

BIOMONITORING OF ^{137}Cs , ^{40}K , ^{232}Th , AND ^{238}U USING OAK BARK IN BELGRADE FOREST, ISTANBUL, TURKEY

by

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In the present study, the bark of oak trees (*Quercus petraea* Liebl.) was collected from Belgrade Forest northwest of Istanbul for determination of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U activity. A gamma spectrometer equipped with high-purity germanium detector was used for radioactivity measurement. Bark samples were collected from the northern and southern sides of trees at a height of 1.5 m above the ground in order to see the effects of rainfall and wind; and they were also collected from the northern side at a height of 0.5 m above the ground to assess the effect of soil splash. The activity concentrations of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U in the bark samples were found to vary in the range of 1.40-27.50, 45.0-221.2, 0.92-9.64, and 4.04-36.10 Bq/kg, respectively. The elevated activity of ^{238}U in bark samples could be attributed to a large amount of coal combustion in the region until the 1990s. According to one-way ANOVA, a significant difference was not found in bark samples collected at a height of 0.5 m and 1.5 m above the ground in terms of radionuclide activity. There was also no significant variation regarding radionuclide accumulation between northern and southern sides.

Key words: ^{137}Cs , natural radionuclides, oak bark, bio-monitoring, Turkey

INTRODUCTION

There are two main sources of radioactivity in the terrestrial environment. One of them is from natural radionuclides such as ^{40}K , ^{232}Th , and ^{238}U series. Among these radionuclides, ^{232}Th and ^{238}U are not generally used; however, ^{40}K is commonly metabolized by organisms with stable potassium without distinction. The other source of the radioactivity is from man-made radionuclides released into the atmosphere as a consequence of nuclear weapon testing, particularly in the 1960s, or nuclear power plant accidents. Many artificial radionuclides (^{134}Cs , ^{137}Cs , ^{131}I , ^{90}Sr , etc.) were released as a result of these events. Among artificial radionuclides, ^{137}Cs is the most important because of its relatively long half-life (30.2 years), high mobility in biological systems, and its chemical properties, which are similar to potassium. ^{137}Cs emits beta particles and gamma rays, and it accumulates in the soft tissues of organisms [1].

Radionuclides are transferred to plants from the soil through their roots and also are adsorbed into the external part of plants through wet and dry atmospheric deposition over long periods of time [2]. Therefore, various parts of plants such as bark, leaves,

crops, and twigs have been used for determination of radionuclides. Recently, bark samples have widely been used for bio-monitoring of radionuclides [2-6]. Zhiyanski *et al.* [6] after comparing bark, young branches, leaves, old branches, and wood, showed that bark is the best accumulator of ^{137}Cs . Similarly, Desideri *et al.* [7] observed the highest activity level in bark samples in comparison to leaves, flowers, fruit, berries, seeds, and roots. Anjos *et al.* reported that cesium concentration ratio in specific compartments depend on plant species (orange, lemon, chili pepper, and guava trees) [8]. In a previous study, it was declared that stem wood and bark become important ^{137}Cs sinks with tree development, because these compartments represent an increasing pool of biomass [9]. Barci Funel *et al.* notified that activity levels of ^{137}Cs were found to be higher for one or two orders of magnitude in bark than in inner parts of pine, spruce and larch [10]. Similarly, McGee *et al.* indicated that ^{137}Cs concentrations were five times higher in the bark than in needle and twig on pine and spruce trees [11].

Belivermis *et al.* [5] reported that the bark of the oak tree could be a good indicator of artificial radionuclides such as ^{137}Cs because of its relatively thick and rough structure. The thick and rough structure of oak bark causes an increase in dead tissues, particularly in the outer layer. Therefore, atmospheric

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pollutants such as metals and radionuclides are immobile and can not be depurated by metabolic processes in these tissues [5].

The aim of the current study was to determine the activity concentrations of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U in oak bark as a bio-monitoring material. Another objective was to observe whether there were differences between the northern and southern sides of trees regarding the radionuclide accumulation, and to investigate the effect of soil splash on deposited radionuclides in bark samples at a height of 0.5 m above the ground.

MATERIAL AND METHODS

Study area

Belgrade Forest located northwest of Istanbul (fig. 1) has a total area of approximately 5400 hectares. The most common tree in the forest is sessile oak (*Quercus petraea*) [12]. The annual average precipitation in the region is approximately 1161 mm [13]. The texture of the soil is shallow to deep, gravelly and sandy-clay loam [14].

Sampling and preparation of samples

In the study area, northern winds are dominant [12]. Therefore the northern side of the hills has richer flora than the southern slopes. Similarly, more moss and lichen vegetate on the northern face of rocks and trees in comparison to their southern side. Bark samples from 15 different oak trees (*Quercus petraea*) were taken at 0.5 m and 1.5 m above the ground on the northern side and 1.5 m above the ground on the southern side in January 2010. Radionuclides in soil can reach height of 0.5 m on trunk above the ground; however, the radionuclides can not reach up to 1.5 m of trunk above the ground as a result of soil splash by

heavy rain. Therefore, these heights (0.5 m and 1.5 m) were chosen to determine the effect of soil splash on deposited radionuclides in bark samples. In total, 45 bark samples were obtained in the study area. The samples were scraped from the outer 0.5 cm of the bark. Approximately 500 g from each sample was dried at 85 °C to a constant weight and homogenized. After determining dry weight, the samples were ignited in a furnace at 400 °C. Then the samples were placed into 170 ml plastic beakers to measure the radioactivity by using gamma spectrometry. The volume, mass, and density of the samples (60-80 ml, 36-64 g, and 0.6-0.8 g/cm³) were adjusted in plastic beakers according to the calibration source.

Analysis using gamma spectrometry

All samples were sealed for four weeks prior to spectrometry measurements in order to establish secular equilibrium between ^{226}Ra , ^{222}Rn and their decay products for radionuclide analysis. Radiation levels were measured using a gamma spectrometer which includes gamma multichannel analyzer equipped with high purity germanium (HPGe) detector (Canberra 2020). The gamma spectra were analyzed using the ORTEC Maestro 32 data acquisition and analysis system. The detector had coaxial closed-facing geometry with the following specifications: resolution (FWHM) at 122 keV ^{57}Co was 1.0 keV and at 1.33 MeV ^{60}Co it was 2.0 keV. Relative efficiency at 1.33 MeV ^{60}Co was 22.1%. The detector was shielded by a cylindrical lead shield with average thickness of 10 cm in order to achieve the lowest background level. Energy-dependent efficiency calibration was carried out by using a standard soil nuclide mixture (Isotope Products Laboratories) containing known activities of ^{210}Pb , ^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{203}Hg , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y , and ^{60}Co . It is known that for accurate calculation of activity values, consistency of samples in terms of volume and density is of overriding importance. In this study, the

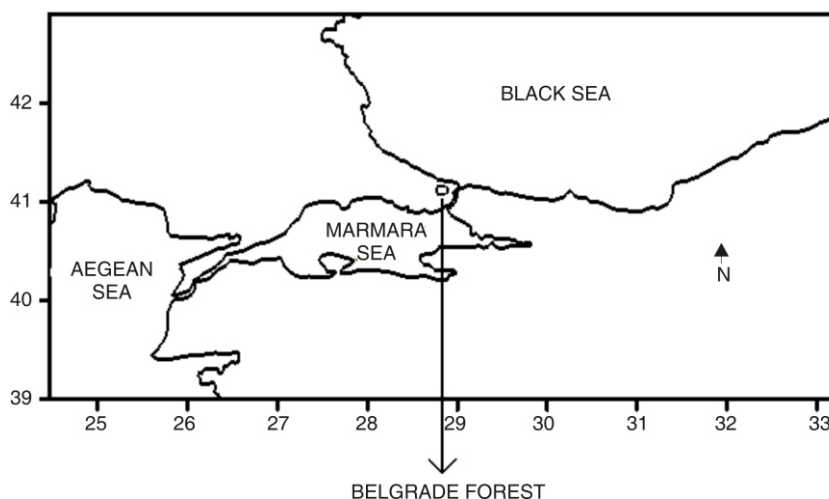


Figure 1. Study area

volume and density of the samples were a standard 70 ml and 0.7 g/cm^3 , respectively. The activity concentrations of ^{137}Cs and ^{40}K were determined directly from the peak areas at 661.6 and 1460.8 keV, respectively. The activity concentrations of ^{238}U and ^{232}Th were calculated assuming secular equilibrium with their decay products. The gamma transition lines of ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 keV) were used to calculate activity concentrations of radioisotopes in the U-series. The activity concentrations of radioisotopes in the Th-series were determined using gamma transition lines of ^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV), and ^{228}Ac (911.1 keV). The counting time for each sample was at least 72 hours. It is known that ^{137}Cs has a physical half life around 30 years. In this study, several months had passed from collecting the samples until the measurement date. Therefore, loss of activity levels of ^{137}Cs occurs due to its half life in the passed time. Thus, decay-correction was made to determine activity levels of ^{137}Cs at the sampling date. Activity values were given as Bq/kg of dry weight. The uncertainty values presented as are the total uncertainties, and were calculated taking into account counting and efficiency calibration errors.

Statistical analysis

Pearson's two-tailed correlation coefficient was carried out in order to clarify the relationship between the radionuclide concentrations found in the samples. One-way analysis of variance (ANOVA) was used to evaluate the differences between radionuclide quantities accumulated on the northern and southern sides of trees and the soil splash effect on deposition of radionuclides in bark samples.

RESULTS AND DISCUSSION

The activity concentrations of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U in bark samples, which are 0.5 m and 1.5 m above the ground on the northern side, and 1.5 m above ground on the southern side, are presented in tab. 1. Table 2 shows Pearson's correlation between the radionuclide activity concentrations. The averages and ranges of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U activity concentrations were found to be 11.87, 1.40-27.50; 81.6, 45.0-221.2; 2.68, 0.92-9.64, and 9.25, 4.04-36.10 Bq/kg, respectively.

Activity concentrations of ^{137}Cs

In previous studies, mean activity levels of ^{137}Cs were found to be 10.2 and 11.87 Bq/kg in oak bark samples collected from South Bulgaria and the Thrace region of Turkey, respectively [5, 6]. In a study carried out in Croatia, the range of ^{137}Cs activity was found to be 15.1-45.1 Bq/kg in the bark of the silver fir tree [4].

In the present study, the average and range of ^{137}Cs activity were found to be 11.87 and 1.40-27.50 Bq/kg. Although the study areas are different, ^{137}Cs activity found in the present study is consistent with other studies.

According to one-way ANOVA ($p = 0.01$), there was no significant difference between samples on the northern and southern sides of trees in terms of ^{137}Cs activity. There was also no significant difference between bark samples collected at 0.5 m and at 1.5 m above the ground. This indicates that there is no soil splash effect on the accumulation level of ^{137}Cs at 0.5 m above the ground, probably due to thick plant cover on the soil surface. However, Zhiyanski *et al.* [6] found higher ^{137}Cs activity in bark samples collected at a height of 1.3 m from the base of the trunk in comparison to those collected at ground level. According to Pearson's correlation, a weak positive correlation coefficient is observed between ^{137}Cs and ^{238}U (tab. 2). This confirms that these radionuclides were mainly taken up by adsorption from atmospheric deposition. Although ^{238}U is not used in metabolism of plants, activity levels were found to be much higher in the present study probably due to widespread coal combustion in Istanbul. Also, ^{137}Cs was found in high activity levels comparing other plants (lichen and moss) [15]. This is presumably related to relatively thick and rough structure of oak bark, and thus oak bark can be easily take up ^{137}Cs radionuclide.

Activity concentrations of ^{40}K

In previous studies, mean activity levels of ^{40}K have been determined to be 123.3 and 72.3 in oak bark samples from Croatia and the Thrace region of Turkey, respectively [3, 5]. Lovrencic *et al.* [4] found ^{40}K activity in the range of 61.2-135.2 in the bark of silver fir. The average and range of ^{40}K activity were found to be 81.6 and 45.0-221.2 Bq/kg in the current study. These activity levels are similar to the values found in previous studies in the literature. As a result of Pearson's correlation analysis, a weak positive correlation coefficient was found between ^{40}K and ^{232}Th . This result is probably related with the fact that K is an essential element for the metabolism of trees while Th is not.

Activity concentrations of ^{232}Th and ^{238}U

Belivermis *et al.* [5] found that activity concentrations of ^{238}U were higher than those of ^{232}Th in oak bark. Similarly, activity levels of ^{238}U were found to be higher than those of ^{232}Th in the current study (tab. 1, figs. 2 and 3). In general, if there is no thorium and uranium source such as a uranium mine in the vicinity of the investigated area, it is expected that biota samples accumulate radionuclides from Th and U series at sim-

Table 1. Activity concentrations of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U in bark samples at 0.5 m and at 1.5 m above ground on north side and at 1.5 m above ground on south side of oak trees

Bark samples	Collected side and height	^{137}Cs	^{40}K	^{232}Th	^{238}U
1	north 0.5 m	6.58 0.47*	159.1 8.26	9.64 0.63	7.47 0.58
	north 1.5 m	8.68 0.68	188.2 9.97	4.28 0.41	4.04 0.40
	south 1.5 m	16.79 1.00	221.2 11.6	3.48 0.33	5.32 0.56
2	north 0.5 m	17.53 0.96	97.8 5.26	5.00 0.42	19.09 1.13
	north 1.5 m	17.64 0.96	91.2 5.01	5.62 0.45	24.79 1.39
	south 1.5 m	11.84 0.65	64.3 3.43	2.25 0.21	33.94 1.79
3	north 0.5 m	14.73 0.81	66.5 3.53	2.05 0.20	18.88 1.06
	north 1.5 m	12.83 0.69	60.4 3.22	1.82 0.17	16.21 0.89
	south 1.5 m	10.12 0.58	52.3 2.83	1.72 0.17	16.12 0.92
4	north 0.5 m	23.49 1.25	82.4 4.42	2.08 0.21	13.49 0.85
	north 1.5 m	27.50 1.43	87.0 4.64	1.70 0.17	13.44 0.81
	south 1.5 m	14.41 0.77	77.9 4.13	1.21 0.14	9.36 0.57
5	north 0.5 m	15.63 0.84	48.5 2.64	1.19 0.11	22.72 1.23
	north 1.5 m	20.32 1.07	60.3 3.28	2.78 0.25	19.58 1.08
	south 1.5 m	21.34 1.09	61.7 3.22	1.28 0.10	22.21 1.17
6	north 0.5 m	20.05 1.08	63.4 3.47	4.31 0.36	23.43 1.31
	north 1.5 m	15.41 0.88	108.4 5.84	4.64 0.44	25.66 1.45
	south 1.5 m	9.61 0.55	87.7 4.61	0.92 0.11	36.10 1.90
7	north 0.5 m	15.72 0.86	57.7 3.14	3.77 0.35	19.97 1.12
	north 1.5 m	16.97 0.91	53.4 2.95	2.94 0.28	23.79 1.30
	south 1.5 m	10.66 0.57	46.1 2.48	1.11 0.13	18.02 0.96
8	north 0.5 m	6.31 0.44	98.0 5.26	2.12 0.22	5.61 0.48
	north 1.5 m	7.39 0.52	90.5 4.90	2.99 0.30	7.12 0.57
	south 1.5 m	3.04 0.28	124.1 6.56	2.29 0.25	5.51 0.50
9	north 0.5 m	5.17 0.45	131.5 7.00	2.89 0.31	5.03 0.50
	north 1.5 m	5.12 0.32	101.4 5.31	1.55 0.14	4.81 0.32
	south 1.5 m	1.40 0.26	115.6 6.12	1.67 0.19	4.88 0.42
10	north 0.5 m	8.93 0.55	61.3 3.28	1.51 0.16	4.16 0.36
	north 1.5 m	7.91 0.46	50.2 2.70	1.58 0.15	4.44 0.33
	south 1.5 m	7.39 0.42	88.1 4.59	0.99 0.11	4.05 0.32
11	north 0.5 m	2.40 0.24	67.8 3.63	2.98 0.27	6.44 0.45
	north 1.5 m	2.39 0.22	59.7 3.18	3.57 0.29	6.72 0.45
	south 1.5 m	2.40 0.21	45.0 2.39	0.96 0.11	10.04 0.59
12	north 0.5 m	9.54 0.59	99.9 5.37	3.99 0.40	14.85 0.99
	north 1.5 m	6.28 0.39	58.1 3.16	2.99 0.29	13.75 0.80
	south 1.5 m	6.85 0.43	59.7 3.21	1.60 0.17	13.92 0.86
13	north 0.5 m	19.92 1.07	77.5 4.18	2.77 0.27	33.42 1.80
	north 1.5 m	11.17 0.62	52.3 2.85	2.34 0.23	17.59 1.01
	south 1.5 m	13.38 0.73	68.5 3.69	3.63 0.34	17.48 0.98
14	north 0.5 m	12.06 0.69	73.4 3.92	2.87 0.27	25.55 1.40
	north 1.5 m	13.60 0.74	46.0 2.54	2.93 0.16	25.04 1.34
	south 1.5 m	13.83 0.74	63.5 3.38	1.75 0.19	33.40 1.74
15	north 0.5 m	13.99 0.77	69.9 3.73	2.61 0.26	20.52 1.13
	north 1.5 m	13.97 0.75	60.8 3.31	2.40 0.22	26.94 1.42
	south 1.5 m	11.98 0.67	74.2 3.96	1.70 0.19	17.98 0.99
Average to	north 0.5 m	12.80 6.18**	83.6 29.7	3.32 2.04	16.04 8.82
	north 1.5 m	12.48 6.57	77.9 36.6	2.94 1.18	15.59 8.55
	south 1.5 m	10.34 5.50	83.3 44.5	1.77 0.84	16.56 10.85

* Uncertainty of measurement

** Standard deviation of mean

Table 2. Pearson's two-tailed correlation coefficients between activity concentrations of radionuclides

Radionuclide and sample	^{40}K N50	^{238}U N50	^{232}Th N50	^{137}Cs N150	^{40}K N150	^{238}U N150	^{232}Th N150	^{137}Cs S150	^{40}K S150	^{238}U S150	^{232}Th S150
^{137}Cs N50	-0.42	0.72*	-0.17	0.86*	-0.17	0.68*	0.04	0.59**	-0.35	0.55**	-0.02
^{40}K N50		-0.46	0.71*	-0.36	0.79*	-0.52**	0.20	-0.15	0.81*	-0.40	0.60**
^{238}U N50			-0.12	0.48	-0.37	0.82*	0.14	0.58**	-0.45	0.74*	0.20
^{232}Th N50				-0.16	0.81*	-0.11	0.64**	0.18	0.68*	0.02	0.51
^{137}Cs N150					-0.06	0.58**	0.03	0.69*	-0.24	0.43	-0.17
^{40}K N150						-0.35	0.44	0.11	0.89*	-0.19	0.44
^{238}U N150							0.31	0.45	-0.53**	0.87*	-0.14
^{232}Th N150								0.14	0.17	0.56**	0.19
^{137}Cs S150									0.07	0.38	0.25
^{40}K S150										-0.42	0.55**
^{238}U S150											-0.12

* Correlation is significant at 0.01 level (2-tailed)

** Correlation is significant at 0.05 level (2-tailed)

Figure 2. Activity concentrations of ^{137}Cs and ^{40}K in oak bark samples

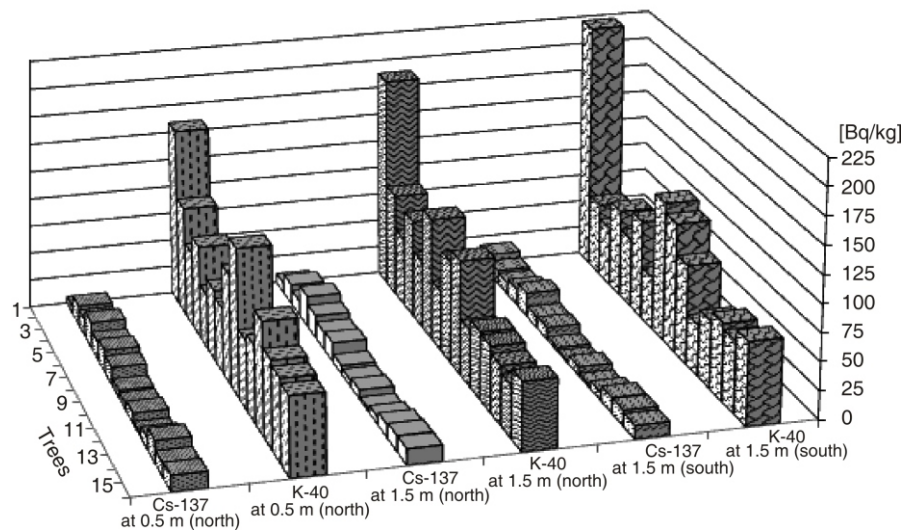
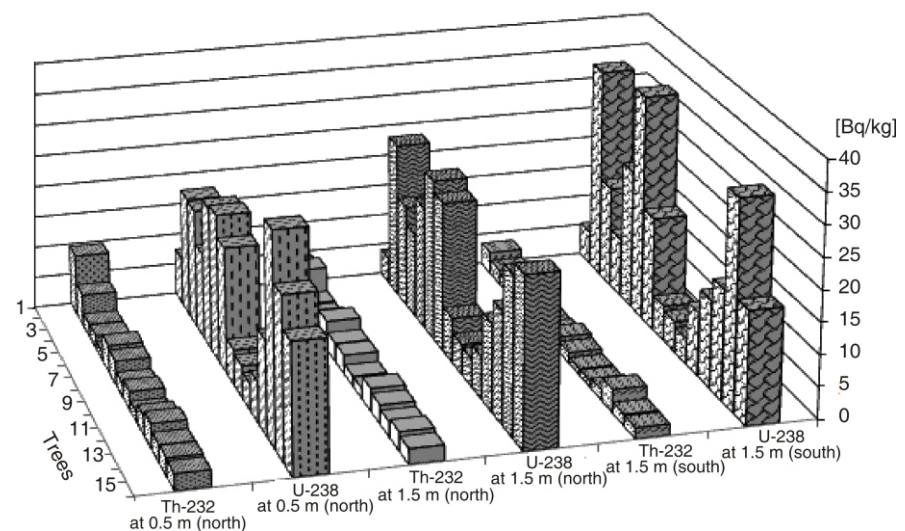


Figure 3. Activity concentrations of ^{232}Th and ^{238}U in oak bark samples



ilar levels [1, 15]. However, in the present study, average ^{232}U activity was found to be six times higher than average ^{232}Th activity in the bark samples. Papastefanou [16] reported that the ^{238}U activity level was higher than the ^{232}Th activity concentration in fos-

sil fuels such as coal. Flues *et al.* [17] documented high ^{238}U activity in the vicinity of a coal-fired power plant whereas ^{232}Th activity was consistent with the control areas. In the current study, higher ^{238}U activity in the bark samples might have been found due to long-term

atmospheric deposition as a result of burning fossil fuels (particularly coal), since coal was the main combustible fuel used by residents of Istanbul to heat houses until the 1990s. After the 1990s, natural gas became the main combustible fuel in the region. However, coal is still used in some areas of Istanbul and the combustion product of natural gas can also contain natural radionuclides. Considering there are approximately 15 million inhabitants in Istanbul, a large amount of U-series radionuclides might have been released into the atmosphere through coal combustion. Furthermore, rather high positive correlation coefficients between ^{137}Cs and ^{238}U activity in samples collected at 50 and 150 cm from the northern side of the oak trees (0.72, 0.58) confirm that the ^{137}Cs and U-series radionuclides have been deposited atmospherically (tab. 2).

Belivermis *et al.* found the mean activity concentrations of ^{232}Th and ^{238}U to be 4.03 and 8.68 Bq/kg in oak bark samples [5]. In the present study, the average activity levels of ^{232}Th and ^{238}U were found to be 2.68 and 16.06 Bq/kg, respectively. In the current study, it is seen that the mean activity concentration of ^{232}Th was lower while the mean activity concentration of ^{238}U was higher in comparison with the levels from Belivermis *et al.* (2010) [5]. This might be related to various degrees of dry and wet atmospheric deposition in both study areas. It is known that these radionuclides are not used in the metabolism of trees. Therefore, the activity values of these radionuclides should be at low levels in biota samples. However, especially concerning ^{238}U , activity found in the bark of oaks is noticeably high (16.06 Bq/kg) compared to other biota samples [15]. While ^{232}Th activity is consistent, ^{238}U activity is several times higher than that found in moss and lichen in a previous investigation carried out in Istanbul [15]. In addition, considering the proximity of a marine environment to the present study area, while ^{232}Th activity is consistent, ^{238}U activity is several times higher than that found in mussels at the coast of Istanbul [1].

CONCLUSIONS

Activity concentrations of ^{137}Cs , ^{40}K , ^{232}Th , and ^{238}U in oak bark samples in the current study are consistent with the results from similar studies in the literature. This study showed that oak bark can accumulate ^{137}Cs in significant amounts. There were no significant variations in terms of accumulation of radionuclides between the samples collected from the northern and southern sides of trees. In addition, no significant variations were found between samples collected at a height 0.5 m and at 1.5 m above the ground on the north side.

To sum up, oak bark can be an ideal bioindicator for the terrestrial environment especially for determin-

ing atmospheric radionuclide deposition. The rough and thick shape of the outer layer of oak bark increases the available surface for adsorption. Also, the lack of phloem and xylem leads to more dead tissues in the outer bark. Due to long life of oak trees and presence of dead tissues in its bark, deposited activity remains for a long time in the outer bark.

Some disadvantages of other bioindicators in determining long term deposition, which can not be applied to oak bark, are as follows:

- in soil samples, the physical and chemical properties of the soil, bio perturbation, cultivation, and vertical and horizontal migration prevent accurate determination of the levels of atmospheric radionuclide deposition, and
- moss and lichen are known to be good bioindicators because of the nourishment they receive directly from atmosphere, however, the effect of soil splash on their talus and depuration of radionuclides by metabolic processes may cause problems, and in the case of vascular plants, they cannot be good indicators due to soil uptake.

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БИОМОНИТОРИНГ ^{137}CS , ^{40}K , ^{232}TH И ^{238}U КОРИШЋЕЊЕМ ХРАСТОВЕ КОРЕ ИЗ БЕОГРАДСКЕ ШУМЕ КРАЈ ИСТАНБУЛА

У овом раду приказано је одређивање активности ^{137}Cs , ^{40}K , ^{232}Th , и ^{238}U сакупљене коре храстових стабала (*Quercus petraea* Liebl.) из Београдске шуме северозападно од Истанбула. За мерење радиоактивности коришћен је гама спектрометар са германијумским детектором високе чистоће. Узорци кора су сакупљени са северне и јужне стране стабала на висини од 1.5 метара од земље како би се видели утицаји кише и ветра, као и са северне стране стабала на висини од 0.5 метара од земље, ради процене утицаја наноса земљишта. Концентрације активности ^{137}Cs , ^{40}K , ^{232}Th , и ^{238}U у узорцима кора биле су у опсегу 1.4-27.5, 45.0-221.2, 0.92-9.64, и 4.04-36.10 Bqkg^{-1} , респективно. Повишена активност ^{238}U у узорцима може се приписати великом сагоревању угља у региону до 1990-тих година. На основу АНОВА теста, у погледу активности радионуклида није пронађена значајна разлика у узорцима прикупљеним на висинама 0,5 и 1,5 метара. Такође, нема значајне разлике у акумулацији радионуклида између северне и јужне стране.

Кључне речи: ^{137}Cs , природни радионуклиди, храстова кора, биомониторинг