Yuzuncu Yıl University
Journal of Agricultural Science

http://dergipark.gov.tr/yyutbd

Research Article (Araştırma Makalesi)

Determination of Radioactivity Levels in Different Mushroom Species from Turkey

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Article Info

Received: 18.09.2020
Accepted: 29.11.2020
Online Published 30.03.2021
DOI: 10.29133/yyutbd.797101

Keywords

Annual effective dose,
Lifetime cancer risk,
Mushroom,
Radioactivity,
Radionuclide.

Abstract: Radioactivity in the environment occurs due to natural, terrestrial, extra-terrestrial factors or caused by human activity. Foodstuffs such as plants and mushrooms that grown in the soil which containing radioactive elements can absorb radioactive elements from the soil. Wild mushrooms can accumulate many types of toxicological, nutritional, and radioactive elements. Knowing the levels of radioactivity in the foodstuffs is of great importance for the protection of human health. In this study, the activity concentrations of the naturally occurring ^{238}U, ^{232}Th, ^{40}K nuclides and artificially occurring ^{137}Cs nuclide were determined and annual effective doses and excess lifetime cancer risk values were calculated in mushrooms commonly consumed by the Turkish people. Fifteen types of mushroom samples were collected from different locations of Turkey. The results showed that the activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs varied from 9.2±1.6 to 75.4±8.8 Bq kg^{-1}, 10.9±1.6 to 76.3±8.9 Bq kg^{-1}, 925.9±29.0 to 3848.0±73.2 Bq kg^{-1} and 6.1±1.1 to 2824.8±79.8 Bq kg^{-1}, respectively. The mean total annual effective dose was found to be 11.5 µSv y^{-1}. ^{40}K radionuclide was the highest contributor to the mean total annual effective dose as 5.35 µSv y^{-1}. The mean excess lifetime cancer risk (ELCR) caused by consumption of mushrooms in the study was determined as 4.6 x10^{-5}.

Türkiye'den Farklı Mantar Türlerinde Radyoaktivite Seviyelerinin Belirlenmesi

Makale Bilgileri

Geliş: 18.09.2020
Kabul: 29.11.2020
Online Yayınlanma 30.03.2021
DOI: 10.29133/yyutbd.797101

Anahtar kelimeler

Yıllık etkin doz,
Yaşam boyu kanser riski,
Mantar,
Radyoaktivite,
Radyonüklid.

Öz: Radyoaktivite; doğal, karasal, dünya dışı faktörler veya insan aktivitesinden kaynaklanmaktadır. Radyoaktif elementler içeren toprakta yetişen bitkiler ve mantarlar gibi gıda maddeleri, topraktaki radyoaktif elemetleri absorbe edebilmektedir. Yabani olarak yetişen mantarlar pek çok türde toksikolojik, besleyici ve radyoaktif element biriktirebilmektedir. Gıda maddelerindeki radyoaktivite seviyelerinin bilinmesi insan sağlığının korunması açısından büyük önem taşımaktadır. Bu çalışmada, Türk halkı tarafından yaygın olarak tüketilen mantarlarda doğal olarak oluşan ^{238}U, ^{232}Th ve ^{40}K nüklidleri ile yapay olarak oluşan ^{137}Cs nüklidinin aktivite konsantrasyonları, yıllık etkin dozlar ve yaşam boyu kanser riski değerleri belirlenmiştir. Türkiye'nin farklı yerlerinden 15 çeşit mantar örneği toplanmıştır. Sonuçlar ^{238}U, ^{232}Th, ^{40}K ve ^{137}Cs aktivite konsantrasyonlarının sırasıyla 9.2±1.6 - 75.4±8.8 Bq kg^{-1}, 10.9±1.6 - 76.3±8.9 Bq kg^{-1}, 925.9±29.0 - 3848.0±73.2 Bq kg^{-1} ve 6.1±1.1 - 2824.8±79.8 Bq kg^{-1} arasında değiştiğini göstermiştir. Ortalama toplam yıllık etkin doz 11.5 µSv y^{-1} olarak bulunmuştur. ^{40}K radyonüklidi, ortalama toplam yıllık etkin doza 5.35 µSv y^{-1} ile

en yüksek katkıda bulunan radyonüklid olarak belirlenmiştir. Çalışmada yer alan mantarların tüketiminden kaynaklanan ortalama yaşam boyu kanser riski (ELCR) değeri 4.6 x10^{-5} olarak belirlenmiştir.

1. Introduction

Mushrooms have been used as an important food in the human nutrition for thousands of years all over the world. However, it is well known that mushrooms have the ability to efficiently accumulate high concentrations of radionuclides from the soil/substrate or environment owing to high surface area of their mycelia (Kalac, 2001; Falandysz and Borovicka, 2013). Wild mushrooms can be exposed to more radioactivity than cultivated mushrooms, and their radioactive element contents can be too high, particularly in areas heavily polluted with radionuclides. Accordingly, mushrooms can be used as a useful bioindicator to determine the level of radioactive contamination in any environment (Kuwahara et al., 2005; Bazala et al., 2008).

Nowadays, the radioactive pollution is a serious problem throughout the world (Hatra, 2018). The radioactive substances adversely affect the health of people, animals and plants and also disrupt the environment and ecological balance. People are continuously exposed to different radiation sources. The major sources of radioactive substances released into the environment are nuclear weapon tests, manufacture of radioactive compounds, nuclear power plants, radioactive waste disposal and nuclear researches. Radionuclides are the radioactive isotopes of elements, are unstable and have excess nuclear energy. Some of them toxic for the human health. They can cause many cancers, birth defects, and cognitive disabilities. Radionuclides may occur naturally or artificially. Humans are exposed to mostly natural radionuclides (Mazzilli et al., 2002). The uranium (^{234}U, ^{235}U and ^{238}U), potassium (^{40}K) and thorium (^{228}Th, ^{230}Th and ^{232}Th) are common natural radionuclides. On the other hand, ^{137}Cs is the most important artificially occurring radionuclide. It is released to the atmosphere as a result of nuclear experiments made for many years or nuclear accidents. It is a crucial fission product due to its high fission yield and its long half-life period (30.17 years). It is estimated that, approximately 3.8x10^{16} Bq of ^{137}Cs was released into the environment after the Chernobyl disaster (UNSCEAR, 1988).

Natural or artificial radiation sources can enter into people's food chain by the consumption of foods containing radionuclides. The wild foods including mushrooms, berries and fishes are principal sources of radionuclides for peoples. The radionuclide contents of mushrooms are generally higher than plants in the same forest (Bannai et al., 2005). Therefore, the determination of radioactivity in edible mushrooms is extremely important in recent years. Many radionuclides including ^{137}Cs, ^{40}K, ^{238}U, ^{234}U, ^{228}Th, ^{230}Th, ^{232}Th, ^{210}Po, ^{85}Sr, ^{90}Sr, ^{238}Pu, ^{226}Ra and ^{60}Co were investigated in the mushrooms in different countries of the world (Baeza et al., 2004; Mietelski et al., 2010; Taira et al., 2011; Castro et al., 2012).

As well as in all of Europe, the principal sources of radioactive elements in Turkey are the fallout from the nuclear weapon tests in the 1950s and 1960s, and from the Chernobyl accident in 1986. Turkey has great wild mushroom potential. Many wild edible mushroom species are fondly consumed by Turkish people. According to Yamaç et al. (2007), Turkey is an important exporter of wild edible mushrooms. Wild edible mushrooms grown in Turkey are exported to mainly European countries, The United States of America, Japan, Central Asia and Middle East countries (Pekşen and Akdeniz, 2012). The radiation certificate is required to export the wild edible mushrooms and the radionuclide contents of mushrooms must be within safe limits for human health. So, it is required to research the levels of radioactive contents in the mushrooms grown in Turkey. However, there are limited works concerning determining the levels of toxicologically important natural and artificial radionuclides such as ^{238}U, ^{232}Th, ^{40}K, ^{134}Cs and ^{137}Cs in the mushrooms grown in Turkey (Karadeniz and Yaprak, 2007; Turhan et al., 2007; Akça, 2011; Akça et al., 2014; Yılmaz et al., 2016; Türkekul et al., 2018; Gürgen et al., 2019).

The objective of this study was to determine the activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides and to calculate annual effective doses and excess lifetime cancer risks in some commonly consumed wild growing and cultivated mushrooms collected from different locations of Turkey.

2. Material and Methods

2.1. Collection of mushrooms and preparation of the samples for radiometric analyzes

The detailed information about mushroom species examined in the study is given in Table 1. These mushroom species except for *Ganoderma lucidum* are commonly grown and consumed in Turkey (Pekşen and Akdeniz, 2012). *G. lucidum* is the most important medicinal mushroom species both in the world and in Turkey (Bulam et al., 2019). The sporocarps of wild edible (*Boletus edulis*, *Craterellus cornucopioides*, *Lactarius deliciosus*, *Laetiporus sulphureus*, *Marasmius oreades*, *Morchella conica, Ramaria botrytis, Tricholoma terreum*) and cultivated mushrooms (*Ganoderma lucidum*, *Hericium erinaceus, Lentinula edodes, Pleurotus ostreatus*-1, 2, 3 and 4) were collected from different locations of Turkey in spring and autumn of 2015-2016. The identifications of the mushroom species were made according to Phillips (1994). After collection, mushroom samples (approximately 0.5 kg for each species) brought to the laboratory were cleaned from the soil, forest debris or substrate debris by gently. Then, whole sporocarps (pileus + stipe) were cut into small pieces, placed ina paper envelope and dried in an oven at 65°C to a constant weight. Afterward, dried mushroom samples were ground into a fine powder using a laboratory mill. The ground samples were put into polyethylene bags, sealed and stored in a refrigerator at 4°C until analysis.

Table 1. Mushroom species examined in the study

No.	Mushroom Species	Family	Local Name	Location	Ecology	Growing	Edilebility
1	*Boletus edulis*	Boletaceae	Ayı mantarı	Giresun	Mycorrhizal	Wild	Edible
2	*Craterellus cornucopioides*	Cantharellaceae	Borazan mantarı	Samsun, Ladik	Saprophytic	Wild	Edible
3	*Ganoderma lucidum*	Ganodermataceae	Reishi	Denizli, Agroma	Saprophytic /Parasitic	Cultivated	Not edible, but medicinal
4	*Hericium erinaceus*	Hericiaceae	Aslan yelesi mantarı	Ondokuz Mayis University	Parasitic	Cultivated	Edible
5	*Lactarius deliciosus*	Russulaceae	Kanlıca mantarı	Giresun, Bektaş plateau	Mycorrhizal	Wild	Edible
6	*Laetiporus sulphureus*	Polyporaceae	Kükürt mantarı	Giresun, Bulancak	Saprophytic /Parasitic	Wild	Edible
7	*Lentinula edodes*	Tricholomataceae	Shiitake, Meşe mantarı	Denizli, Agroma	Saprophytic	Cultivated	Edible
8	*Marasmius oreades*	Marasmiaceae	Cincile, Mıh tepesi	Sinop	Saprophytic	Wild	Edible
9	*Morchella conica*	Morchellaceae	Kuzu göbeği mantarı	Samsun, Vezirköprü	Mycorrhizal/ Saprophytic	Wild	Edible
10	*Pleurotus ostreatus*	Pleurotaceae	Kayın mantarı, Kavak mantarı	Giresun, Eynesil	Saprophytic	Cultivated (Poplar log)	Edible
11	*Pleurotus ostreatus*	Pleurotaceae	Kayın mantarı, Kavak mantarı	Rize	Saprophytic	Cultivated (Poplar log)	Edible
12	*Pleurotus ostreatus*	Pleurotaceae	Kayın mantarı, Kavak mantarı	Bursa	Saprophytic	Cultivated	Edible
13	*Pleurotus ostreatus*	Pleurotaceae	Kayın mantarı, Kavak mantarı	Rize	Saprophytic	Cultivated (Beech log)	Edible
14	*Ramaria botrytis*	Gomphaceae	Pürpürüm	Samsun, Ladik	Mycorrhizal	Wild	Edible
15	*Tricholoma terreum*	Tricholomataceae	Karakız mantarı	Samsun, Vezirköprü	Mycorrhizal	Wild	Edible

2.2. Gamma-ray measurements

Gamma-ray spectrometric analyzes of the samples were performed by using FoodGuard-1 3 x 3-inch NaI (Tl) model radiation detector (ORTEC, Oak Ridge, USA) in the Central Research Laboratory of Kastamonu University. The dried and powdered mushroom samples were placed into plastic boxes with a diameter of 8 cm and a height of 8 cm, prepared in accordance with the geometry of the detector. Then, the mouths of the boxes were tightly closed and the boxes were waited for 1 month. Thus, the formation of radioactive equilibrium between ^{238}U, and ^{232}Th and their decay products was provided and the samples were made ready for counting. The detector was calibrated before the analysis of the samples was started. To analyze the spectra collected in computer memory, it must be known which channel corresponds to which energy. Thus, the types of radioactive nuclei present in the sample can be found.

To accomplish the energy calibration, it is needed a standard source or resources consisting of nuclei previously known their energies. After calibration was completed, each sample was counted in the gamma spectrometer for 50000 sec.

The energy resolution of the detector measured in terms of full width at half maximum is 46 keV at 661.8 keV 137Cs. The light decay time constant for a NaI (Tl) crystal is about 0.23 ms. Typical charge-sensitive preamplifiers translate this into an output voltage pulse with a rise-time of about 0.5 ms. The pulse processing and data analysis system matched to a computer and the spectral analysis was performed using ScintiVision computer software. The detector was housed in a cylindrical lead shield of about 22 cm diameters, 56 cm in overall height and 7 cm thickness. The efficiency calibration of the spectrometer was performed using the IAEA gamma-ray spectrometry reference materials of RG sets (RGU-1, RGTh-1 and RGK-1). The energy calibrations and quality assurance of measurements were carried out by periodical calibrations using calibration sources that contained ^{133}Ba, ^{109}Cd, ^{57}Co, ^{22}Na, ^{137}Cs, ^{54}Mn and ^{60}Co and generated photon emission peaks in the energy range between 80 and 1400 keV and repeating sample measurements. The gamma photopeak of the 2614.4 keV from ^{208}Tl was used to measure the activity concentration of ^{232}Th, while the gamma photopeak of the 1764.5 keV from ^{214}Bi was used to measure the activity concentration of ^{226}Ra. The activity concentrations of ^{40}K and ^{137}Cs were measured directly by its own gamma photopeak at 1460.8 and 661.8 keV, respectively. After correcting for background and Compton contribution, the activity concentrations per unit mass of the above radionuclides were obtained for each sample in units of Bq kg^{-1}. All results in this research are based on dry weight masses (Kurnaz et al., 2016). The combined standard uncertainty of the activity concentration is calculated by the following formula:

$$\Delta A = A \cdot \sqrt{\left(\frac{\Delta C}{C}\right)^2 + \left(\frac{\Delta P_\gamma}{P_\gamma}\right)^2 + \left(\frac{\Delta \varepsilon}{\varepsilon}\right)^2 + \left(\frac{\Delta M}{M}\right)^2} \quad (1)$$

where A and ΔA are, respectively, the activity concentration and its uncertainty; C and ΔC are the count rate and its uncertainty; Pγ and ΔPγ are the gamma emission probability and its uncertainty, Ɛ and ΔƐ are the absolute efficiency of the detector and its uncertainty; M and ΔM are the mass and its uncertainty. The minimum detectable activity (MDA) of the gamma-ray measurement system was calculated by using the following formula (Altıkulaç et al., 2016):

$$MDA = \frac{4.66 \cdot \sqrt{B}}{\varepsilon \, P_\gamma \, .T.M} \quad (2)$$

where, B is the background counts, Ɛ is the absolute efficiency of the detector, Pγ is the gamma emission probability and T is the counting time (s) and M is the mass of the sample (kg). The average value of the MDA for ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs was found as 2.8, 3.4, 8.2 and 1.6 Bq kg^{-1}, respectively.

2.3. Calculation of annual effective dose and excess lifetime cancer risk

The annual effective dose was determined using the following equation as reported by Abbady (2006).

$$AEDE = RAC \times AAMC \times DCC \quad (3)$$

where: AEDE - annual effective dose (μSv y^{-1}), RAC - radionuclide activity concentration (Bq kg^{-1}), AAMC - average annual mushroom consumption (kg y^{-1}), DCC - dose conversion coefficient (Sv Bq^{-1}). In the above equation, average annual mushroom consumption for an adult Turkish people was regarded as 0.4 kg y^{-1} (Çevik, 2019). The dose conversion coefficient were 4.5×10^{-8} Sv Bq^{-1} for ^{238}U, 2.3×10^{-7} Sv Bq^{-1} for ^{232}Th, 6.2×10^{-9} Sv Bq^{-1} for ^{40}K and 1.3×10^{-8} Sv Bq^{-1} for ^{137}Cs (IAEA, 2011).

The excess lifetime cancer risk was determiden by using the following formula.

$$ELCR = AEDE \times DL \times CRF \quad (4)$$

where: ELCR - excess lifetime cancer risk, AEDE - annual effective dose equivalent (μSv y^{-1}), DL - duration of life (y), CRF - cancer risk factor (Sv^{-1}). In the above formula, duration of life was considered as 70 year. Also, cancer risk factor was regarded as 0.057 (ICRP, 2007).

2.4. Assessment of data

For each mushroom species, analyses were performed in triplicate. The mean values and standard deviations were determined and the results were indicated as means ± standard deviations. Principal Components Analysis Biplot (PCA Biplot) was established by using XLSTAT Version 2016.

3. Results

The activity concentrations of radionuclides, annual effective doses and excess lifetime cancer risk values in the examined mushrooms are presented in Table 2. The mean activity concentrations of radionuclides across all the mushrooms studied were in the decreasing order: $^{40}K > ^{137}Cs > ^{238}U > ^{232}Th$.

The accumulation of ^{238}U by different mushroom species was very variable. The activity concentrations of ^{238}U in the mushrooms were found to be ranging from 9.2 to 75.4 Bq kg^{-1}. *T. Terreum* had the lowest activity concentration of ^{238}U, while the highest activity concentration of ^{238}U was determined in *P.* ostreatus-1. It was closely followed by *M. oreades.*

With regard to ^{232}Th, activity concentrations of this radionuclide in the mushroom species varied from 10.9 (*T. terreum*) to 76.3 Bq kg^{-1} (*M. conica*). The activity concentration of ^{232}Th in *M. conica* was seven times that of *T. terreum*

All of the analyzed mushrooms had considerable high activity concentrations of ^{40}K radionuclide. The lowest and the highest activity concentrations for ^{40}K were observed in *C. cornucopioides* (925.9 Bq kg^{-1}) and *L. edodes* (3848.0 Bq kg^{-1}), respectively. Although potassium element found abundant in mushrooms is an essential mineral element for humans, the natural isotope ^{40}K is toxic for health.

The levels of ^{137}Cs in investigated mushrooms had highly high variability ranging from 6.1 to 2824.8 Bq kg^{-1}. Among the studied mushroom species, the lowest activity concentration of ^{137}Cs was determined in *P. ostreatus*-4. On the contrary, *M. conica* which is a commercially very important species for Turkey accumulated by far the highest level of ^{137}Cs.

The mean activity concentrations of ^{137}Cs and ^{238}U in wild mushrooms were found to be higher than cultivated mushrooms. Conversely, activity concentrations of ^{40}K and ^{232}Th were higher in cultivated mushrooms when compared with wild ones. In this study, radioactivity of cultivated mushrooms was caused mostly by natural ^{40}K radionuclide.

As seen in Table 2, the total annual effective dose values in the mushrooms varied widely ranging from 4.49 (*T. terreum*) to 30.20 μSv y^{-1} (*M. conica*). The mean total annual effective dose was found to be 11.5 μSv y^{-1}. ^{40}K radionuclide (5.35 μSv y^{-1}) was the highest contributor to the total annual effective dose. It was followed by ^{232}Th (4.13 μSv y^{-1}). The annual effective dose values for ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs radionuclides in the mushrooms observed in this study ranged from 0.17 (*T. terreum*) to 1.36 μSv y^{-1} (*M. oreades*), 1.00 (*T. terreum*) to 7.02 μSv y^{-1} (*M. conica*), 2.30 (*C. cornucopioides*) to 9.54 μSv y^{-1} (*L. edodes*), and 0.03 (*P. ostreatus*-4) to 14.69 μSv y^{-1} (*M. conica*), respectively.

Table 2. The activity concentrations of radionuclides, annual effective dose and lifetime cancer risk values in various mushroom species

Mushroom species	Activity concentrations of radionuclides (Bq kg^{-1})				Annual effective dose (μSv y^{-1})					Excess lifetime cancer risk
	^{238}U	^{232}Th	^{40}K	^{137}Cs	^{238}U	^{232}Th	^{40}K	^{137}Cs	Total	
B. edulis	52.5±4.4	33.5±3.9	2306.3±43.9	53.3±12.0	0.94	3.08	5.72	0.28	10.00	4.00×10^{-5}
C. cornucopioides	58.5±4.1	29.8±4.9	925.9±29.0	90.7±12.9	1.05	2.74	2.30	0.47	6.56	2.62×10^{-5}
G. lucidium	57.2±3.9	56.2±6.2	3288.9±72.1	24.1±5.1	1.03	5.17	8.16	0.13	14.50	5.78×10^{-5}
H. erinaceus	13.6±2.2	58.9±9.7	2442.4±77.1	156.0±9.7	0.25	5.42	6.06	0.81	12.50	5.00×10^{-5}
L. deliciosus	69.4±7.8	49.0±8.4	2885.7±74.5	25.7±2.2	1.25	4.51	7.16	0.13	13.00	5.21×10^{-5}
L. sulphureus	27.4±6.9	53.0±5.2	1816.7±25.1	17.6±1.7	0.49	4.88	4.50	0.09	9.97	3.98×10^{-5}
L. edodes	37.6±4.7	20.9±1.7	3848.0±73.2	87.2±6.7	0.68	1.92	9.54	0.45	12.60	5.03×10^{-5}
M. oreades	75.3±7.6	47.4±6.8	1415.2±67.2	18.5±1.9	1.36	4.36	3.51	0.10	9.32	3.72×10^{-5}
M. conica	66.9±7.9	76.3±8.9	2922.3±65.6	2824.8±79.8	1.20	7.02	7.23	14.69	30.20	12.00×10^{-5}
P. ostreatus-1	75.4±8.8	64.3±6.1	3133.3±68.1	25.3±2.3	1.36	5.92	7.77	0.13	15.20	6.05×10^{-5}
P. ostreatus-2	26.4±2.3	58.1±7.9	1672.7±53.0	44.9±1.7	0.47	5.35	4.15	0.23	10.20	4.07×10^{-5}
P. ostreatus-3	49.8±3.1	22.2±1.9	1744.4±75.4	32.6±2.3	0.90	2.04	4.33	0.17	7.43	2.97×10^{-5}
P. ostreatus-4	53.1±6.7	37.9±1.6	1547.2±56.7	6.1±1.1	0.96	3.49	3.84	0.03	8.31	3.32×10^{-5}
R. botrytis	25.2±2.3	54.4±6.8	1268.4±39.5	13.1±1.2	0.45	5.00	3.15	0.07	8.67	3.46×10^{-5}
T. terreum	9.2±1.6	10.9±1.6	1145.0±58.0	92.7±5.1	0.17	1.00	2.84	0.48	4.49	1.79×10^{-5}

Excess lifetime cancer risk values (ELCR) in investigated mushrooms were between 1.79 x 10^{-5} and 12.00 x 10^{-5} and were in the following descending order: *M. conica* > *P. ostreatus*-1 > *G. lucidium* > *L. deliciosus* > *L. edodes* > *H. erinaceus* > *P. ostreatus*-2 > *B. edulis* > *L. sulphureus* > *M. oreades* > *R. botrytis* > *P. ostreatus*-4 > *P. ostreatus*-3 > *C. cornucopioides* > *T. terreum.* There was no remarkable difference in terms of the mean lifetime cancer risk values between wild growing (4.59 x 10^{-5}) and cultivated mushrooms (4.60 x 10^{-5}). According to the values observed, the mean excess lifetime cancer risk of fifteen mushroom samples was determined as 4.6 x10^{-5} (Table 2).

The results of the PCA analysis are presented in Table 3 and 4. The first three principal components, which are the most important components, explained 100% of the total variability for activity concentrations and annual effective doses of radionuclides in examined mushroom species. In addition, biplots of the first two principal components based on PCA for activity concentrations and annual effective doses of radionuclides in examined mushroom species are shown in Figure 1 and 2. Biplot graphics provide visual evaluation of the relationships between mushroom species and characteristics. There were two values of each principal component, positive and negative. The blue points in Figure 1 and Figure 2 represent the activity concentrations and annual effective doses of radionuclides (n = 45). With regard to the activity concentrations of radionuclides, *M. conica* was quite far from other species and the biplot origin. *T. terreum, H. erinaceus* and *C. cornucopioides* were quite close to each other. Except for the mentioned above 4 mushroom species, all species showed similar characteristics in terms of the activity concentrations of radionuclides. *M. conica* was also far from other species with regard to the annual effective doses of radionuclides. *G. lucidium* and *L. deliciosus* were the closest species to each other.

Table 3. Principal component analysis of activity concentrations of radionuclides for examined mushroom species

	PC axis		
	PC1	PC2	PC3
Eigen values	14.364	0.634	0.002
Explained variance (%)	95.758	4.227	0.016
Cumulative variance (%)	95.758	99.984	100.000

Table 4. Principal component analysis of annual effective doses of radionuclides for examined mushroom species

	PC axis		
	PC1	PC2	PC3
Eigen values	12.391	1.574	1.035
Explained variance (%)	82.608	10.493	6.900
Cumulative variance (%)	82.608	93.100	100.000

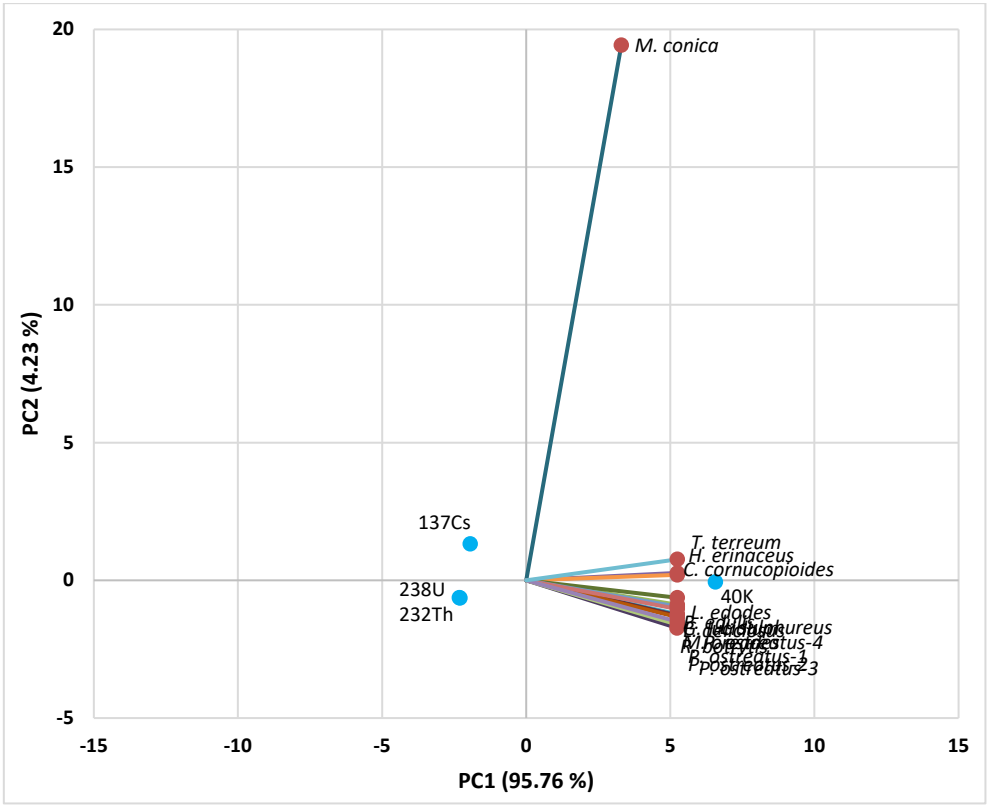

Figure 1. Biplot graph of the first two principal components for activity concentrations of radionuclides in examined mushroom species.

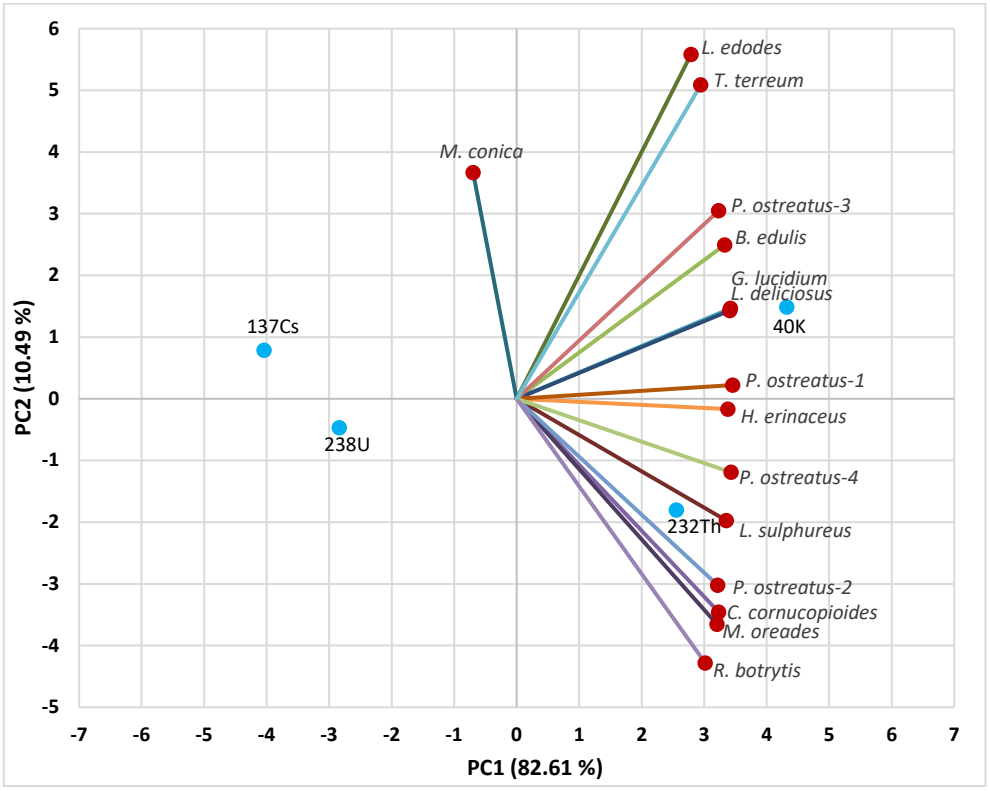

Figure 2. Biplot graph of the first two principal components for annual effective doses of radionuclides in examined mushroom species.

4. Discussion and Conclusion

In this study, the mean activity concentrations of radionuclides across all the mushrooms studied were in the decreasing order: $^{40}K > ^{137}Cs > ^{238}U > ^{232}Th$. In previous studies, activity concentrations of ^{40}K in the mushrooms were also found higher than that of other radionuclides examined (Akça, 2011; Changizi et al., 2012; Akça et al., 2014; Yılmaz et al., 2016; Gürgen et al., 2019).

The activity concentrations of ^{238}U in the mushrooms were found to be varied from 9.2 to 75.4 Bq kg^{-1}. The obtained values in this study related to the activity concentrations of ^{238}U in the mushroom samples were in accordance with the results of Baeza et al. (2006). The range for ^{238}U was 0.0011-0.270 Bq kg^{-1} in Poland and the Czech Republic (Mietelski et al., 2002; Borovicka et al., 2011). However, our results were higher than those of Akça (2011), Akça et al. (2014) and Yılmaz et al. (2016). ^{238}U values of mushrooms were found to be ranging from 19-168 Bq kg^{-1} (Türkekul et al., 2018) and 14.6-26.6 Bq kg^{-1} (Gürgen et al., 2019).

With regard to ^{232}Th, activity concentrations of this radionuclide in the mushroom species varied from 10.9 to 76.3 Bq kg^{-1}. These results were compatible with Rosa et al. (2011), who reported that the activity concentrations of ^{232}Th in wild mushrooms collected from different locations of Brazil were 0.6-142 Bq kg^{-1}. ^{232}Th was reported in Poland and the Czech Republic within the range 0.0008-0.496 Bq kg^{-1} (Mietelski et al., 2002; Borovicka et al., 2011). Our results were higher than the values reported by Akça (2011), Akça et al. (2014) and Yılmaz et al. (2016) for mushrooms collected from different provinces of Turkey.

The values determined in this study for activity concentrations of ^{40}K (925.9-3848.0 Bq kg^{-1}) were similar to the results of Mietelski et al. (2010) for different mushroom species collected from Poland. Karadeniz and Yaprak (2010) found that activity concentration of ^{40}K were 588-2024 Bq kg^{-1} in 25 different mushrooms collected from Turkey. Castro et al. (2012) determined to be as 461-1535 Bq kg^{-1} activity concentration of ^{40}K in 17 mushroom samples belonging to *Agaricus*, *Pleurotus* and *Lentinula* species in Brazil. The minimum and maximum values for ^{40}K of 50 mushroom samples obtained from the same station were 201-966 Bq kg^{-1} (Türkekul et al., 2018). ^{40}K values of *Agaricus bisporus* and *Pleurotus ostreatus* mushrooms cultivated in different commercial companies were varied between 330.4-739.7 Bq kg^{-1} (Gürgen et al., 2019). In contrast to our results, Gaso et al. (2000), Akça (2011), Akça et al. (2014) and Yılmaz et al. (2016) observed lower activity concentrations of ^{40}K in various mushrooms.

The activity concentrations of natural radionuclides for agricultural products can be until is 2000 Bq kg^{-1} according to WHO (1989). So, the mushrooms investigated in this study do not cary any health risk for consumers in terms of ^{238}U and ^{232}Th. However, mushroom species with high activity concentrations of ^{40}K such as *L. edodes*, *G. lucidium*, *P.* ostreatus-1 should be consumed with caution. On the other hand, radionuclide concentration in the portion may decrease due to the procedures to be applied according to the consumption type of the mushrooms such as soaking, cooking, boiling, freezing, drying, pickling or blanching (Saba and Falandysz, 2020).

The levels of ^{137}Cs in investigated mushrooms ranged from 6.1 to 2824.8 Bq kg^{-1}. Yoshida et al. (2004) reported that the activity concentrations of ^{137}Cs in wild mushrooms significantly increased after Chernobyl nuclear accident. Baeza et al. (2004) stated that the ^{137}Cs accumulation among nutritional types of mushrooms was in the following order: mycorrhizal > saprophytic > parasitic. When compared to other radionuclides, ^{137}Cs were investigated more intensively in wild mushrooms in different countries of the world. The levels of ^{137}Cs for many mushroom species were found in the ranges of 9.84-401 Bq kg^{-1} in Turkey (Karadeniz and Yaprak, 2007), 2-15000 Bq kg^{-1} in Germany (Kammerer et al., 1994), 2-1357 Bq kg^{-1} in Mexico (Gaso et al., 1998), 110-4290 Bq kg^{-1} in Poland (Malinowska et al., 2006), 167-376 Bq kg^{-1} in Norway (Gwynn et al., 2013) and 200-19900 Bq kg^{-1} in Finland (Lehto et al., 2013). Our findings were within the limits reported in the studies mentioned above. The guideline level reported by IAEA (2016) for activity concentration of ^{137}Cs is 1000 Bq kg^{-1}. Accordingly, all other species except for only *M. conica* among mushroom species studied in this study did not exceed the permissible limit.

The accumulation of radionuclides by mushrooms is actually an extremely complicated event that depends on many factors such as ecological/environmental factors, mushroom species/strain, structure and characteristics of substrate/soil, concentration of radionuclides in the substrate/soil, development and age of mycelium, the maturity degree of fruiting body, fruiting body size,

morphological parts of fruiting body, the nutritional type of mushroom (mycorrhizal, saprophytic, parasitic), distance from any source of pollution and time after the disaster or fallout (Kalac and Svoboda, 2000; Kuwahara et al., 2005; Guillen and Baeza, 2014).

The total annual effective dose values in the mushrooms varied from 4.49 to 30.20 μSv y^{-1}. Our values were higher than those of Karadeniz and Yaprak (2010) and Akça et al. (2014). However, our results were lower than the reported levels by Yılmaz et al. (2016). The annual effective dose values obtained in this study were found to below reference level (1 mSv y^{-1}) reported by ICRP (2007).

Excess lifetime cancer risk values (ELCR) in investigated mushrooms were between 1.79 x 10^{-5} and 12.00 x 10^{-5}. The mean excess lifetime cancer risk of fifteen mushroom samples was determined as 4.6 x10^{-5}. These results were lower than that of Yılmaz et al. (2016). According to Taskin et al. (2009), the avarege excess lifetime cancer risk value in the world is 0.29 x 10^{-3}. Therefore, our results were lower than avarege excess lifetime cancer risk value of world.

Turkey has a very rich wild mushroom diversity. Many wild edible mushroom species are fondly consumed as a popular and favourite delicacy in Turkey. However, mushrooms can accumulate toxicologically significant amounts of radionuclides. Therefore, the determination of radionuclide concentrations in the edible mushrooms is highly important. In this study, the activity concentrations of natural (^{238}U, ^{232}Th ^{40}K) and artificial (^{137}Cs) radionuclides, annual effective doses and excess lifetime cancer risks in various mushroom species collected from Turkey were determined.

The results revealed that the activity concentrations of radionuclides varied considerably depending on the species. The activity concentrations of radionuclides of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs were found in the ranges of 9.2-75.4 Bq kg^{-1}, 10.9-76.3 Bq kg^{-1}, 925.9-3848.0 Bq kg^{-1} and 6.1-2824.8 Bq kg^{-1}, respectively. The mean activity concentrations of ^{137}Cs and ^{238}U in wild mushrooms were found to be higher than cultivated mushrooms. The mean total annual effective dose in mushroom samples was found to be 11.5 μSv y^{-1}. The mean excess lifetime cancer risk of mushrooms was determined as 4.6x10^{-5}. The considerable consumption of mushroms which have high concentrations of radionuclides is toxic. In the present study, *M. conica* collected from Samsun accumulated fairly high levels of ^{137}Cs. So, this species should be consumed with caution. In conclusion, most of the mushrooms analyzed in this study can be consumed safely.

Acknowledgment

This study was published as a summary report in the Abstract Book of Ecology Symposium, 11-13 May 2017 Kayseri, Turkey.

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