}Th and ^{210}Po (0.01–0.03%). The removal amounts of ^{210}Pb are 0.02–0.06%. The removal of heavy natural radionuclides from soils by irrigation waters is associated with the irrigation rate [20].

Malanca et al. have reported the distribution of ^{226}Ra, ^{232}Th and ^{40}K in fifty-two undisturbed soil samples in Brazil and the average activity concentration of ^{226}Ra were found 29.2 Bq kg^{-1} [21]. Quindos et al. have studied natural radioactivity level in Spanish soils and the ^{226}Ra activity concentration was in the range from 28 to 94 Bq kg^{-1} by dry weight [22]. Also, comparison of point estimation techniques in the spatial analysis of ^{226}Ra and ^{228}Ra in soil has been made by Dowdall and O'Dea. Godoy et al. have reported that ^{226}Ra and ^{228}Ra concentrations in 0–5 cm soil samples collected from several south Shetland Islands were in the range from 2.74 to 20.5 Bq kg^{-1} for ^{226}Ra and from 3.5 to 24.3 Bq kg^{-1} for ^{228}Ra [23]. Karunakara et al. have investigated the vertical distribution and enrichment of ^{210}Po in the environment of Kaiga in South India. ^{210}Po activity in the soil varied from 17.1 to 228.2 Bq kg^{-1} with a mean value of 83.3 Bq kg^{-1} [24].

El-Daoushy and Garcia-Tenorio have reported that unsupported ^{210}Po is accumulated by different sedimentological and biogeochemical processes in various depository systems, such as rivers, lakes, marine coasts, spills, pestlands and ice-layers. They have studied the aquatic deposition of ^{210}Po and ^{226}Ra in the lake sediments and observed that ^{226}Ra activity increased with increasing depth while ^{210}Po decreased [29]. Karunakara et al. have also pointed that ^{210}Po concentration decreased with increasing soil depths in all sampling stations in the environment of Kaiga nuclear power plant site in South India. According to Karunakara et al. the main source of ^{210}Po in the surface soil is from the deposition of ^{222}Rn daughters. They also indicated that ^{210}Po deposited on the soil from the atmosphere is able to move down further the soil in the environs [24]. It was found that there is no correlation between ^{210}Po and the grain size of the soil. The low ^{210}Po activity in the upper strata of the soils in the present study may be due to the cultivation process.

The addition of fertilizer, particularly superphosphates, to the soil may increase its ^{226}Ra and ^{210}Po content. Sidle et al. have reported that common ammonium phosphate fertilizers may contain an average of 185 m Bq g^{-1} ^{226}Ra [25]. However, it was observed that radium was not excess in the upper parts of the cores. The fertilizer loadings may enter the deeper strata during the washout process and possibly leachate at the deeper parts of the soil by the ground water [26].

According to Zhanguo et al., the radionuclides which are soluble and exchangeable are transported easily during erosion process of surface soils, and ^{226}Ra and ^{228}Ra should be combined in the crystalline skeleton of clay minerals. 75%–83% of ^{226}Ra was remained in the residues, while only 10%–20% of ^{226}Ra was bounded to the organic and Fe-Mn oxy-hydroxide fractions, with very little bound to the exchangeable and carbonate fractions [2].

The figures for the radium profiles of various soil types up to depths of 100 and 200 cm are particularly interesting. Analyses of the sample show that the concentration of radium in soil depends mainly on the abundance of parent radioactivity in parent rock. It was found that the distribution depends on the type of soil profile [20].

The soil types in the Buyuk Menderes Basin are following: Alluvial, limeless brown soil, colluvial, reddish Mediterranean soil, regosol, brown forest soil, salty, alkaline and alkaline + salty in Eastern part of the Basin and Alluvial, colluvial, limeless brown soil, brown forest soil, rendzina, red-brown Mediterranean soil, regosol in Western part of the Basin [12,13].

The interaction between radionuclide and the soil matrix is an important parameter in estimating radionuclide transfer processes. Organic matter (%), clay (%), silt (%), sand (%) contents and pH values of the surface soils for the sampling sites in which the core samples were taken were determined in our previous study [10] and given in Table 1. ^{210}Po is very immobile in soil and therefore it is likely irreversibly sorbed on clay and organic colloids. The amount of ^{210}Po contained in the top layer of soil can be correlated with the amount of precipitation [27]. There is a close correlation between the ^{210}Po levels of soil and its organic substance [24]. ^{226}Ra and ^{210}Po activity values change depend on soil type and texture. Table 2 shows ^{226}Ra and ^{210}Po activities in some countries.

Karunakara et al. have pointed that ^{210}Po concentration decreased with increasing soil depths in all sampling stations

Table 1
Soil site location and soil texture

Site	Latitude (N)	Longitude (E)	Organic matter (%)	Clay (%)	Silt (%)	Sand (%)	Soil (pH)
1	37°31′27	27°16′27	8.78	7.84	1.28	90.88	7.00
2	37°39′06	27°22′51	5.83	–	6.52	93.92	7.27
3	37°38′76	27°22′94	4.58	2.08	6.82	91.10	7.25
4	37°53′42	27°29′12	2.25	5.68	1.78	92.54	6.05
5	37°48′64	27°42′84	3.56	–	12.28	89.76	7.90

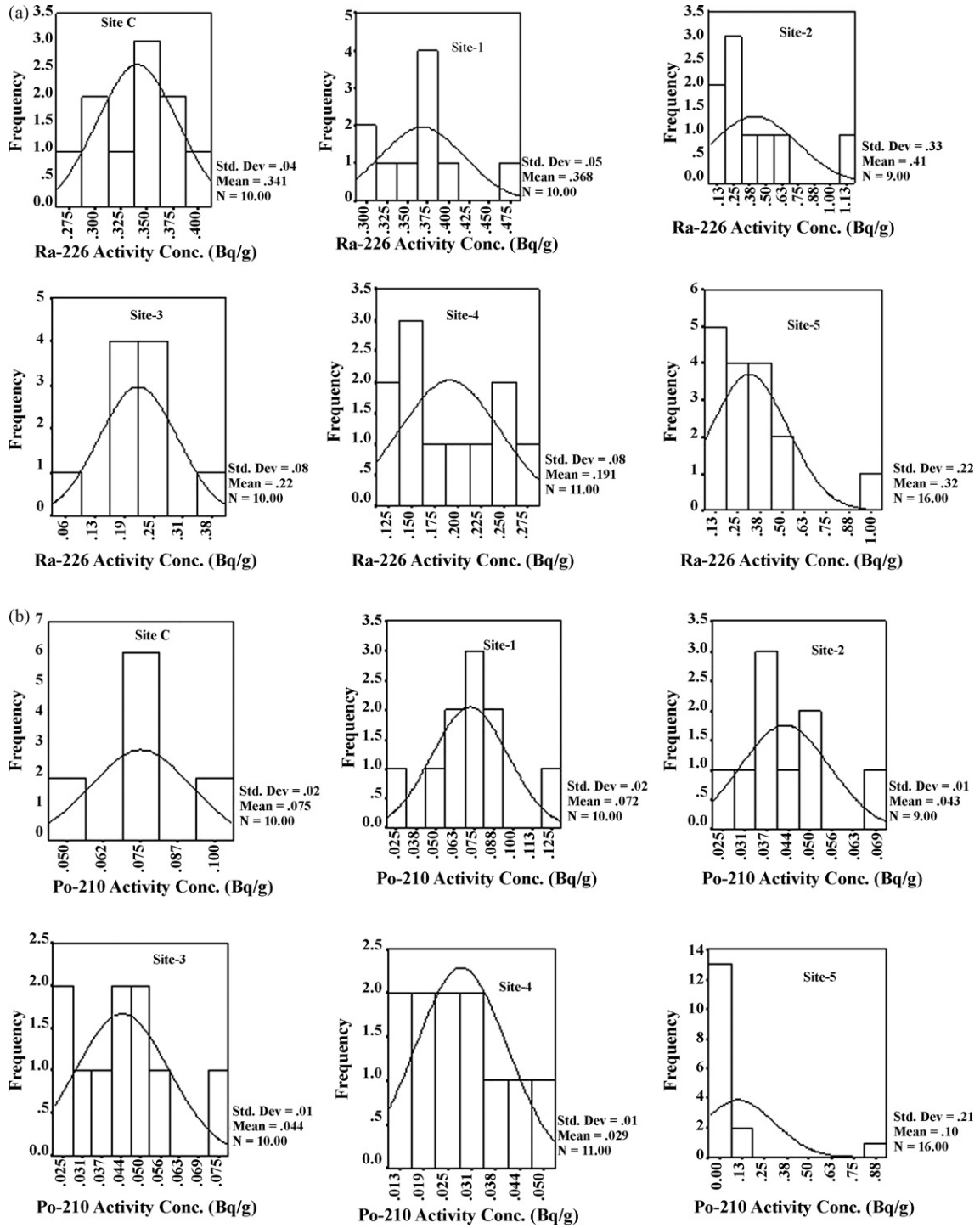

Fig. 3. Frequency distributions of ^{226}Ra and ^{210}Po activity concentrations.

Table 2
^{226}Ra and ^{210}Po activities in soils in some countries

Country	^{226}Ra (Bq kg^{-1})	^{210}Po (Bq kg^{-1})	References
Russia	10–30	30–50	[23]
South India		17.1–228	[18]
Brazil	2.74–20.5		[28]
Spain	28–94		[16]
Brazil	29		[28]
Switzerland	10–900		[28]
Greece	1–240		[28]
Estonia	6–310		[28]
India		17–228	[23]
USA		8–128	[29]
Germany		33–207	[30]
Brazil		32–70	[31]
Our previous study	37–298	13–135	[10]
Qur present study	80–1170	10–870	
World range	3.7–125.5	8.14–219	[18,24]

Table 3
Statistical data of ^{226}Ra and ^{210}Po activity concentrations (Bq g^{-1}) for five sampling sites of Buyuk Menderes Basin

Sample point	Skewness*		Kurtosis		Median	
	^{210}Po	^{226}Ra	^{210}Po	^{226}Ra	^{210}Po	^{226}Ra
Site 1	0.259	0.659	2.105	1.032	0.070	0.372
Site 2	0.869	1.829	0.792	3.355	0.038	0.245
Site 3	0.867	0.790	1.093	1.874	0.043	0.208
Site 4	0.586	0.504	−0.297	−1.075	0.026	0.170
Site 5	3.928	2.163	15.588	5.632	0.045	0.253

* Critical values for rejection ($n = 10$, $\alpha = 0.05$) are <-0.79, >0.79.

in the environment of Kaiga. They also indicated that ^{210}Po deposited on the soil from the atmosphere is able to more down of the soil in the environs. It was found that there is no correlation between ^{210}Po concentration and the grain size of the soil [24].

A software program is used to compute the basic statistics. For the collected soils, the frequency distributions of ^{226}Ra and ^{210}Po activity concentrations are represented in Fig. 3 and their fits plotted considering the skewness and the kurtosis coefficients that show the distribution of variables whether it obeys the normal or lognormal distribution. The skewness, kurtosis coefficients and median are given in the Table 3.

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 of ^{226}Ra and ^{210}Po suggest that the distributions are close to normal except for site-5. Negative kurtosis values at site-4 indicate that more values are located in the tails than in a standard normal distribution.

The skewness test indicated that the ^{226}Ra and ^{210}Po concentrations for depth increment were found to be normally distributed at site-1 and site-4 and also ^{226}Ra concentration at site-3. On the other hand, the means of site-2 and site-5 for ^{226}Ra and site-5 for ^{210}Po differs from the median this indicates that distribution at these sites is skewed. Comparison of median concentrations for ^{210}Po and ^{226}Ra indicate that ^{226}Ra is the dominant α-emitting source in soils, particularly at site-1. ^{226}Ra concentrations are greater than ^{210}Po concentrations for all sites.

4. Conclusions

The ^{226}Ra activities in soil core samples from the Buyuk Menderes Basin were determined to be higher than the world average. The depth profiles for ^{226}Ra show an increasing trend. The ^{210}Po activity remains almost constant with increasing depth. ^{226}Ra and ^{210}Po activity concentrations in five core samples ranged from 80 to 1170 Bq kg^{-1} and from 10 to 870 Bq kg^{-1}, respectively. The ^{226}Ra and ^{210}Po activity concentrations in the soil core samples can be attributed to the soil type and to the distribution of uranium anomalies in the Buyuk Menderes Basin.

Obtained data can help to explain the migration of ^{226}Ra and ^{210}Po in the cultivated soil. In conclusion, investigation of the distribution of radium and polonium in agricultural soil profiles for their pathway and behaviour is very important to understanding the possible impact on the ecological system.

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