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A study on the activity concentrations of natural radionuclides and annual effective dose values in some tobacco samples

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Abstract

Tobacco samples were collected from different tobacco markets from Antalya in Turkey. To calculate natural radionuclides activity concentration, samples were counted for 86400 seconds by using high purity germanium (HPGe) detector. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for tobacco samples found higher than minimum detectable activity (MDA). ^{137}Cs was not detected in any sample. Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K range from 83±15 Bqkg^{-1} to 325±45 Bqkg^{-1}, 29±5 Bqkg^{-1} to 207±37 Bqkg^{-1} and (2.09±0.28)x10^{3} Bqkg^{-1} to (5.07±0.90) x10^{3} Bqkg^{-1}, respectively. In addition, annual effective dose value was calculated. The annual effective dose values owing to inhalation for adults change from 2.76 to 9.91 µSvy^{-1} for ^{214}Pb, from 5.69 to 27.69 µSvy^{-1} for ^{214}Bi, from 5.72 to 40.41 µSvy^{-1} for ^{228}Ac, from 42.23 to 102.37 µSvy^{-1} for ^{40}K. The total annual effective dose values change from 0.05 to 0.16 mSvy^{-1}.

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1. Introduction

Determination of the activity concentrations of the natural radionuclides (^{238}U, ^{232}Th and ^{40}K) and artificial radionuclides (^{137}Cs, ^{134}Cs, ^{90}Sr and ^{131}I) are significant to protection of the radiation. ^{238}U, ^{232}Th and ^{40}K are major natural radionuclides which are toxic for health as well as environment. Cosmic rays and naturally occurring radioactive materials being in the earth itself are natural sources of the radiation whereas nuclear accidents, nuclear power plants, medical application of the radionuclides are man-made source of radiation [1].

Uranium has both radionuclide and metallic properties as well as its biological properties outcome from the chemical effects and natural occuring radionuclide. The internal contact with Uranium reveals radiological and chemical consequences which depend partly on chemical form of the intake and the route of intake (mainly ingestion and inhalation). The chemical toxicity effects of uranium species are namely as the renal effects, however others are associated with radiological toxicity of uranium. If we consider of the chemical effects of the uranium species, the kidneys are the easiest and sensitive target organ. Also, it effects central nervous system at higher radiation exposure. Such as, it has been repoted that it occured abnormal behaviours in animals at higher uranium doses [2].

Exposure to long term radiation causes bone weaking, necrosis of the nasal and mouth tumors, leucopoenia and large number of diseases. Such as, ingested or inhaled radium enhance the risk of developing following diseases as bone cancer, lymphoma, and diseases concerning the blood formation, such as aplastic anemia and leukemia etc. The emergences of these diseases are time taking. The hazard effect of the radiation depends upon the exposure time and intensity of radiation [1-3].

With the population growth, smoking becomes fashion of being modern specially in youngsters these days. But there is strong correlation between tobacco, cigarette, its smoke and lungs cancer [4]. Risk of occuring lungs cancer among non-smokers is ten times less than that of smokers [5]. Because tobacco which is main product of cigarette contains radioactive isotopes of ^{238}U and ^{232}Th and their decay series which are carcinogenic in nature [6]. The use of tobacco and its products for smoking increase the coincidental internal intake and radiation dose due to presence of these radioisotopes [7]. It is stated that there is strong

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correlation between lung cancer and smoking as well as there is positive correlation between the carciogenic effect of smoking and radiation [8].

Discussing some serious impacts about radionuclides present in the tobacco samples, lung cancer is very critical and serious issue. Though in recent times cancer gains more attention regarding health agenda all over the world [6]. Therefore, determination of the radioactivity concentration and annual effective dose values of the tobacco samples is significant to protection of the radiation. The aim of this study is to calculate activity concentration of natural radionuclides and dose evaluation in some tobacco samples.

2. Materials and Methods

In Sixteen different types of tobacco samples were collected randomly from different tobacco markets. The samples were numbered randomly from 1 to 16 and kept in plastic containers of 100 ml capacity. Plastic cups were closed firmly to prevent radon emission. Samples waited for 30 days before counting to come into secular equilibrium between radium and radon (uranium and its daughter nuclei) [6].

Samples were counted for 86400 seconds to calculate activity concentration of natural radionuclides using high purity germanium (HPGe) detector (AMATEK-ORTEC GEM40P4 – 83) which has 40% relavite efficiency, p type, electrically cooled. The detector is located in department of Physics in Akdeniz University. The energy resolution of the dedector is 1.85 keV at 1332 keV for ^{60}Co and 768 eV at 122 keV for ^{57}Co.

Before counting the samples, point sources [^{22}Na (1274 keV), ^{57}Co (122 and 136 keV), ^{54}Mn (834 keV), ^{60}Co (1173 and 1332 keV), ^{133}Ba (80, 276, 302, 356 and 384 keV) and ^{137}Cs (661 keV)] were counted for 1000 seconds to check energy calibration. Multinuclides standard source (IAEA 1364- 43 – 2) was used to determine efficiency calibration from Cekmece Nuclear Research and Training Center in Turkish Atomic Energy Authority (TAEK) [9, 10].

Spectra of tobacco samples were obtained by using MAESTRO-32 computer program [11]. Before analyzing of spectra, background spectrum was subtracted from each spectra of tobacco to reduce background effect. Spectra were analyzed automatically by using Gamma-W computer software [12].

Minimum detectable activity (*MDA*) was calculated by Currie equation as shown by following relation [13,14];

$$MDA = \frac{2.71 + 4.66\sigma}{t.\varepsilon.I_\gamma.m} \quad (1)$$

where σ, t, ε, I_γ and m are standard deviation of count, counting time, efficiency, probability of gamma-ray emission and mass of sample, respectively. Minimum detectable activities of ^{214}Pb (351.93 keV), ^{214}Bi (609.32 keV), ^{228}Ac (911.20 keV), ^{40}K (1460.82 keV) and ^{137}Cs (661.66 keV)] radionuclides for 1-kg sample size and 53999 seconds live counting time were calculated using background spectrum.

Activity concentration of radionuclides A ($Bqkg^{-1}$) were calculated using well-known following formula [15],

$$A = \frac{N}{m.t.\varepsilon.I_\gamma} \quad (2)$$

where N, m, t, ε and I_γ shows number of counts of under the peak with background subtraction, mass of the sample, counting time, detector efficiency of gamma-ray energy and probability of gamma-ray emission, respectively.

Uncertainty of activity concentration of radionuclides was calculated by using following relation [16]

$$\Delta A = A\sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta \varepsilon}{\varepsilon}\right)^2 + \left(\frac{\Delta I_\gamma}{I_\gamma}\right)^2 + \left(\frac{\Delta m}{m}\right)^2} \quad (3)$$

where ΔN, Δt, $\Delta \varepsilon$, ΔI and Δm show uncertainties of count of under peak with background, counting time, detector efficiency of gamma-ray energy, probability of gamma-ray emission and mass of sample.

Radium equivalent activity (Ra_{eq}) is calculated using following equation [17]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

here A_{Ra}, A_{Th} and A_K present activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively.

Annual effective dose value $(H_E)$is calculated as following [18]:

$$H_E = 0.75 \times M_T \times A_i \times F \quad (5)$$

where M_T (kgy^{-1}) is annual consumed quantity of tobacco mass which is estimated as 8.979 kg y^{-1} [4], M_T

(kgy^{-1}) is calculated estimating a smoker consumes 30 cigarette per day or 24.6 g of tobacco per day in Ref. [4]. A_i stands the concentration of the ith radionuclide, F (SvBq^{-1}) is dose conversation factor which is 4.8 × 10^{-9} SvBq^{-1} for ^{214}Pb, 1.2 ×10^{-8} SvBq^{-1} for ^{214}Bi, 2.9 × 10^{-8} SvBq^{-1} for ^{228}Ac and 3.0 × 10^{-9} SvBq^{-1} for ^{40}K [19].

Excess lifetime cancer risk ($ELCR$) is calculated using following equation [4,20]:

$$ELCR = (\sum H_E) \times DL \times RF \quad (6)$$

where DL is duration of life (~70 years) and RF is the risk factor (0.05 Sv^{-1}) [18,21],

Total annual effective dose $\sum H_E$ value is calculated as following:

$$\sum H_E = \frac{\left(H_{E(^{214}Pb)} + H_{E(^{214}Bi)}\right)}{2} + H_{E(^{228}Ac)} + H_{E(^{40}K)} \quad (7)$$

3. Results and Discussion

MDA of ^{214}Pb (351.93 keV), ^{214}Bi (609.32 keV), ^{228}Ac (911.20 keV), ^{137}Cs (661.66 keV) and ^{40}K (1460.82 keV) are 4.64 Bqkg^{-1}, 4.37 Bqkg^{-1}, 8.44 Bqkg^{-1}, 1.80 Bqkg^{-1} and 32.71 Bqkg^{-1}, respectively. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for tobacco samples found higher in present studies in compare with MDA. ^{137}Cs was not detected in any sample.

Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K are shown in Table 1. Activity concentration of ^{226}Ra calculated using mean value of ^{214}Pb and ^{214}Bi. Activity concentration of ^{226}Ra ranges from 83±15 Bqkg^{-1} to 325±45 Bqkg^{-1}, the mean value of ^{226}Ra is 157±20 Bqkg^{-1}. The activity concentration of ^{226}Ra is the highest in sample-1 while the activity concentration of ^{226}Ra is lowest in sample-8.

Table 1. Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in tobacco samples

Sample No	^{226}Ra			^{232}Th	^{40}K (x10^3)** (Bqkg^{-1})
	^{214}Pb (Bqkg^{-1})	^{214}Bi (Bqkg^{-1})	Mean Value (Bqkg^{-1}) (^{214}Pb+^{214}Bi)/2	^{228}Ac (Bqkg^{-1})	
1	307±46	343±46	325±45	183±33	4.22±0.53
2	177±21	202±20	190±21	131±18	4.29±0.39
3	149±19	183±19	166±19	98±13	3.17±0.25
4	122±14	133±21	128±18	38±12	2.44±0.35
5	162±24	188±14	175±19	104±14	4.17±0.34
6	147±15	173±14	160±15	118±15	3.13±0.26
7	85±14	109±13	97±14	111±18	2.25±0.19
8	95±16	70±14	83±15	29±5	2.09±0.28
9	153±18	169±13	161±16	95±14	3.61±0.36
10	147±20	135±17	141±19	109±16	3.03±0.25
11	233±36	229±34	231±35	207±37	5.06±0.90
12	101±15	116±12	108±14	91±12	3.10±0.29
13	110±15	126±12	118±14	99±13	3.03±0.25
14	128±14	164±17	146±16	127±18	4.41±0.36
15	185±23	208±20	197±21	125±19	4.52±0.48
16	133±20	186±16	159±18	122±21	4.08±0.36
Mean Value	152±21	163±19	157±20	112±17	3.54±0.37

**Data can be found in Ref. [22]

Activity concentration of ^{232}Th varies from 29±5 Bqkg^{-1} to 207±37 Bqkg^{-1}. The mean value of activity concentration of ^{232}Th is 112±17 Bqkg^{-1}. The activity concentration of ^{232}Th is highest in sample-11 while the activity concentration of ^{232}Th is lowest in sample-8. The activity concentration of ^{40}K varies from (2.09±0.28)$x10^3$ Bqkg^{-1} to (5.06±0.90)$x10^3$ Bqkg^{-1}. The mean value of activity concentration of ^{40}K is

(3.54±0.37) $x10^3$ $Bqkg^{-1}$. Also, the activity concentrations of ^{40}K can be found previous work in Ref. [22]. Among all the samples the highest value of activity concentration for ^{40}K is found in sample-11 while the lowest value in sample-8.

Activity concentration of ^{40}K is much higher than activity concentration of ^{226}Ra and ^{232}Th. This result is in aggreement with Ref. [23]. Activity concentration of ^{40}K and ^{226}Ra are higher than ^{232}Th because tobacco cultivated soil may be including high radioactivity or fertilizer and high activity concentration radionuclides can be transferred from roots to tobacco plant. General components of fertilizer are potassium, nitrogen, and phosphorus which are commonly used in the agriculture of tobacco plant [4]. The use of fertilizer in soil or cultivated area may bring enhancement in the radionuclide components. The radionuclide (uranium, thorium, radium, and polonium) may be transferred from soil to root and different parts of plants such as leafs, grain and stem [24]. Distribution of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in tobacco samples are shown in Fig.1, Fig.2 and Fig.3, respectively.

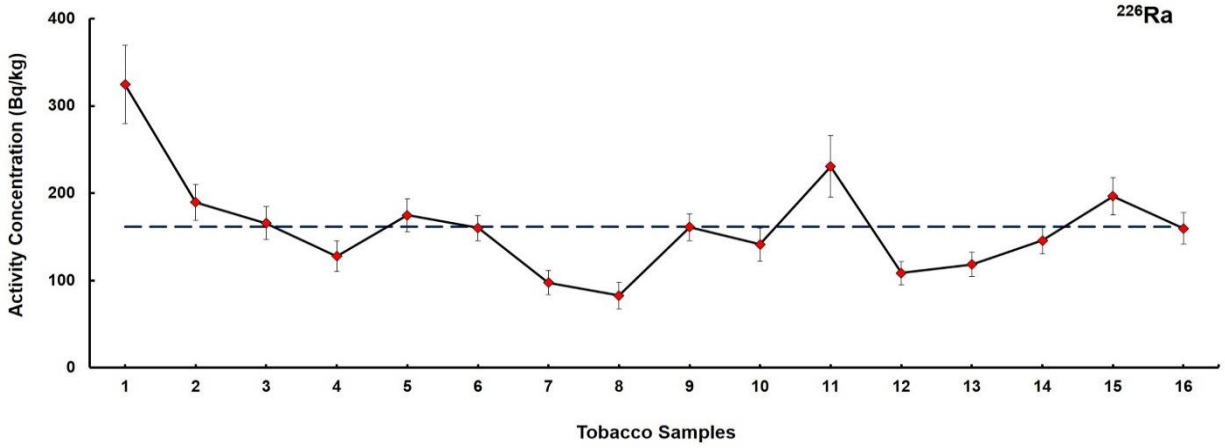

Figure 1. Distribution of radioactivity concentration of ^{226}Ra for tobacco samples and blue-dashed line represents mean value of ^{226}Ra for tobacco samples in this study.

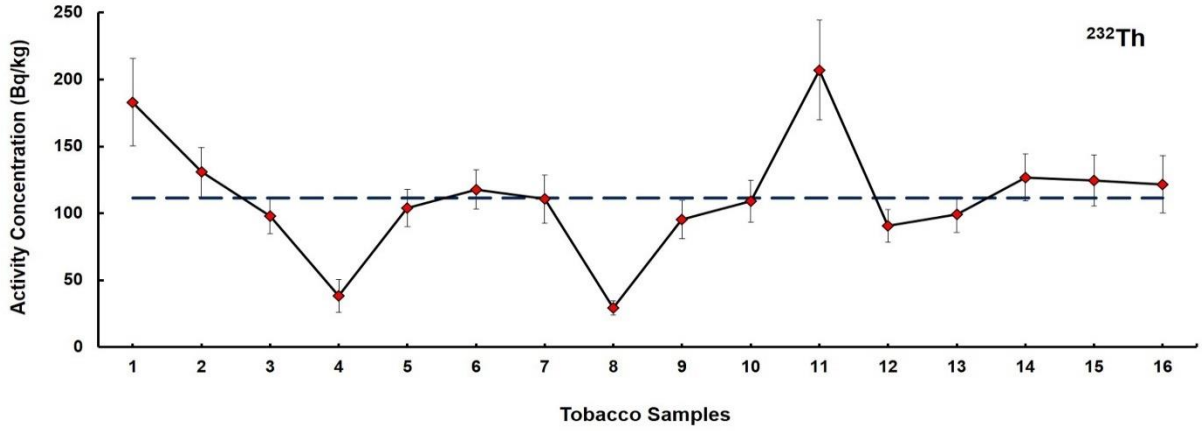

Figure 2. Distribution of radioactivity concentration of ^{232}Th for tobacco samples and blue-dashed line denotes mean value of ^{232}Th for tobacco samples in this study.

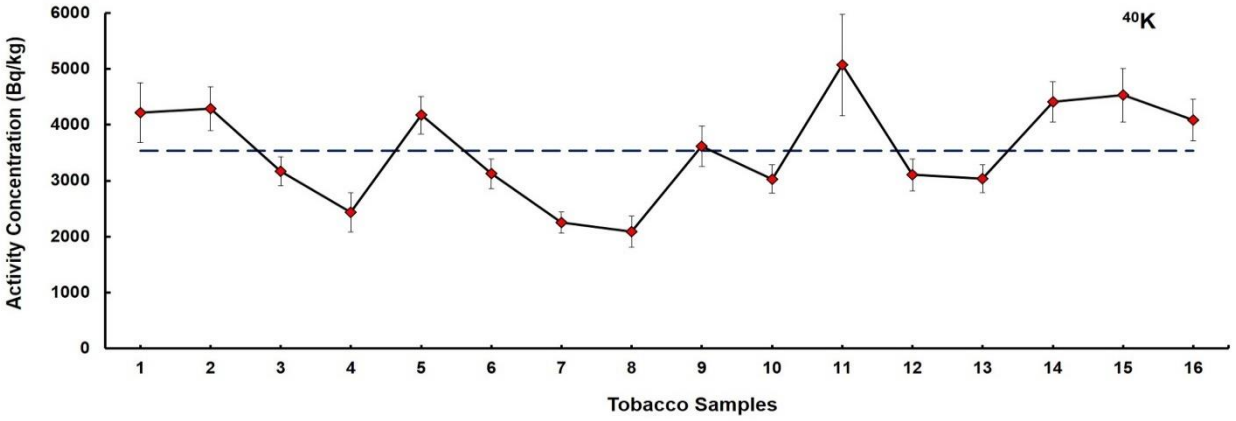

Figure 3. Distribution of radioactivity concentration of ^{40}K for tobacco samples and blue-dashed line denotes mean value of ^{40}K for tobacco samples in this study.

Values of radium equivalent activity (Ra_{eq}), annual effective dose (H_E) for ^{214}Pb, ^{214}Bi, ^{228}Ac and ^{40}K, seperately and total value of annual effective dose $(\sum H_E)$ and excess lifetime cancer risk *(ELCR)* of tobacco samples were calculated and these values are presented in Table 2.

Table 2. Annual effective dose evaluations for tobacco samples

Sample No	Ra_{eq} $(Bqkg^{-1})$	H_E (^{214}Pb) (μSvy^{-1})	H_E (^{214}Bi) (μSvy^{-1})	H_E (^{228}Ac) (μSvy^{-1})	H_E (^{40}K) (μSvy^{-1})	$\sum H_E$ $(mSvy^{-1})$	$ELCR$ $(x10^{-3})$
1	911	9.91	27.69	35.71	85.21	0.14	0.49
2	707	5.71	16.36	25.54	86.70	0.12	0.43
3	550	4.81	14.75	19.12	63.99	0.09	0.33
4	370	3.93	10.79	7.46	49.21	0.06	0.23
5	644	5.23	15.16	20.29	84.28	0.11	0.40
6	569	4.75	14.01	23.01	63.19	0.10	0.33
7	429	2.76	8.84	21.59	45.54	0.07	0.26
8	285	3.06	5.69	5.72	42.23	0.05	0.18
9	509	4.94	13.69	18.61	72.98	0.10	0.35
10	530	4.76	10.91	21.27	61.23	0.10	0.32
11	917	7.52	18.47	40.41	102.37	0.16	0.55
12	477.	3.25	9.37	17.71	62.73	0.09	0.30
13	493	3.57	10.17	19.33	61.35	0.09	0.31
14	667	4.13	13.25	24.75	89.19	0.12	0.43
15	723	5.99	16.80	24.33	91.50	0.13	0.45
16	648	4.29	15.05	23.74	82.53	0.12	0.41
Mean Value	594	4.91	13.81	21.79	71.51	0.10	0.36

Total annual effective dose (H_E) varies from 0.05 $mSvy^{-1}$ to 0.16 $mSvy^{-1}$, the mean value of total annual effective dose is 0.10 $mSvy^{-1}$. The value of ELCR gives information about possibility of growing cancer excess of lifetime at a certain exposure level and so this value has no units [4] .Value of excess lifetime cancer risk *(ELCR)* varies from 0.18×10^{-3} to 0.55×10^{-3}, the mean value of *ELCR* is 0.36×10^{-3} which is higher than mean value of 0.29×10^{-3} reported by the UNSCEAR report [1, 17]. Increasing value of ELCR brings about breast, prostate or even blood cancer [25]. Value of radium equivalent activity, total annual effective dose and excess lifetime cancer risk are the highest in sample-11 and sample-8 is the lowest.Comparison between natural radioactivity concentrations in tobacco samples with previous studies and this study is shown in Table 3.

Table 3. Natural radioactivity concentrations in tobacco samples other studies.

Country (samples collected)	^{238}U (^{226}Ra) (Bqkg^{-1})	^{232}Th (Bqkg^{-1})	^{40}K (Bqkg^{-1})	Ref.
Antalya Market (Turkey)	161.46±19.88	111.56±17.20	3539.82±366.43	Present Study
Iraqi Market	14.86 ±3.76	10.84 ± 3.13	1050.64 ± 47.57	[4]
Greece	2-8		273-2080	[19]
1 Pakistan, 1 Singapore, 1 Egypt, 1 Germany,1 Switzerland, 11 UK, 8 USA)	7.0	7.8	876	[22]
Baghdad Market	52.543 (^{214}Bi)	1.742 (^{228}Ac) 7.730 (^{212}Pb) 12.122 (^{212}Bi)	75.267	[26]
Serbian Market	9.4±3.2		1160±120	[27]

4. Conclusion and Outlook

Natural radioactivity concentration of ^{226}Ra, ^{232}Th, ^{40}K, radium equivalent activity, annual effective dose, total annual effective dose and excess lifetime cancer risk were calculated using high purity gemanium detector (HPGe).

As shown in Table-3, activity concentration of the ^{238}U, ^{232}Th and ^{40}K are the lowest value in Greece region [19] and Baghdad Market [28], respectively. In present study, ^{238}U, ^{232}Th and ^{40}K activity concentration of tobacco samples are higher than other studies. Because the activity concentration of natural radionuclides can be affected by the area where the tobacco is planted and the geological characteristics of the zone [28]. As a result, the activity concentrations of radionuclides may vary depending on the geological characteristics of the soil where tobacco plants are grown and the amount of fertilizer.

Distribution of particulate size, inhalation type, aerodynamic and electrical properties of particle, dissipation and solubility in the lung as well as the rate of the radionuclides in the smoking products are required to determine prediction of dose effect from smoking [22]. This study is first step to prediction of the dose effect from smoking. In the future, all these paramaters can be investigated.

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Conflicts of interest

The authors state that there is no conflict of interests.

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