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Original article

Assessment of Natural Radioactive Elements and Health Risk Values in Imported Cocoa Samples at Some Libyan Markets

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ABSTRACT

This study investigated the presence of natural radioactive isotopes in imported cocoa samples collected from Derna, Libya, using gamma-ray spectrometry with a Sodium Iodide (NaI) detector. The analysis focused on the activity concentrations of radionuclides such as ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K. The average activity concentrations were 46.96 Bq/kg (^{226}Ra), 49.77 Bq/kg (^{238}U), 53.57 Bq/kg (^{232}Th), and 123.75 Bq/kg (^{40}K). Values for ^{226}Ra, ^{238}U, and ^{232}Th exceeded the global recommended limits, while ^{40}K remained below the permissible limit. The absorbed dose rate (DR) ranged from 42.42 to 80.50 nGy/h, with an average of 58.27 nGy/h, lower than the global reference of 84 nGy/h. Internal and External annual effective doses were 0.29 mSv/y and 0.07 mSv/y, respectively, both within safe limits. The Excess Lifetime Cancer Risk (ELCR) averaged 0.26×10^{-3}, slightly higher than the global recommended threshold (0.29×10^{-3}). The Annual Gonadal Equivalent Dose (AGED) averaged 0.41 mSv/y, which is above the safety limit of 0.3 mSv/y. Hazard indices (Radium Equivalent, Gamma Index, Alpha Index, Hin, Hex) were all below international safety limits, indicating no immediate health risk.

Introduction

Natural radioactive isotopes, which are a component of naturally occurring radioactive materials (NORM), and technologically enhanced naturally occurring radioactive materials (TENORM) are the two primary categories of radioactive sources that are found in our bodies and the environment. Furthermore, industrially generated radioactive materials are man-made radioactive materials. Those with incredibly lengthy half-lives, sometimes longer than the age of the Earth itself, are among the main radioactive isotopes. These isotopes, uranium (^{235}U-^{238}U) and thorium (^{232}Th), are two prominent examples that are essential to the radioactive activity of the Earth. Secondary radioactivity. These fundamental isotopes decay to produce elements, which are naturally present on Earth. Radium (^{224}Ra, ^{226}Ra, and ^{228}Ra), radon (^{220}Rn and ^{222}Rn), lead (^{210}Pb), polonium (^{210}Po), and thorium (^{228}Th, ^{230}Th, and ^{232}Th) are some of the significant radioactive isotopes in these decay chains. Furthermore, a frequent radioactive isotope that greatly increases the yearly radiation dosage that people receive is potassium-40 (^{40}K) [1]. Radioactive materials that are found naturally in the environment, such as anthropogenic, biological, and primordial radioactive isotopes, are referred to as naturally occurring radioactive materials (NORM) [2].

Environmental and geographic factors affect natural radioactive activity and external radiation exposure, particularly gamma rays, which manifest in varying intensities. Radiation activity levels can be used to forecast changes in natural radioactive activity brought on by nuclear accidents or other industrial and human activities, monitor radioactive pollution, and evaluate radiation dose rates [2]. Evaluating human radiation exposure has become essential due to the growing necessity for radiation usage and its uses in many different industries. Additionally, there are cosmic radioactive isotopes such as carbon-14(^{14}C), tritium (^{3}H), and beryllium-7(^{7}Be) that can contribute to the comprehension of naturally occurring radioactive activity. Radioactive elements and heavy metals are naturally occurring chemicals that are present in the water and the atmosphere in different concentrations. Because of their environmental prevalence, they may also show up as residues in food. When these heavy metals build up in the body, they can cause harm to organs or disruptions in essential processes [3].

No studies were carried out on different samples by different methods, such as sodium iodide detectors, gamma rays, and germanium detectors [4-6]. The studies of the chemical compounds that have high risks on human health were established on heavy metals [7-35], Hydrocarbon compounds [36-42], plants and vegetables [43-75]; on the other hand, some studies were carried out on some treatments for the environmental danger's materials [76-87]. This study aims to determine the radioactive nuclides in imported Cocoa samples and their hazard indexes.

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Methods
Sampling
In this study, ten imported cocoa samples were collected from local markets in Derna city, Libya, 2024. The types of samples were given in (Table 1).

Table 1. The studied cocoa samples

Sample. No	Name sample
1	Nesquik
2	Said
3	Ovaltine
4	Darlet
5	Corona
6	ALwdaq
7	CaCao Drink
8	Katsan Ovalite Cocoa powder
9	Raw cocoa
10	Dutch cocoa

Radioactive nuclides analysis
Samples preparation and measurements
The samples were dried in an oven set at 500 ^{0}C for two hours. After allowing the samples to cool, they were weighed and safely placed in plastic bottles. Following transportation and sealing, these samples were kept for around 30 days to create secular equilibrium between the ^{232}Th and ^{238}U series and their corresponding daughter radionuclides [5].

Gamma Spectroscopy
Gamma-ray spectroscopy is used to identify the various radioactive isotopes in a sample. This technique measures the energy of gamma rays that reach the sample using a particular detector. By comparing the measured energy with the known energies of gamma rays emitted by radioactive isotopes, the source of the radiation may be located. This method stands out for its numerous applications, particularly in fields that need quick analysis without causing sample degradation. As stated by [6]. An alternative definition of Gamma-ray spectroscopy is an analytical technique based on the analysis of the energy spectrum of gamma rays emitted by radioactive materials.
Because detecting and analyzing the energy distribution of gamma rays allows for the exact identification of radioactive isotope types and the calculation of their numbers, these rays are a Distinctive fingerprint of every radioactive material. To capture gamma ray energy and produce the proper spectrum, this technology relies on the employment of specialized reagents like semiconductors (such as high-purity germanium detectors) or phosphorescent compounds (such as sodium iodide detectors). Nuclear physics, environmental monitoring. Nuclear medicine and many other scientific and applied domains depend on gamma-ray spectroscopy. Internal security. In the field of Environmental Sciences, this technique plays a pivotal role in assessing the levels of radioactive contamination, both resulting from natural radioactive substances (NORMs) and artificial radioisotopes caused by nuclear activities [5-6].

The Sodium Iodide Detector (NaI)
Sodium iodide is a radioactive inorganic material. It is a scintillation detector. Standard compounds called scintillants, which release light when they meet particles, are used to detect radiation [5]. A single crystal of pure sodium iodide connected to the photocathode of an optical photomultiplier tube makes up the construction of the thallium-doped sodium iodide detector. The crystal is excited when gamma rays contact the sodium iodide and ionize it as they reach the detector. It breaks down by releasing photons of visible light. We call this emission scintillation. This detector is hence referred to as a scintillation detector. To move the visible photons' wavelength into the photocathode's sensitive region, the crystal must be doped with thallium.
The scintillation crystal is encased in a thin aluminum sheath with a glass window at the interface with the photocathode to prevent moisture absorption of the hygroscopic NaI [6]. This detector interacts with gamma rays at room temperature and produces a small amount of scintillation. NaI becomes more efficient at generating light photons after interacting with gamma rays by adding a small amount of doped thulium to the pure NaI detector (0.1-0.4%). This interaction ionizes or excites NaI (Ti) molecules through

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the emission of light photons. The energy drops from the high state to the ground state again, and approximately 20-30 photons of 1 keV energy are generated.

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 [5,6].

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 construction materials. The Raeq was calculated using the established mathematical formula from previous studies [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.
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 [5&6].
Raeq = ARa + 1.43ATh + 0.077AK (2)
Where: ARa, ATh, AK = Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

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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 the 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 (Ia)

External radiation, particularly radon and its short-lived decay products, poses a risk to the respiratory system. They 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 results recorded different levels of ^{232}Th, ^{238}U, and ^{40}K, and radionuclides. Consequently, the radionuclides' concentrations in cocoa samples vary. To determine the concentration of natural radionuclides ^{232}Th, ^{238}U, and ^{40}K in cocoa samples. (Table 2) shows the contents of detected radioactive isotopes of ^{226}Ra,^{238}U,^{232}Th, and ^{40}K of examined samples, different obtained values for cocoa samples 28.25 to 61.43, 35.68 to 65.94, 42.43 to 81.90, 31.57 to 262.14 Bqkg^{-1} for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K, respectively, with average values of 46.96, 49.77, 53.57, 123.75 Bqkg^{-1}, respectively.
The results of specific activity of some cocoa samples have radioactive quantities of ^{226}Ra, ^{238}U, and ^{232}Th that are greater than globally advised levels of 33, 32, and 40 Bq kg^{-1}, respectively. However, samples under evaluation had a higher activity content of 40K, which was less than the 400 Bq kg^{-1} worldwide recommended threshold in previous studies [88]. Also, the results showed that some samples did not contain radioactive elements according to the method used in this study.

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

Sample code	^{226}Ra	^{238}U	^{232}Th	^{40}K
C1	54.37	65.94	44.13	262.14
C2	41.66	44.22	51.20	225.09
C3	49.43	54.15	57.93	71.37
C4	46.60	41.35	43.82	31.57
C5	61.43	57.28	81.90	63.13
C6	28.25	35.68	42.43	89.21
Average	46.96	49.77	53.57	123.75
P.L	33	32	40	400

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Radioactivity values
By computing radiation parameters, the concentration of radionuclide activity is determined for ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K. To estimate radiation doses and assess biological effects on the human body, several pertinent quantities were used, such as absorbed dose rate (D_R), annual effective dose equivalents for Internal and external radiation (E_{in} and E_{out}), cancer risk factors (ELCR), and annual gonadal equivalent dose (AGED) for terrestrial gamma radiation. (Table 3) displays values of computed radiation hazard parameters.

Table 3. Absorbed dose rate, Internal and external annual effective dose, excise life cancer risk, and annual gondola effective dose of the cocoa sample.

Sample code	DR ($nGy.h^{-1}$)	Ein ($mSvy^{-1}$)	Eout ($mSvy^{-1}$)	ELCR $\times 10^{-3}$	AGED ($mSvy^{-1}$)
C1	62.78	0.31	0.08	0.25	0.47
C2	53.75	0.26	0.07	0.22	0.37
C3	60.82	0.30	0.07	0.25	0.42
C4	49.32	0.24	0.06	0.20	0.34
C5	80.50	0.39	0.10	0.32	0.55
C6	42.42	0.21	0.05	0.317	0.29
Average	58.27	0.29	0.07	0.26	0.41
P.L (UNSCEAR, 2010)	84	1	0.07	0.29	0.3

Absorbed Dose Rate (DR)
Estimated absorbed dose rate values for examined cocoa samples ranged from 42.42 to 80.50 $nGyh^{-1,}$ but the average absorbed dose rate value of cocoa samples was 58.27$nGyh^{-1}$. Values of absorbed dose rate for all studied samples were lower than the world-recommended value of 84$nGyh^{-1.}$ The calculated absorbed dose rate DR for all samples is shown in (Table 3).

Internal Annual Effective Dose (Ein)
values obtained for Internal annual effective dose Ein of Egyptian granite samples vary from 0.21 to 0.39 $mSvy^{-1}$, with an average value of 0.29 $mSvy^{-1}$ as shown in (Table 3). The average values for all measured samples were less than 1 $mSvy^{-1,}$ a limit previously studied [90]. Th annual effective dose calculated for all Internal samples analyzed in this study falls below the globally accepted safety threshold for public radiation exposure. Consequently, these samples do not pose any health risks to passengers, aligning with the conclusions of our investigations.

External Annual Effective Dose (Eout)
Yearly external effective dose Eout values derived from Table 3) ranged from 0.05 to 0.08 $mSvy^{-1}$, with an average of 0.07 $mSvy^{-1}$. All cocoa samples' external yearly effective dose levels were found to be equal to the suggested values of 0.07 $mSvy^{-1}$.

Excess Lifetime Cancer Risk (ELCR)
ELCR was determined using an algorithm based on life expectancy (70 years), with a risk factor (RF) equal to 0.05 Sv^{-1}. With an average value of 0.26, recorded values for cancer risk factor during additional life span for Egyptian granite samples vary from 0.20 Sv^{-1} to 0.32 Sv^{-1}, as shown in (Table 3). All cocoa samples had ELCR levels more than 0.29*10-3, the global recommended threshold 89].

Annual Gonadal Equivalent Dose (AGED)
According to (Table 3), the average annual gonadal equivalent dose for cocoa samples was 0.41 $mSvy^{-1}$, with values ranging from 0.29 $mSvy^{-1}$ to 0.55 $mSvy^{-1}$. According to findings, average yearly gonadal equivalent dose levels for every sample were higher than the 0.3 $mSvy^{-1}$ threshold value.

Radioactive elements
Natural radioactive nuclei were determined by the Sodium Iodide (NaI) detector method. Values of natural radioactive nuclei are shown in (Table 3). Also, some radioactivity indices were calculated according to the following methods: Radium Equivalent Dose (Ra_{eq}), Radiation Level Index ($I\gamma$), Alpha index ($I\alpha$), Internal Hazard Index (H_{in}), External Hazard Index (H_{ex}), values of which are shown in (Table 4):

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Table 4. Hazard indices of the cocoa sample

Samples code	Hazard indexes				
	Raeq	I□	Iα	Hin	Hex
C1	137.66	0.49	0.27	0.52	0.37
C2	121.44	0.42	0.21	0.44	0.33
C3	137.76	0.48	0.25	0.51	0.37
C4	111.70	0.38	0.23	0.43	0.30
C5	183.41	0.63	0.31	0.66	0.49
C6	95.79	0.34	0.33	0.33	0.26
Average	131.29	0.45	0.26	0.48	0.35
P.L	370	1	1	1	1

Radium Equivalent Dose (Raeq)

Estimated Radium Equivalent Dose values for the examined cocoa samples ranged from 95.79 to 183.41 $Bq.Kg^{-1,}$ but the average radium equivalent dose value of cocoa samples was 131.29 $Bq.Kg^{-1}$. The values of radium Equivalent dose for all studied samples were lower than the world-recommended value of 370 $Bq.Kg^{-1}$, calculated Radium Equivalent Dose (Ra_{eq}) for all samples is shown in (Table 5).

Gamma Radiation Level Index (Iγ)

According to (Table 4), the average value of the samples' Gamma Radiation Level Index (I) is 0.45 Bq.kg-1, with values ranging from 0.34 to 0.63 Bq.kg-1. The average readings for all analyzed samples were less than 1 Bq.kg-1limit. All of the samples examined in this investigation had Gamma Radiation Level Index values that are below the generally recognized safety threshold for radiation exposure to the general population. As a result, no health hazards were associated with the studied samples.

Alpha index

Alpha index values derived from (Table 4) ranged from 0.21 to 0.33 $Bq.kg^{-1}$, with an average of 0.26 $Bq.kg^{-1}$. All cocoa samples Alpha index values fall short of the suggested threshold of 1 $Bq.kg^{-1.}$

Internal Hazard Index (Hin)

As shown in (Table 4), Internal Hazard Index (H_{in}) values for cocoa samples ranged from 0.33 to 0.66 $Bq.kg^{-1}$, with an average of 0.48 $Bq.kg^{-1}$. Results showed that the average values, internal hazard index for each sample, were more than the 1 $Bq.kg^{-1}$ threshold value.

External Hazard Index (Hex)

External Hazard Index (Hex) values of samples range from 0.26 to 0.49 $Bq.kg^{-1}$, with an average of 0.35 $Bq.kg^{-1}$, as indicated in (Table 4). Average levels for all analyzed samples were below the 1 $Bq.kg^{-1}$ limit, and UNSCEAR (2010). All of the samples examined in this investigation had External Hazard Index values below the generally recognized safety level for radiation exposure to the general population. As a result, these samples do not present any health hazards to humans.

Discussion

Although levels of radioactive isotopes uranium, potassium, and thallium in examined cocoa samples varied from sample to sample, they were all over internationally advised limits, suggesting long-term health hazards. High radiation exposure can cause major health issues, such as an elevated risk of cancer. Findings of this investigation align with those of the study. This research focused on cocoa. Radioactivity levels in cocoa were found to be high. Hazard Index (HI) and Target Hazard Quotient (THQ) were also used to calculate health hazards. The Hazard Index 1.49 exceeded the internationally advised safe threshold. Suggesting that ingesting these goods in high numbers may pose a health risk. The findings align with research conducted by previous studies [88-91], which demonstrated that consistent cocoa eating, chocolate that has been contaminated, can have major negative health impacts, such as an elevated risk of cancer and renal disease. It is advised to use aqueous extracts sparingly to get vital nutrients and safeguard consumers.

To guarantee that daily consumption restrictions are not exceeded and to ensure the public's health from potential hazards related to heavy metals and radiation, it is crucial to monitor and examine cocoa samples from Libyan markets. Different cocoa brands and geographical areas have been found to contain

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varying levels of heavy metals, underscoring the necessity of routine testing and food product quality management [91-93].

Differences in fundamental physical and chemical properties of various agricultural regions can account for notable diversity in metal levels throughout these locations. The type, water quality, mineral content, climate, pesticides, and processes utilized to change the product into its consumable form are a few examples of se variables. Accumulation of metal in plant components is influenced considerably by each of these factors [94]. Thus, radioactive elements in different chocolate brands were significantly influenced by their places of origin. It is crucial to remember that the water solubility of various metals varies. Previous studies have reported similar variations in radioactive elements in different chocolate brands. To ensure the safety of food products and consumers, research and quality control in this area are suggested [94].

Conclusion

The study confirms that cocoa samples from Libyan markets contain natural radionuclides above recommended levels for ^{226}Ra, ^{238}U, and ^{232}Th, but overall radiation dose parameters (DR, Hex, Hin, Iγ) remain within internationally accepted safety standards. However, slightly elevated ELCR and AGED values suggest that long-term exposure through consumption could pose potential health risks.

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Conflict

No conflict of the results recorded in this study with the other studies or authors.

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