Alqalam Journal of Medical and Applied Sciences. 2025;8(4):2330-2340
https://doi.org/10.54361/ajmas.258434

Original article

Assessment of Natural Radioactivity and Health Risk Indices in Commercial Tea Samples from Libyan Markets Using Gamma-Ray Spectrometry

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Abstract
This study aims to evaluate the concentration of naturally occurring radioactive elements in commercial tea samples available in Libyan markets using gamma-ray spectrometry. Twelve different tea brands were analyzed to determine the levels of Uranium-238 (U-238), Thorium-232 (Th-232), and Potassium-40 (K-40). The average activity concentrations for each of the examined samples. The levels of the detected elements were as follows: (7.07 - 54.74), (2.14 - 31.42), (12.74 - 50.95), and (63.48 - 156.12 Bq kg-1) for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, respectively. The results of the specific activity concentrations of radionuclides ^{226}Ra and ^{40}K for all tea samples are lower than the world recommended values 50 and 15 Bqkg^{-1}, respectively. But the higher value of activity concentration of ^{238}U and ^{232}Th for most samples under investigation was lower than the internationally recommended value of 20 and 420 Bq kg-1. The results showed varying concentrations across samples, but all were within internationally accepted safety limits, indicating no significant radiological health risk from consuming these products. The results revealed that the differences in the radioactive levels in the studied tea samples were mainly attributed to many reasons as the use of pesticides, water irrigation, type of soil around the collections of tea samples
Keywords. Natural Radioactivity, Gamma-ray Spectrometry, Tea Imported.

Introduction

Naturally occurring radioactive elements are present in the environment and can accumulate in plants through soil and water absorption. Given the widespread consumption of tea as a daily beverage, assessing its radioactive content is essential to evaluate potential long-term exposure risks. assessed the levels of naturally occurring radioactive nuclides and their associated effective radiation doses in commonly consumed foodstuffs in Kut, Iraq. They analyzed twenty-nine food samples that were collected from Wasit Governorate Markets for uranium (U-238), thorium (Th-232), and potassium (K-40) using a gamma-ray spectroscopy system equipped with a 3x3 inch sodium iodide (Tl) detector. The results revealed that the concentrations of these radionuclides and the calculated annual effective dose, equivalent radium concentration, and external and internal risk factors were within acceptable limits, despite variations in the concentrations across different samples [1].

Tea, a beloved beverage consumed globally, originates from the Camellia plant. The tender shoots, leaves, and buds of this plant are harvested and processed to yield a diverse array of tea types. Beyond traditional loose-leaf tea, the market is inundated with a plethora of tea products, encompassing flavored, instant, and ready-to-drink varieties. These convenient tea options are often packaged in eye-catching glass containers, catering to the modern consumer's preference for on-the-go consumption [2]. Some studies assessed the levels of naturally occurring radioactive nuclides and their associated effective radiation doses in commonly consumed foodstuffs in Kut, Iraq. They analyzed twenty-nine food samples that were collected from the Waist Governorate Markets for uranium (U-238), thorium (Th-232), and potassium (K-40) using a gamma-ray spectroscopy system equipped with a 3x3 inch sodium iodide (Tl) detector. The results revealed that the concentrations of these radionuclides and the calculated annual effective dose, equivalent radium concentration, and external and internal risk factors were within acceptable limits, despite variations in the concentrations across different samples [3].

The determination of radioactive elements in different samples was taken in different studies as soils, plants, and water [4-6]. In Libya, the environmental studies on different health risk compounds as hydrocarbons [7-15], heavy metals [16-44]. Plant extracts and their chemical and biological effects [45-87], pesticides [88] were established. The aim of this study is to measure the levels of radioactive compounds in some imported tea samples collected from local markets in Libyan cities.

Methods

Sampling: Twelve tea samples were collected from markets in Al-Bayda city, Libya. The types of the studied were given in the (Table 1).

Samples Preparation and Measurements

The samples were dried in an oven at 95°C for 2 hours to eliminate residual moisture. Subsequently, the dried samples were ground into a fine powder using a mortar and pestle and sieved through a fine mesh to

Received*: 15-08-2025 -* ***Accepted****: 14-10-2025 -* ***Published****: 20-10-2025*

obtain a homogenous composite sample (Faanu et al., 2010). The samples were weighed and placed in 250 cm^3 polyethylene bottles. These samples were then transported and stored for approximately 30 days to establish secular equilibrium between the ^{238}U and ^{232}Th series and their respective daughter radionuclides.

Table 1. The studied tea samples.

Sample. No	Name
1	Barka
2	AlZahra
3	Al Taj
4	AL Aross
5	ALNajma
6	AlMorog
7	AlDalil
8	AlASeel
9	AlOadg
10	AlWardsa
11	ALShera
12	AlRaeg

Gamma Spectroscopy

The detection technique for gamma rays is determined by the gamma-ray photons with the detector material. Although the photoelectric effect and Compton scattering are the most prominent types that play essential roles in photon detection. The main gamma-ray photons and secondary scattered photons interact with the detector atoms, generating fast electrons with an energy proportional to that of the source photon within the detector volume. These fast electrons can then produce, as they move through the detector volume, secondary electrons. These electrons are secondary electrons. may be gathered and used to create electrical pulses. A preamplifier can be used to transform the charges.

A voltage proportional to the amount of initial gamma-ray energy deposited in the detector. A functional gamma spectroscopy detector must perform two independent roles. To begin, the detector must function as a radiation converter medium with a high chance of contact in order to create fast electrons, which must be able to flow quickly through the detector. The detector's background noise must be low enough to collect data; second, the detectors. Only electrons with the same energy as the observed gamma-ray photons are counted. If the detector can withstand a strong electric field and function at a low temperature, this can be accomplished.

The Sodium Iodide Detector (NaI) Detector

Gamma spectroscopy systems that employ Nal as a detection material are considered suitable in-situ measuring methods because they operate at room temperature. The electrical pulses are formed by converting the electrons that are generated through the radiation interaction into photoelectrons. The energy of the gamma-ray photons is deposited in the scintillator (a fluorescence material), causing atoms to rise to excited states. In Nal, the energy of excited electrons crossing the energy gap from the normally full valence band to the empty conduction band is roughly 4 eV. The excited atoms eventually lose their energy by producing visible photons, and the electrons return to the valence band.

Photoelectrons are created when visible photons released as a result of this DE excitation reach a photosensitive surface. Pulses of electricity using a photomultiplier tube, the photoelectrons are multiplied and accelerated**.** Self-absorption of visible photons can occur in a pure Nal crystal. Small amounts of impurities called "activators" are added to the crystal to diminish this effect and improve the possibility of visible photon transmission, such as the element thallium (Tl). The wavelength of Nal(Tl) is increased by adding activators to pure Nal crystals. Maximum visible photon emission from 303 to 410 nm while decreasing scintillation light self-absorption within the crystal.

Description of The System

Sodium iodide scintillation NaI (TL) detectors are used to measure the samples. The detector was inserted in the cylindrical lead shield to reduce the background and noise radiation from many natural radionuclides such as ^{40}K, decay series, and cosmic rays. The basic system of gamma ray spectroscopy consists of a NaI(TL) detector, a high voltage power supply, a unlit channel Analyzer (MCA box), and a sensor Cassy. The gamma ray spectra data were analyzed by using Cassy Lab software on a PC[2]. The block diagram of the equipment's setup of the sodium iodide detector is shown in (Figure 1).

Received*: 15-08-2025 -* ***Accepted****: 14-10-2025 -* ***Published****: 20-10-2025*

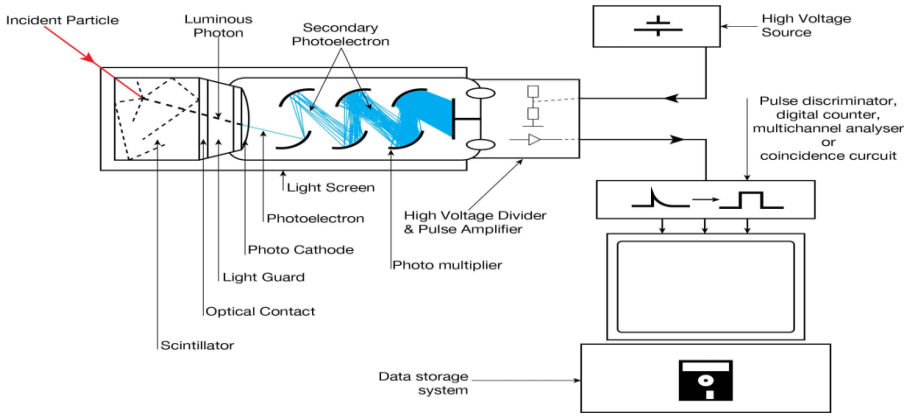

Figure 1. Description of the block diagram of the system

Measurements of radioactivity

Detection of Radioactive Elements

The identification and quantification of natural radionuclides were performed using gamma-ray spectrometry. The measurement system consisted of a NaI(Tl) scintillation detector coupled with a high-voltage power supply and a computer equipped with software for spectral acquisition and analysis. System calibration was executed using certified gamma-ray standard sources. All measurements were conducted at the Physics Department, Faculty of Science, Omar Al-Mukhtar University [4&5].

Estimation of Radium Emanation Rate in the Samples

The radium emanation rate was determined using the "can technique". Initially, 250 g aliquots of the dried and pulverized samples were sealed in 1-liter glass containers for 45 days to ensure equilibrium between radium and its progeny. Following this, LR-115 Type II solid-state nuclear track detectors (SSNTDs) were affixed to the inner upper surface of the containers, which were then resealed for a 90-day exposure period to record alpha particles from radon decay. Post-exposure, the detectors were chemically etched in a 2.5 N NaOH solution maintained at 50°C for 3 hours in a constant-temperature water bath. The resulting track densities (tracks cm^{-2}) were measured using an Olympus optical microscope at 400X magnification, and radium concentrations were calculated accordingly.

Estimation of Uranium in Samples

Uranium concentrations were quantified via the fission track technique, the samples were pulverized and homogenized with a polyvinyl chloride (PVC) binder, and the mixture was hydraulically pressed into thin pellets. These pellets were encapsulated in aluminum foil and placed in close contact with Lexan plastic track detectors alongside certified reference standards (Fischer glass). The assembly was irradiated with a thermal neutron fluence of approximately 1×10^{15} n cm^{-2}. Fission fragments from the $^{235}U(n,f)$ reaction induced latent tracks in the Lexan detectors, which were subsequently revealed by chemical etching in a 6.25 N NaOH solution at 60°C for 50 minutes. The density of the fission tracks was determined using an optical microscope at 400X magnification, and uranium concentrations were calculated by comparing the track densities of the samples to those of the standards.

Calculation of Radiation Hazard Indices

To assess the potential radiological health risks associated with elevated concentrations of naturally occurring radionuclides, several internationally recognized hazard indices were computed from the measured activity concentrations of ^{238}U, ^{232}Th, and ^{40}K.

Radium Equivalent Activity (Raeq)

The Radium Equivalent Activity (Raeq) was calculated to evaluate the gamma radiation hazard from the combined activity of ^{238}U, ^{232}Th, and ^{40}K. This index provides a single weighted value that is commonly used to assess the suitability of materials for construction. The Raeq was calculated using the established mathematical formula from previous studies [4&5].

$Raeq = AU + 1.43ATh + 0.07AK$

Radionuclide Determination via Gamma Spectrometry

Natural radionuclides were identified using sodium iodide (NaI) gamma-ray spectrometry. Specific radiation indices were calculated as follows: Radium Equivalent Activity (Raeq): The activity concentration (Bq/kg) for each radionuclide was determined using the equation:

$A = N / (\varepsilon\gamma * I\gamma * m * t)$ (1)

Where: N = Net sample count (or net area under the photopeak), $\varepsilon\gamma$ = Detector efficiency, $I\gamma$ = Gamma yield, m = Sample mass, t = Counting time.

Received*: 15-08-2025 -* ***Accepted****: 14-10-2025 -* ***Published****: 20-10-2025*

To evaluate radiation exposure, concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were analyzed, and a standard radiation hazard index was established. This index, known as the Radium Equivalent Activity (Raeq), is mathematically defined as [4&5].

$$Raeq = ARa + 1.43ATh + 0.077AK \quad (2)$$

Where: ARa, ATh, AK = Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Radiation Hazard Indices (Hex and Hin)

The detrimental effects of gamma radiation from radionuclides present in the samples were estimated by calculating various hazard indices. While total activity concentration serves as a precise indicator of overall radiation risk, the hazard indices also guide selection of suitable materials for constructing dwellings, bricks, and or materials for human habitation. Two hazard indices were employed: External Hazard Index (Hex) and Internal Hazard Index (Hin). External Hazard Index (Hex): external hazard index (Hex) due to gamma ray emission from samples was calculated to estimate biological risk using the relationship:

$$Hex = ARa/370 + ATh/259 + AK/4810 \leq 1$$

Where: ARa, ATh, and AK are activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg

Internal Hazard Index (Hin)

Internal exposure arises from inhalation of radon gas (^{222}Rn) and its short-lived decay products or ingestion of radionuclides. As radon is carcinogenic and present in all building materials, measurement of radon exposure is termed the Internal Hazard Index, calculated as follows:

$$Hin = ARa/185 + ATh/259 + AK/4810$$

Gamma Level Index (Iγ): The gamma level index (Iγ) is used to assess risk level from ^{238}U, ^{232}Th, and ^{40}K radionuclides. It is calculated using the relationship:

$$I\gamma = ARa/150 + A\ Th/100 + AK/1500$$

Where: ARa, ATh, and AK are activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg.

Alpha Index (Iα)

External radiation, particularly radon and its short-lived decay products, poses a risk to the respiratory system. se emit alpha particles that adhere to airborne aerosols, dust, and or particles. Upon inhalation, radon decay products deposit on cells lining airways, where alpha particles can damage DNA and potentially cause lung cancer. Excess alpha radiation due to radon inhalation is estimated by the alpha index (Iα), defined as follows:

$$I\alpha = ARa/200$$

Recommended upper limit for ^{226}Ra concentration is 200 Bq/kg, corresponding to Iα = 1.

Computer Software

Microsoft Excel and Origin software were utilized for data analysis and calculations in this study.

Results

Radioactive Nuclide results

The amounts of ^{232}Th, ^{238}U, and ^{40}K are not uniform over the planet; there are certain regions where one species is more abundant than the other, and vice versa. As a result, the concentrations of these nuclides in tea samples vary. This work aims to measure the concentration level of natural radionuclides ^{232}Th,^{238}U, and ^{40}K in tea samples collected from the local market in Al-Beida city-Libya. The radioactivity measurement was performed using gamma-ray spectrometry at the Advanced Nuclear Physics Laboratory, Department of Physics, Omar Al-Mukhtar University. The radionuclide contents analysis and the determination of their activity concentration and radiological hazards in samples will be presented in this study.

Calculations of Gamma Spectrometer results

The calculated activity concentrations for selected gamma-ray transitions from the decays of ^{214}Pb, ^{226}Ra, and ^{214}Bi (from the ^{238}U series), ^{228}Ac, ^{212}Pb, ^{212}Bi, and ^{208}Tl (from the ^{232}Th series), and ^{40}K. By picking the largest value of their activities, the specific activity of the samples selected on the Lead isotope (^{214}Pb) at energies (295.21 and 351.92) keV and Bismuth (^{214}Bi) at energies (609.37, 768.4, 1120.3, 1238.1, 1377.7, and 1764.5keV) is comparable to the specific activity of Uranium (^{238}U). While the specific activities of Bismuth (^{212}Bi) at 727.2 keV, Thallium (^{208}Tl) at 583 keV, and Actinium (^{228}Ac) at energies of (92, 209.5, 338.5, 911.1, and 968.9) keV, respectively, are identical to the specific activity of Thorium (^{232}Th) by selecting the greatest value of their activities.

Using their energies, 1460.80 keV. The specific activity concentration of radioactive element ^{40}K was computed at energy 1460.8 keV. To quantify the gamma-emitting radionuclides within the samples, a direct counting geometry was employed. In this approach, the samples were strategically positioned in close proximity to, or directly above, the detector surface to optimize detection efficiency. Each sample underwent a prolonged counting period of 7200 seconds (two hours) to ensure sufficient accumulation of counts for accurate activity determination. The resulting activity concentrations were expressed in Becquerels per kilogram (Bq kg^{-1}), which signifies the number of nuclear disintegrations (decays) occurring per unit mass

Received: *15-08-2025* - ***Accepted***: *14-10-2025* - ***Published***: *20-10-2025*

of the sample per second. This unit provides a standardized metric for comparing the radioactivity levels within different samples [4].

Activity Concentration of Radionuclides

The activity concentrations of radionuclides were determined using a gamma-ray spectrometer (NaI (Tl) detector), and the gamma dose rate from these radionuclides was calculated in this work. (Table 2) summarizes the ^{226}Ra,^{238}U,^{232}Th, and ^{40}K average activity concentrations for each of the examined samples. The obtained values differed for tea samples 7.07 to 54.74, 2.14 to 31.42, 12.74 to 50.95, 63.48 to 156.12 Bqkg^{-1}for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, respectively, with average values of 32.82, 21.67, 22.32, and 110.66 Bq kg^{-1}, respectively, as shown in (Figure 1). The results of the specific activity concentrations of radionuclides ^{226}Ra and ^{40}K for all tea samples are lower than the world recommended values 50 and 15 Bq kg^{-1}, respectively. But the higher value of activity concentration of ^{238}U and ^{232}Th for most samples under investigation was lower than the international recommended value of 20 and 420 Bq kg^{-1} (UNSCEAR, 2010).

Table 2. Specific activity concentrations of natural radionuclides in tea samples

Sample code	^{226}Ra	U^{238}	^{232}Th	^{40}K
T_1	43.26	23.75	12.97	156.12
T_2	28.26	25.27	42.02	104.65
T_3	43.26	28.62	40.18	140.68
T_4	32.67	11.88	46.30	109.80
T_5	7.07	2.14	27.58	87.50
T_6	42.38	24.82	32.13	133.82
T_7	37.97	20.15	12.74	106.37
T_8	34.43	31.02	45.14	118.38
T_9	26.49	15.72	34.46	85.78
T_{10}	28.26	31.42	50.95	63.48
T_{11}	54.74	24.47	28.33	123.52
T_{12}	15.02	13.54	13.87	97.79
Average	32.82	21.67	22.32	110.66
P.L (UNSCEAR, 2010)	50	20	15	420

Radiological Hazard Indexes

These radiological parameters are calculated by measuring the radionuclide activity concentrations ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, but also for estimating radiation doses and assessing biological effects on the human body. Several relevant quantities were used in the current study, including absorbed dose rate (D_R). Annual effective dose equivalents for Internal and External (E_{in} and E_{out}), cancer risk factors (*ELCR*), and annual Gonadal Equivalent Dose (AGED) for terrestrial gamma radiation. The values of these calculated radiological hazard parameters are given in (Table 3).

Table 3. Absorbed dose rate, Internal and External annual effective dose, excise life cancer risk, and annual gondola effective dose of tea samples

Sample Code	DR (nGy.h-1)	Ein mSvy-1))	Eout mSvy-1))	ELCR × 10-3	AGED (mSvy-1)
T1	34.37704	0.16864	0.042146	0.139083	0.23691
T2	42.8315	0.210114	0.052511	0.173288	0.295827
T3	50.1634	0.25	0.06	0.20	0.35
T4	47.67034	0.233852	0.058444	0.192865	0.328962
T5	23.59966	0.12	0.028933	0.09548	0.164606
T6	44.60652	0.218822	0.054688	0.180469	0.307277
T7	29.70464	0.145719	0.036418	0.120179	0.203981
T8	48.14318	0.236171	0.059024	0.194778	0.332245
T9	36.65498	0.179815	0.044939	0.148299	0.252832
T10	46.49608	0.228091	0.057004	0.188114	0.320227
T11	47.58904	0.233453	0.058344	0.192536	0.326351
T12	19.4239	0.095286	0.02	0.08	0.135094
Average	39.27	0.19	0.05	0.16	0.27
P.L (UNSCEAR, 2010)	84	1	0.07	0.29	0.3

Received*: 15-08-2025 -* ***Accepted****: 14-10-2025 -* ***Published****: 20-10-2025*

The Absorbed Dose Rate (DR)

The calculated absorbed dose rate DR for all samples is shown in (Table 3), The estimated absorbed dose rate values for the examined tea samples ranged from 19.42 to 50.16 nGyh-1, but the average absorbed dose rate value of the tea samples was 39.27 nGyh-1. (Figure 3) shows these results. The values of the absorbed dose rate for all studied samples were lower than the world recommended value of 84 nGyh-1 (UNSCEAR, 2010).

The Internal Annual Effective Dose (E_{in})

As shown in (Table 3). The values obtained for the Internal annual effective dose E_{in} of Egyptian granite samples vary from 0.12 to 0.25 mSvy^{-1}, with an average value of 0.19 mSvy^{-1}, as shown in (Figure 4). These average values for all measured samples were less than the 1 mSvy^{-1} limit. The annual effective dose calculated for all Internal samples analyzed in this study falls below the globally accepted safety threshold for public exposure to radiation. Consequently, these samples do not pose any health risks to passengers, aligning with the conclusions of other investigations.

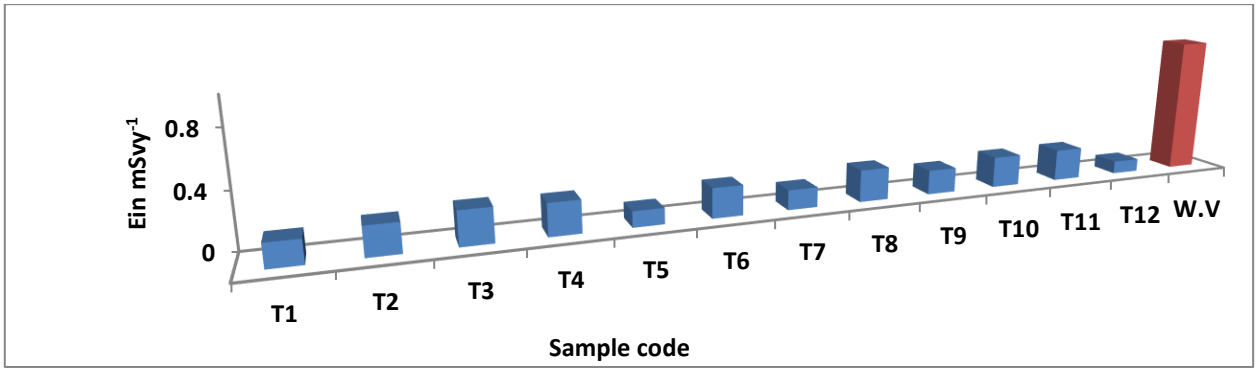

Figure 2. Internal annual effective of tea samples

The External Annual Effective Dose (E_{out})

From (Table 3), the values obtained for the annual external effective dose E_{out} varied from 0.02 to 0.06 mSvy^{-1}, with an average value of 0.05 mSvy^{-1}. The values obtained for the External annual effective dose for all tea samples are lower than the recommended values of 0.07 mSvy^{-1}. As shown in (Figure 3).

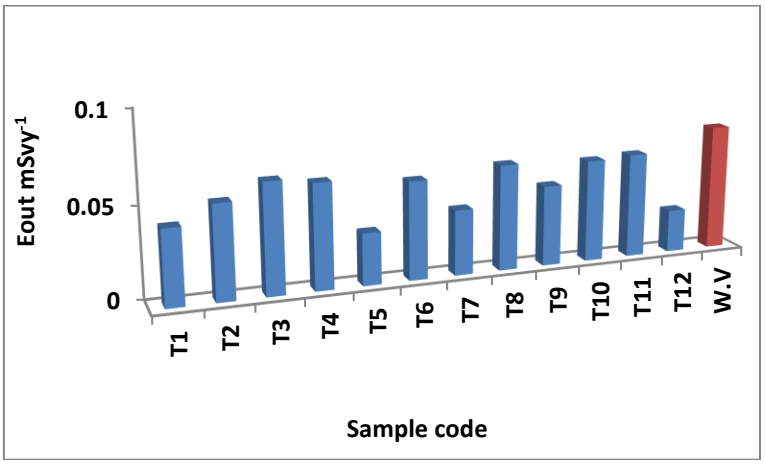

Figure 3. External annual effective of tea samples

Excess Lifetime Cancer Risk (ELCR)

The ELCR was calculated, where the equation is based on life expectancy (70 years) and (*RF*) is the risk factor (0.05 Sv^{-1}). (Table 3) shows the cancer risk factor for all samples, and the recorded values for the risk of cancer over the extra life span for Egyptian granite samples range from 0.08 to 0.20, with an average value of 0.16. The *ELCR* values for all tea samples are lower than the recommended world value 0.29 × 10^{-3} (UNSCEAR, 2010). As shown in (Figure 4).

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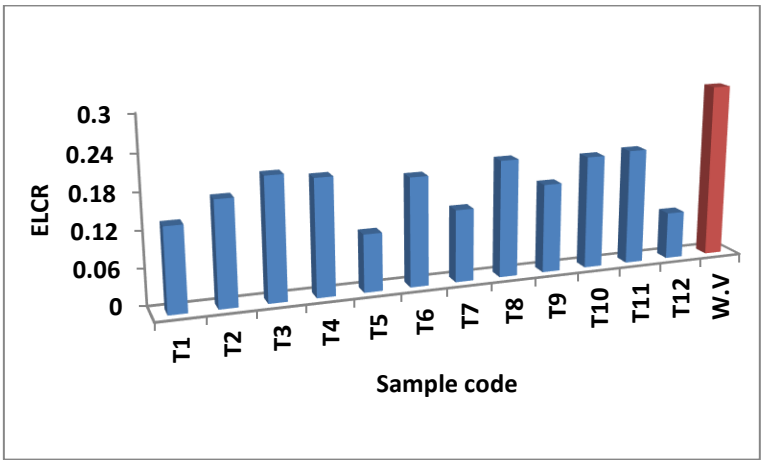

Figure 4. Excess lifetime cancer risk of tea samples

Annual Gonadal Equivalent Dose (AGED)

AGED as shown in Table 3). The obtained values of (AGED) for tea samples ranged from 0.13 mSv.y^{-1} to 0.35 mSv.y$^{-1,}$ with an average of 0.27 mSv.y^{-1}. The results showed that the average values of annual gonadal equivalent dose for all samples were lower than the threshold value of 0.3 mSv.y^{-1}. As shown in (Figure 5).

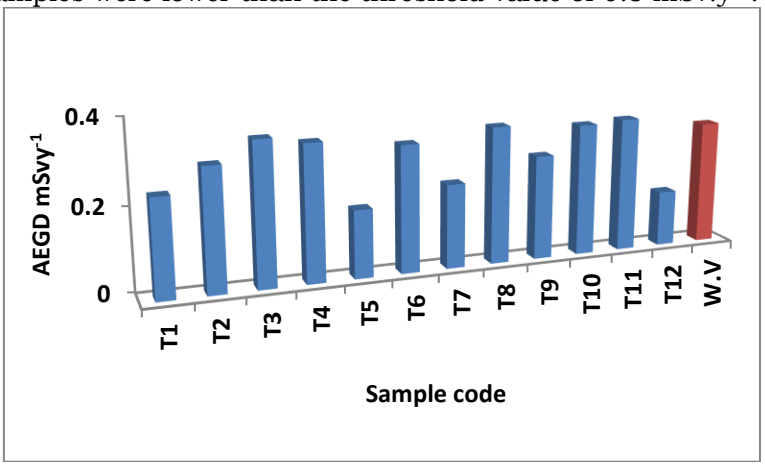

Figure 5. Annual Gonadal Equivalent Dose of tea samples

All values were below global safety thresholds (UNSCEAR, 2010).The annual effective dose was below 1 mSv, the recommended safety limit. Lifetime Excess Cancer Risk (ELCR) ranged from 0.08 to 0.20 ×10^{-3}, lower than the global reference value of 0.29 ×10^{-3}.

Discussion

The findings suggest that the analyzed tea samples pose no immediate radiological hazard to consumers. However, the variation in radioactive concentrations reflects possible influences from soil composition, irrigation water, and agricultural practices. Periodic monitoring of radioactive levels in agricultural products is recommended, especially for widely consumed items. The equivalent concentrations of radium-226 measured in this study were below permissible limits, with the highest value reaching 17.56 Bq/(kg or L). This is further supported by the assessment of both internal and external risk factors. The study found an average external risk factor (Hex) of 0.02 Bq/kg and an average internal risk factor (Hin) of 0.06 Bq/kg. The annual effective dose for all samples was below the 1 mSv standard set by the International Commission on Radiological Protection, falling within safe limits. These radioactivity levels align with the recommendations of international organizations and bodies, which were the primary guidelines used in this study. Some studies stated that the main sources of radioactive materials mainly come from natural sources, especially from the natural elements present in many tea samples. The differences between the detected radioactive in this study are attributed to the different tea sources from many countries, water irrigation, and pesticides uses [4-6].

Conclusion

This study identified the levels of radioactivity in tea samples collected from local Libyan markets. Notably, uranium exhibited the highest concentration among the detected radioactive elements. The study concluded that the presence of radioactive elements in some studied sapless mainly due to the differences in the sources of tea samples.

Received*: 15-08-2025 -* ***Accepted****: 14-10-2025 -* ***Published****: 20-10-2025*

Acknowledgement
Special thanks to the physics department, Faculty of Science, Omar Al-Mukhtar University for their help during the analysis of radioactive elements.

Conflict
The authors state that no conflict of the results recorded in this study with the other ones.

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