**Measurement of Natural Radionuclides Concentration in Libyan Food Spices Samples**

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**ABSTRACT:**

Awareness of radioactivity concentrations in human diet are of high concern for the estimation of radiological hazards to human health. However, not many studies of radioactivity in food have been conducted in Libya. In this study the activity concentration of naturally occurring radionuclide levels in a set of 30 samples of food spices was determined. Samples were collected from one of the biggest local markets in Tripoli, Libya; Souliman Khater. All samples were prepared and analyzed at the department of Nuclear Engineering, University of Tripoli to establish activity concentrations associated with the $^{238}$U and $^{232}$Th natural decay chains and the $^{40}$K.

The activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K based on their weighted average values across samples were (1.92±0.72), (2.70±0.61) and (529.50±0.04) Bq kg$^{-1}$, respectively.

Achieved results showed that the activity concentration was within the acceptable level for $^{238}$U, $^{232}$Th while, the $^{40}$K level was high in samples of Cardamom and Hot red pepper imported from India, Nalta jute from Tunis, Turmeric, Mixed Turmeric from Libya, and they were significantly higher than

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as reported by the UNSCEAR (2000).

The highest activity concentration of $^{238}U$, $^{232}Th$ was recorded for Cinnamon from India, which were 16.32±1.64 and 16.30±1.65 Bq kg$^{-1}$ respectively. The range of $^{40}K$ was found to be between 66.41±5.11 Bq kg$^{-1}$ for White pepper and 1284.33±88.32 Bq kg$^{-1}$for Cardamom. The annual effective dose equivalent in μSv y$^{-1}$ ranged between 9.52 and 72.57 μSv y$^{-1}$while the world average annual effective dose equivalent (AEDE) from outdoor or indoor terrestrial gamma radiation was 290 μSv y$^{-1}$.

The results of this study were slightly higher than those achieved in many other studies conducted in Egypt, Iraq, and other countries. High concentration of potassium was recorded in many samples. Therefore, it is important to concern about the daily potassium intake by looking at the combination of food and potassium concentration in it, such as by increasing consumption of food spices.

**KEYWORDS:** Natural occurring radioactive, radioactivity, Gamma spectrometry, annual effective dose.

**INTRODUCTION**

People are exposing to the natural source of ionizing radiation continuously (Awudu et al., 2012, pp. 635–6410). Naturally Occurring Radioactive Materials (NORMs) produced from terrestrial, and artificial sources utilised in various applications in industry, agriculture, research and medicine (WHO and FAO, 2011, pp. 1–5). The natural radioactivity can be detected in food and water, the NORM concentration varies depending on several factors such as regional geology, climate, and agricultural methods. The hazardous from the radionuclides on human health depends on their type and the exposing time. In addition, individuals' radiation dose varies depending on the location as the radioactivity release following the radiological emergency in places close to the nuclear power plant may become more contaminated.

Phosphate fertilization leads to increase the uranium series radionuclides in food crops, which may be subjected to the contamination of agricultural land, air, and water supplies during cultivation. In addition, phosphates may elevate the concern regarding the radioactivity levels. (Ugolini, E. et al. 2020).

The concentration of radionuclides in foods depends on several factors; the food type and the geographic region of produced food. Generally, potassium-40 ($^{40}K$), radium-226 ($^{226}Ra$) and uranium-238 ($^{238}U$) and their associated progeny are the common natural radionuclides in food (WHO and FAO, 2011, pp. 1–5). About one- eighth of the mean annual effective dose is due to natural sources results from foodstuff consumption(Awudu et al., 2012, pp. 635–6410). Potassium-40 accounts for about 60% of the total annual effective dose due to ingestion.

Due to the health concerns associated with the indoor radiation exposure, many international and governmental bodies such as the international commission on Radiological Protection (ICRP), the World Health Organization (WHO), and others, have taken on powerful measures aimed at control such exposures (Amin and Ahmed 2013, pp. 350–354). Cancer is the main health concern for individuals in the long term due to high radiation exposure. IAEA estimates that on average, people radiation exposure due to all natural sources about 2.4 mSv a year. This can vary, based on the geographical regions. In addition, it was reported that about 0.7 mSv per year, occur from cosmic and terrestrial gamma radiation. The fraction of the background dose resulting from ingestion of NORM in food is about 0.25-0.4 mSv per year.

Consumption of contaminated food increases the internal exposure of people, thereby increasing the health risks associated with the high radiation exposure (WHO and FAO, 2011, pp. 1–5).

Food spices come from the bark, roots, seeds, fruit of plants and trees, can be contaminated either through deposition in the atmosphere or transferred by water through the soil (Al-Mashhadani 2020, pp. 0–7& Ononugbo, et al. 2019, pp. 1–9). The natural radionuclides $^{226}$Ra, $^{228}$Ra, $^{228}$Th and $^{40}$K were measured in the foodstuffs using gamma ray spectrometry. Findings showed that the average activity concentration of $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in the natural spices were $0.8 \pm 0.2$ Bq kg$^{-1}$, $0.98 \pm 0.4$ Bq kg$^{-1}$, $2.19 \pm 0.9$ Bq kg$^{-1}$ and $0.06 \pm 0.01$ Bq kg$^{-1}$ respectively ( Awudu et al. 2012, pp 635–641). The results indicated insignificant radiological health hazard to the public due to the consumption of spices via foods comparing to the UNSEAR 2000 report (U. Nations, S. Committee, A. Radiation, G. Assembly, and S. Annexes,2008).

The activity concentration of radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K was estimated using gamma ray spectrometry in several spices consumed in Iraqi kitchen (Al-Mashhadani,et al.2020, pp. 0–7). The study showed that the total annual effective dose received from all radionuclides was below the average radiation dose of 290 μSv y$^{-1}$ received worldwide due to ingestion of natural radionuclide in food spices.

Rafat, et al., in another study measured the concentration of radioactivity in Egyptian food spice. Their findings showed that the activity of $^{226}$Ra, $^{232}$Th were, 40.8, 18.7 and 578.8 Bq kg$^{-1}$ respectively (Amin and Ahmed 2013, pp. 350–354). There was no significant health hazard and Egyptian spices were reported to be radiologically safe, according to the international standards.

Food spices commonly used in Iraqi cuisine were studied(Al–Mashhadani,et al.2020, pp. 0–7). The specific activity of the $^{40}K$, $^{238}U$, $^{232}Th$ and $^{137}Cs$ chains was identified and the doses taken by food consumption were also evaluated. There average activity were, 72.00 Bq kg$^{-1}$, 106.576Bq kg$^{-1}$, 148.74 and 191.88 Bq kg$^{-1}$ respectively. The highest concentration of activity of $^{40}K$, $^{238}U$, $^{232}Th$ and $^{137}Cs$ was recorded in latency (401.7Bq kg$^{-1}$) for $^{40}K$, bay leaf (260.67 Bq kg$^{-1}$) for $^{238}U$, Spice Brianna (733.7 Bq kg$^{-1}$) for $^{232}Th$, and chamomile (833.4Bq kg$^{-1}$) for 137Cs. The maximum value of the total annual effective dose received due to the consumption of natural spices by the population was 169.83 μSv y$^{-1}$.

In Libya, only few studies have been performed to determine the radionuclides concentrations in spices and the dose assessment from consumption of these spices, and there was no reported data provided for these measurements.

## Materials and Methods

### *Sample collection and preparation*

A total of thirty natural food spices samples were purchased from the largest local market (Suleiman Khater) in Tripoli. Samples of about 200 grams were packed in plastic bags labelled and transferred to the laboratory for the purpose analysis. The samples were then ground into fine powder and weighed, dried in oven at 100°C and sieved through a 0.1–mm mesh sieve to remove the impurities and to get a uniform particle size. These homogenized samples were kept in cylindrical plastic containers of diameter 71 mm, 47 mm height and thickness of 1.1 mm, then were well–sealed with adhesive tape. Each sample was weighted and labelled according to its symbol, weight, and the date of storage. The container dimensions were chosen to match the reference source geometry (Figure 1). Samples then stored for about one month at the Department of Nuclear Engineering, the University of Tripoli to reach the state of the equilibrium (Garcêz, et al. 2019, pp. 120–126). Table 1 lists the collected samples.

Figure 1 :Samples preparation

Table 1: Food spices samples

| sample code | Sample name | country of origin |
|---|---|---|
| Sp-1 | Rosemary | Libya |
| Sp -2 | Spice | Tunis |
| Sp -3 | Garlic | India |
| Sp -4 | Nutmeg | India |
| Sp-5 | Cardamom | India |
| Sp-6 | Rice mix | India |
| Sp-7 | White pepper | India |
| Sp-8 | Hot red pepper | India |
| Sp-9 | Paprika | Spain |

| | | |
|---|---|---|
| Sp-10 | Cinnamon | India |
| Sp-11 | Clove | Iran |
| Sp-12 | Curry | India |
| Sp-13 | Kerwia | India |
| Sp-14 | Smoked crushed | Tunis |
| Sp-15 | Turmeric | India |
| Sp-16 | Clove | Iran |
| Sp-17 | Cumin | Libya |
| Sp-18 | Nalta jute | Tunis |
| Sp-19 | Meat sticks | Libya |
| Sp-20 | Medium red | Libya |
| Sp-21 | Onion spices | India |
| Sp-22 | Coriander | India |
| Sp-23 | Thyme | Tunis |
| Sp-24 | Onion | India |
| Sp-25 | Ginger | India |
| Sp-26 | Sumac | India |
| Sp-27 | Black pepper | India |
| Sp-28 | Turmeric | Libya |
| Sp-29 | Mixed turmeric | Libya |
| Sp-30 | Chicken | Tunis |

**Gamma-ray spectrometry:**

The samples were counted using a gamma-ray spectrometer housed in one of the nuclear radiation laboratories of the University of Tripoli at the Nuclear Engineering Department. The high-resolution hyper-pure germanium detector (HPGe), low background counting system by ORTEC was utilized in this work. A lead castle of approximately 10 cm thick with a copper lining on the inside to absorb lead X-rays, shields the detector. The analysis was conducted using the software Ortec Gamma Vision® (Ortec GammaVision-32

v. 6 2010). The nominal relative efficiency to the scintillation counter (86.4%), while the measured relative efficiency of the detector found to be 76.2%.

**Energy and Efficiency Calibration:**

HPGe detector energy calibration was performed over a wide range of energies. The calibration was conducted by using full-energy peaks from a standard source (SYRNORM 2005) in the shape of a cylindrical plastic container with Ra-226 activity of 9775 Bq $kg^{-1}$.

**Minimum Detectable Activity (MDA):**

The minimum detectable activity (MDA) for each radionuclide was obtained from the background radiation spectrum for the same counting time (24hr) as for the soil samples. The values were calculated using equation (1) (H. I. Kohn,1989).

$$MDA = \frac{N_d}{\varepsilon P_\gamma T\, m} \quad (1)$$

Where ;

$$N_d = L_c + 2.706$$

$$L_c = 4.653\,\sigma_{NB}$$

and, $N_d$ ,$\sigma_{N_B}$, $\varepsilon$, $P_\gamma$, $L_c$ are the counts, their standard deviation, detector efficiency, the emission probability of the gamma line corresponding to the peak energy, critical level, respectively, and T is the sample counting time. m is sample mass.

## RESULTS AND DISCUSSION:

### Detection Limit and Minimum Detectable Activity (MDA):

The minimum detectable activity obtained was in the range of $0.2549 \pm 0.001$ Bq $kg^{-1}$ to $1.864 \pm 0.002$ Bq $kg^{-1}$ as shown in Figure 2. This range of values was used to check if the activity concentrations found within the samples are of measurable values throughout this study.

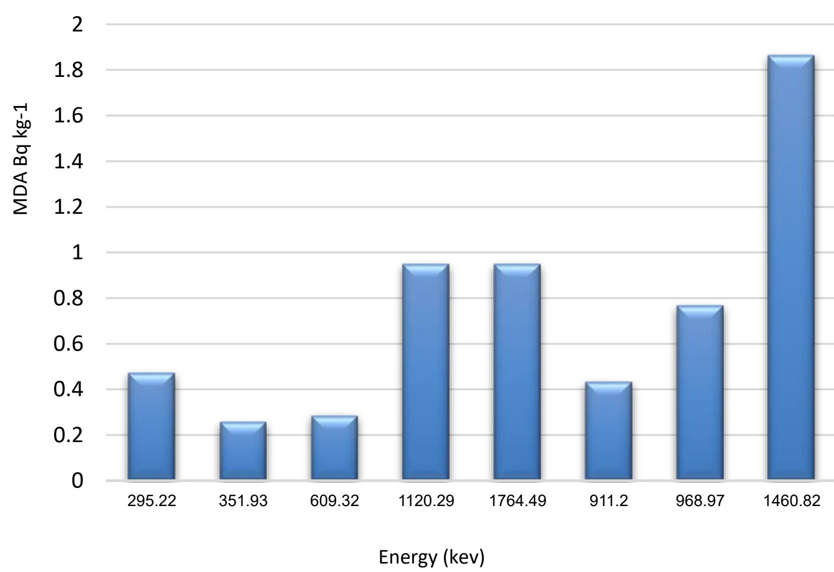


Figure 2: Calculated MDA for listed radionuclide energy

**Samples Specific Activity Concentration Measurement (Ac):**

The activity concentration of an identified radionuclide produced by $^{238}U$, $^{232}Th$ chains and $^{40}K$ were calculated using the following equation:-

$$Ac(Bq/kg) = \frac{N-B}{\varepsilon \times t \times I_{\gamma} \times m} \qquad (2)$$

Where $\varepsilon$ is the absolute efficiency at a given photo peak energy, $I_{\gamma}$ is the emission probability of the gamma line corresponding to the peak energy, $m$ is the mass of the measured sample (kg), $N$ and $B$ are the areas under the photopeaks of the sample and the background, respectively (P. Sola, U. Injarean 2022, pp. 98–111& F. Caridi et al.2016).

Activity concentration of $^{238}U$, $^{232}Th$ series, and $^{40}K$ are shown in Figures 3 and 4. The gamma-energy lines of 609.32 (45.4%) keV, 1120.29 (14.8%) keV from $^{214}Bi$ were used to represent the $^{238}U$ ($^{226}Ra$) series, while 911.20 (25.8%) keV and 968.97 (15.8%) keV from $^{228}Ac$ were used to represent the $^{232}Th$ series, and 1460.82 (10.66%) keV for $^{40}K$ gamma-line. Activities were compared with the world limit (33 Bq kg$^{-1}$ for $^{238}U$, 45 Bq kg$^{-1}$ for $^{232}Th$, and 420 Bq kg$^{-1}$ for $^{40}K$) reported by UNSCEAR 2008 (G. Silini 1981).

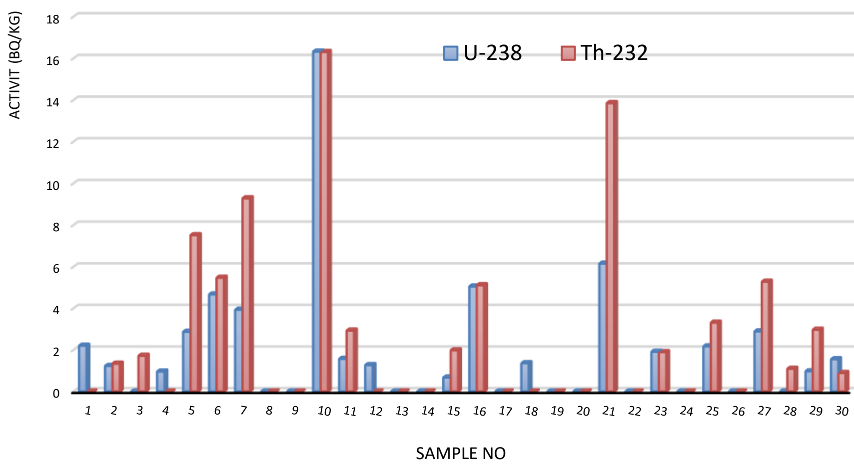


Figure 3: Activity concentrations (Bq $kg^{-1}$) distribution of U−238, Th−232 for all samples

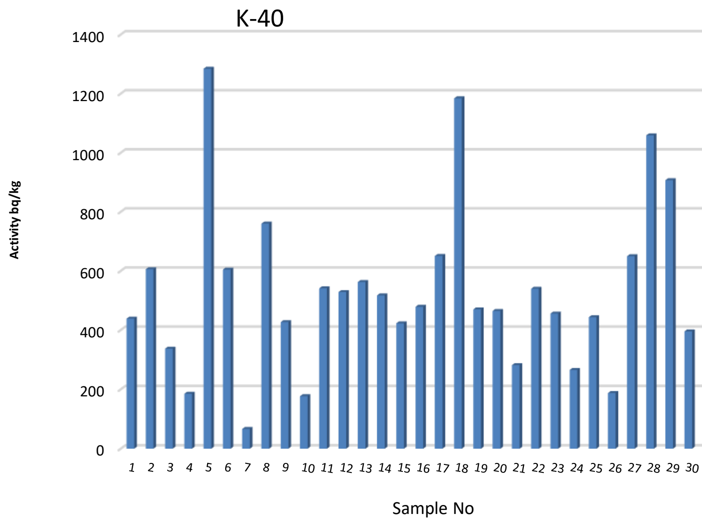


Figure 4: Activity concentrations (Bq $kg^{-1}$) distribution of Potassium K-40 for all samples

Results of this study were compared with a study conducted in Nigeria (Ononugbo, et al., 2019, pp. 1–9), it was found that the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ in (nutmeg, garlic, ginger, white onion and black pepper). The concentration of $^{238}U$ was slightly higher than the indicated study, while, the $^{232}Th$ and $^{40}K$ concentrations were lower than the indicated results.

Another comparison of some of the results of this study was made with a study conducted in Egypt (Amin and Ahmed 2013, pp. 350–354), and it was found that the activity concentrations (turmeric, rosemary, thyme, red pepper, cumin, coriander, cardamom, black pepper and nutmeg), were in $^{238}U$ and $^{232}Th$ were much lower than those in the indicated study, while $^{40}K$ was higher in (red pepper, cumin, cardamom and black pepper) than the indicated results.

**Gamma Absorbed Dose Rate Evaluation:**

The Radium equivalent specific activity in all samples was estimated using equation 3:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$

Where $A_{Ra}$ , $A_K$ and $A_{Th}$ , are the specific activities of $^{238}U$, $^{40}K$ and $^{232}Th$ in Bq $kg^{-1}$. The calculated $Ra_{eq}$ found to be in the range between 7.743±0.359 and 44.681±0.149 Bq $kg^{-1}$, which were below the world's mean values of 370 Bq $kg^{-1}$ (H. S. Eissa, M. et al.2011, pp. 533–540 & El-Kameesy, et al. 2008, pp. 245–251 ).

The gamma dose rate (D) in nGy $hr^{-1}$ from the NORM in air at 1 m above the ground surface can be calculated using the following equation (F. Caridi et al.2016):-

$$D = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_K \quad (4)$$

Where $A_{Ra}$ , $A_{Th}$ and $A_K$ are the specific activities of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in (Bq $kg^{-1}$).

The range for gamma dose rate calculated in this study was found to be between 59.20±0.13 and 7.77±0.36 nG $hr^{-1}$. $Ra_{eq}$ ranged between the

14.44±0.26 and 112.50±0.10Bq $kg^{-1}$ and below the recommended maximum value of 370 Bq $kg^{-1}$ in UNSCEAR(2000). Figure 5 and Figure 6 show the distribution of Dose rate (nGy $hr^{-1}$) and $R_{eq}$, for all samples. Results revealed that the lowest values were recorded for Nutmeg (SP4), (8.11±0.35 nGy $hr^{-1}$ ;15.23±0.26 Bq $kg^{-1}$), and Sumac (SP5), (7.77±0.36 $nGyhr^{-1}$; 14.44±0.26 Bq $kg^{-1}$), while the highest values were obtained from sample Cardamom (SP5), (59.20±0.12 $nGyhr^{-1}$;112.50±0.10 Bq $kg^{-1}$), and Nulta jute (SP18), (49.65±0.14 nGy $hr^{-1}$; 92.54±0.0.10 Bq $kg^{-1}$).

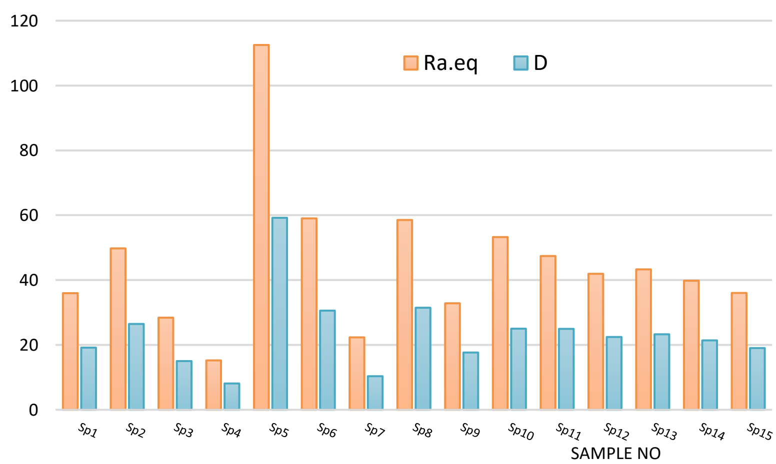


Figure 5: Dose rate (nG/h) and $Ra_{eq}$ (Bq $kg^{-1}$) distribution for samples Sp1 to Sp15.

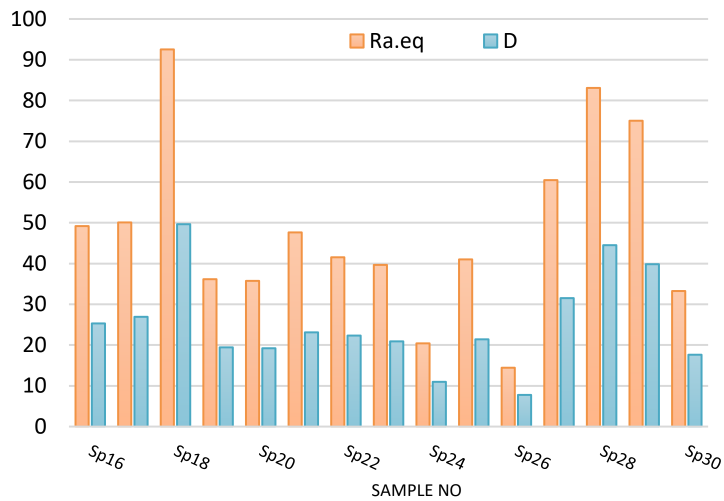


Figure 6: Dose rate (nG/h), $Ra_{eq}$ (Bq $kg^{-1}$) distribution for samples (Sp16 to Sp30)

**The Annual Effective Dose Rate:**

The annual effective dose equivalent in μSv/y was calculated using equation (Al-Mashhadani et al. 2020, pp. 0-7).

$$AEDE(\mu Sv/y)=D(nGy/h) \times 8760(h/y) \times 0.2 \times 0.7(Sv/Gy) \times 10^{-3} \qquad (5)$$

The annual effective dose equivalent was reported to be 70 μSv $y^{-1}$ in UNSCEAR 2000 and UNSCEAR 1993, for environmental exposure to gamma rays of moderate energy. Figure 7 shows the AEDE in μSv $y^{-1}$. The estimated values of AEDE were compared with some previouse studies and findings showed that the range was slightly higher in this study and far from the risk limit where the range was from 9.52 to 72.57 μSv $y^{-1}$, while it was between 0.85 and 46.76 μSv $y^{-1}$ in other study.

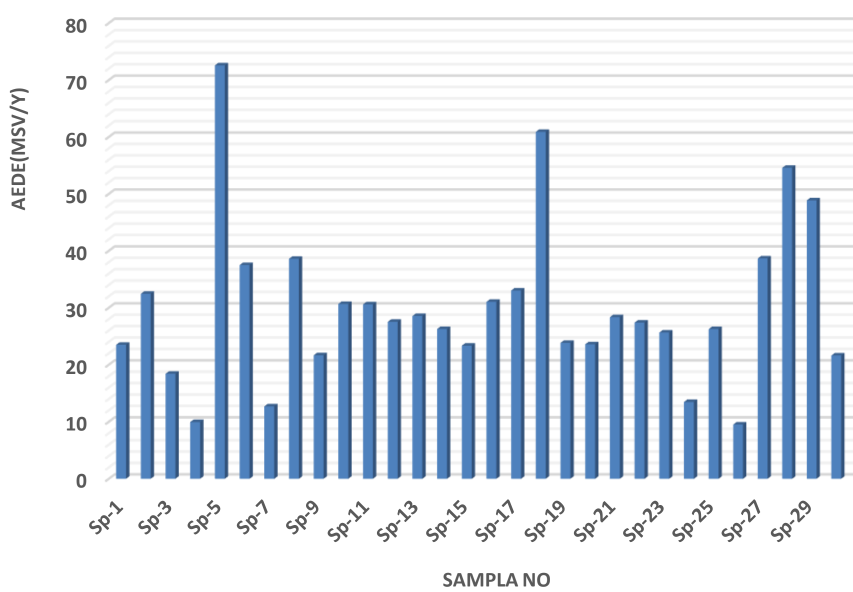


Figure 2: annual effective dose equivalent for each sample

**CONCLUSION:**

This study was conducted to estimate the activity concentration of radionuclides $^{238}U$, $^{232}Th$ and $^{40}K$ using gamma ray spectrometry in 30 different samples of food spices widely used in Libyan cuisine. All samples were prepared and analysed at the department of Nuclear Engineering, University of Tripoli. The activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ based on the weighted mean values across samples were (1.92±0.72), (2.70±0.61) and (529.50±0.04) Bq $kg^{-1}$, respectively. Some measured samples revealed an activity concentration of $^{238}U$ and $^{232}Th$ below the detection limit. The highest activity was recorded in cinnamon contained $^{238}U$ (16.32±1.64 Bq $kg^{-1}$ and $^{232}Th$ 16.30±1.65 Bq $kg^{-1}$. The lowest activity concentration of $^{40}K$ recorded in the white pepper with 66.41±5.11 Bq $kg^{-1}$, while the highest value in the sample was recorded in cardamom with 1284.33±88.32 Bq $kg^{-1}$.

The annual effective dose equivalent in μSv $y^{-1}$ ranged between 9.52 and 72.57 μSv $y^{-1}$ while the world average annual effective dose equivalent (AEDE) from outdoor or indoor terrestrial gamma radiation is 290 μSv $y^{-1}$. The results of this study were slightly higher than those achieved in many other studies conducted in Egypt, Iraq, and other countries. High concentration of potassium was found in many samples. Therefore, it is vital to monitor to daily potassium intake by wathcing the combination of food and potassium concentration in it. On the other hand, high potassium intake needs to be followed by a reduction in sodium and salt intake in food.

**الخلاصة**

تعد تركيزات النشاط الإشعاعي في النظام الغذائي البشري مصدر قلق كبير لتقدير المخاطر الإشعاعية على صحة الإنسان. ومع ذلك، لم يتم إجراء العديد من الدراسات حول النشاط الإشعاعي في الغذاء في ليبيا. في هذه الدراسة تم تحديد تركيز النشاط لمستويات النويدات طبيعية المنشأ في مجموعة من 30 عينة من التوابل الغذائية. تم جمع العينات من أحد أكبر الأسواق المحلية في طرابلس، ليبيا، تحديدا سوق سليمان خاطر. تم تحضير وتحليل جميع العينات بمعامل قسم الهندسة النووية بجامعة طرابلس لتحديد تركيزات النشاط المرتبطة بسلاسل الانحلال الطبيعي U238 $^{238}$، $^{232}Th$ و $^{40}K$ .

كان متوسط تركيز النشاط الاشعاعي في العينات لل $^{238}U$ و$^{232}Th$ و $^{40}K$ ، ($0.72 \pm 1.92$) و ($0.61 \pm 2.70$) و ($0.04 \pm 529.50$ (بيكريل/كجم على التوالي.

أظهرت النتائج المحققة أن تركيز النشاط كان ضمن المستوى المقبول لكل من $^{238}U$ ، $^{232}Th$ بينما كان مستوى تركيز $^{40}K$ مرتفعًا في عينات الهيل والفلفل الأحمر الحار المستورد من الهند ، والكركم ، والكركم المختلط من ليبيا حيث كانت أعلى بكثير مما ورد في تقرير (2000) UNSCEAR.

تم تسجيل أعلى تركيز نشاط اشعاعي ل $^{238}U$، $^{232}Th$ في القرفة المستوردة من الهند والتي كانت $16.32 \pm 1.64$ و $16.30 \pm 1.65$ بيكريل/كجم على التوالي. اما النشاط الاشعاعي ل $^{40}K$ فقد حدد بين $66.41 \pm 5.11$ بيكريل/كجم للفلفل الأبيض و $1284.33 \pm 88.32$ بيكريل/كجم للحبهان. وتراوحت الجرعة الفعالة السنوية المكافئة بين 9.52 و 72.57 ميكروسيفرت/سنة بينما كان المتوسط العالمي للجرعة الفعالة المكافئة (AEDE) من إشعاع جاما الأرضي الخارجي أو الداخلي 290 ميكروسيفرت/سنة.

كانت نتائج هذه الدراسة أعلى قليلاً من تلك التي تحققت في العديد من الدراسات الأخرى التي أجريت في مصر والعراق ودول أخرى. تم العثور على تركيز عال من البوتاسيوم في معظم العينات. لذلك، من المهم الاهتمام بكمية البوتاسيوم اليومية من خلال النظر إلى مزيج الطعام وتركيز البوتاسيوم فيه، مثل زيادة استهلاك البهارات الغذائية.

**الكلمات الرئيسية** – المواد المشعة طبيعية المنشأ، النشاط الاشعاعي، مطياف جاما، الجرعة الفعالة السنوية:

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