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Evaluation of Radioactivity and Heavy Metal Concentrations in Some Liquid Supplements Samples

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Abstract
In recent years, it has become clear that consumers of nutritional supplements, especially adults, tend to consume these supplements for the purpose of improving health and delaying chronic diseases associated with age, in addition to improving physical and mental performance. These supplements are widely available in all pharmacies and medical stores in Iraq. Therefore, the current study aims to determine the concentrations of uranium and its isotopes and the concentration of radon gas and calculate the associated radiological hazard using the CR-39 detector by the exposure and irradiation method, and to calculate the concentration of heavy metals and hazard coefficients using the flame atomic absorption spectroscopy technique. The results showed that for Uranium isotopes activity values for (U^{238}, U^{235}, U^{234}) were (5.23±0.3144Bq/L), (0.244±0.014Bq/L), and (5.402±0.324Bq/L), and The average value of the total annual effective dose (AED) and for the risk of deaths from cancer per million individuals (RECFPMP) were ($2.02\times10^{-6}\pm3.83\times10^{-7}$) (mSv/y) ($6.84\times10^{-6}\pm1.437\times10^{-6}$) respectivly, while for radon the radon concentration in the air and The annual effective doses (AED) and the lung cancer cases per million (CPPP) have an average value were (22.421±17.389 Bq/L), (0.565 ±0.155mSv/y) and (10.181±7.896), and for heavy metals The concentrations were measured for each (Cd, Cr, Cu, Zn and Fe) were (0.307±0.020ppm), (0.028±0.005ppm), (0.714±0.048ppm), (2.714±0.188ppm)and (0.145±0.035ppm), and for The daily intake of carcinogens(CD_{Ica}) The highest value was for Zn with the average value equal ($8.92\times 10^{-9} \pm 6.19\times10^{-10}$ mg/kg/day), and lowest value was for Cr with average value ($9.33\times10^{-11}\pm1.57\times10^{-11}$ mg/kg/day) , and The risk of cancer (CR) was calculated for both Cr and Cd with an average are ($4.66\times10^{-11}\pm7.86\times10^{-12}$), ($6.36\times10^{-9}$ $\pm4.07\times10^{-10}$) ,and finally for Total Cumulative Cancer Risk (TCCR) with an average is (6.41×10^{-9} $\pm1.147\times10^{-9}$),all measured results were less than the acceptable limits.
Keywords: Radioisotopes, Dietary supplements, Cancer Risk, Flame atomic absorption spectroscopy, Heavy metal.

1. Introduction

The use of nutritional supplements for adults aims to improve health and delay chronic diseases. However, most studies related to the use of nutritional supplements among adults have proven that these supplements have good health benefits, specifically with regard to primary prevention of some diseases such as cardiovascular disease, stroke, and depression [1]. In general, the safety of nutritional supplements is not subject to any specific challenges when marketing them, unlike herbal nutritional supplements, which are always subject to rigorous testing before being put on the market. Some studies have shown that some nutritional supplements contain chemicals or radioactive materials that lead to liver damage. In addition, it has been noted that some companies are working to add adulterated substances with synthetic drugs and their

analogues, which include stimulants and antidepressants. Therefore, harmful effects are not diagnosed early, but the damage is discovered after a period of time. Most supplements are also sold without a prescription, so they are consumed more frequently than traditional, controlled medications. It is also possible for nutritional supplements to interact with medical treatment and lead to a certain disorder [2]. Foodstuffs, and even supplements, are not safe. In its entirety, some of them are contaminated to a certain degree, especially with heavy metals and even radioactive materials [3-5]. Contamination may occur due to the frequent use of medicinal plants as raw materials that carry heavy metals due to the environment in which they are grown, in addition to their contamination from the soil [6].
The aim of this study was to measure concentrations and risk factors by using CR-39 solid-state nuclear track detector to measure uranium and radon in nutritional supplements for adults through irradiation and exposure. As for heavy metals, the technique of atomic absorption spectroscopy was used.

2.Materials and Methods

2.1. Sample Collection and Preparation

Uranium, radon and heavy metal concentrations were calculated by collecting 8 samples of nutritional supplements from Pharmacies and medical stores. The samples were classified based on sample name, code, manufacturer, and origin, as shown in **Table 1**:

Table 1. Samples of nutritional supplements used by adults and approved in the study

Sample	Code	Manufacturer's name	Origin
For ferro	S1	abmsvl	Italy
Hemafer	S2	Unipharma	Greece
Omega 3 fish oil	S3	NOW	USA
Fex Plex	S4	Versalya	Italy
Alca-plex	S5	Roxy	USA
Glovit	S6	Global pharma	UAE
Zinc Cris	S7	Cipla	India
Skin age	S8	Yaseka	Croatia

2.2. Experimental part

About 8 samples of nutritional supplements used by adults were collected, to ensure sample preparation, the samples were mechanically ground using an electric grinder to obtain a homogeneous powder. The material was then filtered by passing it through 0.75 µm pores. A sensitive digital balance with an accuracy of ±0.01% was used to measure and record the weight of each sample used, Irradiation method for calculate the uranium concentration was to expose the samples to neutron radiation, and the track densities were measured the uranium concentrations. Each sample weighed about (1ml) and was pressed into 1.6 cm diameter and 1 cm thick pellets. The pellets were fixed on a CR-39 detector measuring 1.5 cm x 1.5 cm. The specific symbol was placed next to each sample. The samples were arranged in a circular ring and placed on a paraffin wax plate (5 cm) from a ^{241}Am-^{9}Be neutron source, as shown in **Figure 1**[7]. The samples were irradiated for 7 days to produce fission tracks to detect alpha particles produced by fission fragments. The fission track densities were measured on CR-39 detectors and then they were chemically etched using a NaOH solution (N = 6.25g) for 2 hours at 60 °C [8]. The tracks that developed on the surface of each detector were carefully counted using an optical microscope.

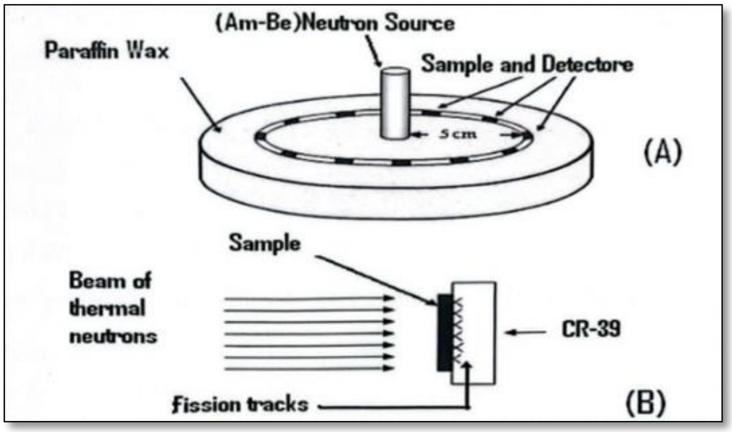

Figure 1. Irradiation of the sample with the detector[7]

And for radon exposure for measuring radon concentration, (1mL) of supplement powder was taken and placed in tightly sealed plastic containers with a height of 6.5 cm and a diameter of 3.5 cm, as shown in **Figure 2**, Each container was weighed and ensured to be empty before and after adding the supplement samples, to determine the exact sample weight. A 250µm thick CR-39 tracer (from Teasel, England) was then cut into (1.5 cm x 1.5 cm) and glued to the inside of the container lid. The plastic containers were tightly sealed to prevent radon from escaping and to allow the radon and its progenitors time to establish long-term equilibrium. The detectors were kept in the plastic containers for 90 days. After exposing the CR-39 detectors to radiation, chemically etched as previously documented in the section above:

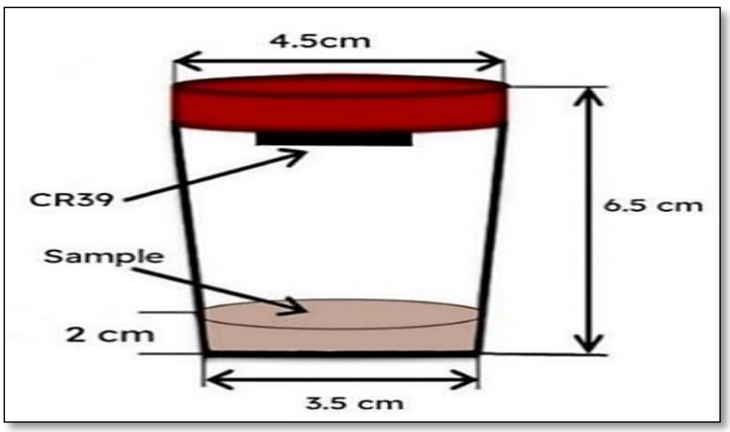

Figure 2. CR-39 detector in the supplement sample

Finally, use flame atomic absorption spectroscopy (FAAS) technique was used to determine the concentrations of heavy metals in food supplement samples. It is important to know that this method enables the analysis and determination of the concentration of many different metals in materials. In this study, the concentrations of (Zn, Cr, Cd, Fe, and Cu) were measured using the (NOV AA model) at wavelengths ranging between (350 and 400 nm).

2.3. Calculations

2.3.1.Measuring Uranium Concentration and Calculating Coefficients

Uranium concentration can be calculated by first calculating the track density using the **Equation (1)** [9]: $(N_A) = N_A\ (\text{track}) / A\ (\text{mm}^2)$ **(1)**

Where N_A represents the total track rate and A represents the field of view, which is 0.0676 mm^2. Uranium concentrations in food supplement samples are measured by comparing the track density recorded on detectors in contact with the sample and the standard track density, using the **Equation (2)** [9]:

$$C_x / \rho_x = C_s / \rho_s \quad \textbf{(2)}$$

When C_x is the uranium concentration in the unknown sample in ppm ،C_s is the uranium concentration in the standard sample in ppm، ρ_x is the track density in the unknown sample in track/mm^2، ρ_s is the track density in the standard sample in track/mm^2 ، and to calculate the specific activity in (Bq/L or Bq/kg): For uranium isotopes, **Equation (3)** [10], can be used:

A(Bq/kg) = Uc (mg/ kg) x I.M.A(%) x S.P.A (Bq/mg) **(3)**

Where I.M.A represents the relative abundance of uranium isotopes(U^{238}-U^{235}-U^{234}) (99.27, 0.72, 0.0055 respectively) and S.P.A is the specific activity of uranium (12.44, 80, 230700) respectively [11], The annual effective dose (E_U) in (Sv/y) committed to the population due to uranium isotopes (U^{238}, U^{235}, U^{234}), resulting from the intake of nutritional supplements in this study, is given by the following **Equations (4 and 5)** [12]:

$$E_{Ui}\,(Sv/y) = I_{Ui}\,(Bq/\ ) \times e(g)_{Ui}\,(Sv/Bq) \quad \textbf{(4)}$$

$$I_{Ui}\,(Bq/\ ) = \mathrm{A}\,(Bq/L\ or\ kg) \times dt\,(L\ or\ kg/y\ ) \quad \textbf{(5)}$$

Where I_{Ui} is the annual intake of three isotopes, and $e(g)_{Ui}$ are the ingestion dose coefficients for three Isotopes ((U^{238}, U^{235}, and U^{234}) have the following values: (4.40×10^{-8} Sv/Bq, 4.60×10^{-8} Sv/Bq and 4.90×10^{-8} Sv/Bq) respectively [12]. As for dt which is the total daily intake of the samples in the current study in L/day, according to recent studies, is a daily dose given to athletes as supplements [13]. In addition, the equation was also used to determine (RECFPMP) due to uranium intake from food supplement samples in our current study using **Equation (6)** [14]:

$$\mathrm{RECFPMP} = Total_{EU} \times DL \times RF \quad \textbf{(6)}$$

Where DL is the life expectancy (70 years), and RF is the radiation dose recommended by the International Commission on Radiological Protection (0.05 Sv^{-1}) [15].

2.3.2. Measuring Radon Concentration and Calculating Coefficients

For radon, the alpha particle track density recorded on the CR-39 is the Rn component, so its concentration is directly proportional to the track density readings. Therefore, the measured:

Radon concentrations in the air: Radon concentrations in the air above the sample inside the enclosure are represented by (Bq/m³) can be calculate according to the following **Equation (7)** [16]:

$$C(Bq/m^3) = \rho\ /kt \quad \textbf{(7)}$$

where ρ is the track density, which represents the average number of total tracks for each area of the field of view of the ten images, t is the exposure time (90 days) , and k is the slope of the calibration curve obtained theoretically using the **Equation (8)** from which we obtain the constant [9]:

$$K=0.25r\,(2 \cos \theta_c - r/r_a) \quad \textbf{(8)}$$

where r is the radius of the enclosure in centimeters, r_a is the range of alpha particle movement in the air (4.15 cm), and θ_c represents the critical angle of the detector (45^0), The value of k Constant was equal = 0.046.

Annual Effective Dose: be calculated in (mSv/y) by using the **Equation (9)** [17]:

AED (msv/y) = F x C x 0.80 x 8760 x D **(9)**

Where F is the equilibrium factor, with a value of (0.4) recommended by the United Nations Scientific Committee on Atomic Radiation Research in 2000 [17], the value of 0.80 represents the occupancy factor, which is the amount of time a person spends at home, 8760 is the number of hours per year, and D represents the ICRP dose conversion factor, with a value of $9*10^{-6}$ (mSv/Bq.h.m^{-3}) [18].

Lung Cancer Cases: This quantity can be calculated using the **Equation (10)** [17]:

CPPP = AED x $18*10^{-6}$ y/mSv **(10)**

2.3.3. Measuring concentration of Heavy Metal and Calculating Coefficients

Many health risk factors were calculated due to Zn, Fe, and Cu, Cd, and Cr concentrations in nutrition supplement samples in the study by using carcinogens parameters as follows:

Chronic Daily Intake for Carcinogens (CDI_{ca}): can calculate by use **Equation (11)** [19]:

$$CDI_{ca}(mg/kg.\,d^{-1}) = Cs \times IR \times EF \times ED \times CF/BW \times AT \quad \textbf{(11)}$$

where IR equal to (0.24 mg/day) of supplement sample 20, and Cs is the concentrations of (Zn, Fe, Cu, Cr and Cd (in mg/kg or ppm), EF is exposure frequency (350 d/y) [21], ED is exposure duration (30 y) [19], CF is conversion factor (10^{-6} kg/mg), BW is body weight (70 kg), AT is equal to 365.70 days [21].

Cancer Risk (CR): can calculate by use **Equation (12)** [22]:

$$CR = CDI_{ca} \times SF \quad \textbf{(12)}$$

Where SF was the slope factor equal for Cr, and Cd have constant values [23].

Total Cumulative Cancer Risk (TCCR): can calculate by use **Equation (13)** [22]:

$$TCCR = \sum CDI_k \times SF_k \quad \textbf{(13)}$$

where (TCCR) is the total cumulative cancer risk, which is the result of multiplying the chronic daily intake of carcinogens (CDI_{ca}) and the slope factor (SF) for each element.

3.Results

3.1. Uranium concentration and risk factors in dietary supplements

Uranium concentration and risk factors were measured in 8 samples of dietary supplements, **Figure3** shows the values of Uranium isotope activity, for U^{238} activity ranged values from (6.919-4.097Bq/L), with average (5.23±0.3144Bq/L), U^{235} activity ranged from (0.323-0.191Bq/L), with an average (0.244±0.014Bq/L), and U^{234} activity ranged from (7.141-4.228Bq/L), with an average value equal to (5.402±0.324Bq/L).

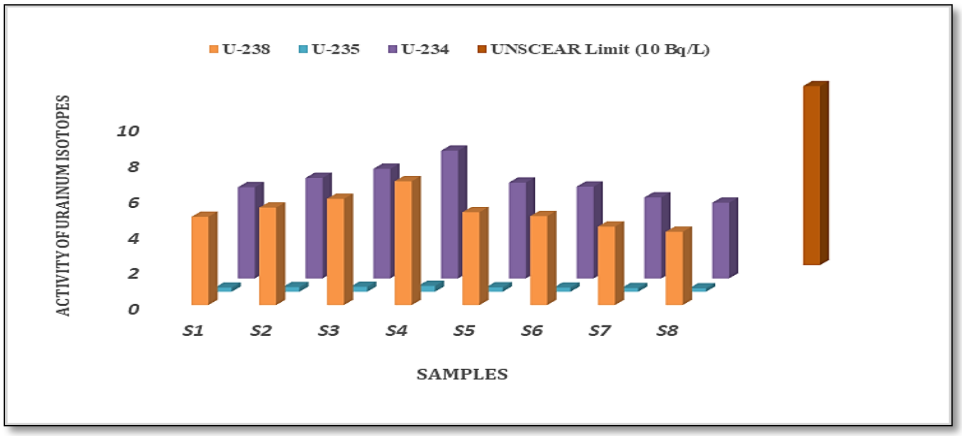

Figure 3. Activity of Uranium isotopes values for supplement sample with acceptable limit (UNSCEAR)

The annual effective dose (AED) was calculated for all samples as shown in **Figure 4**, where the total AED values range from (2.616×10^{-6} mSv/y) for sample 6 to (1.446×10^{-6} mSv/y) for sample 8.

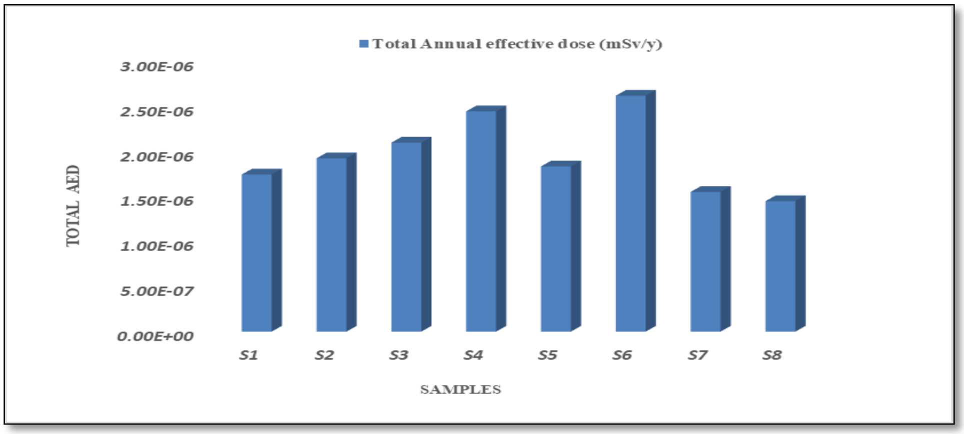

Figure 4. Total annual effective doses of isotopes in supplement samples

Figure5, shows the statistical distribution of the measured annual effective dose (AED). The results showed that the average dose with a standard deviation ($2.02\times10^{-6}\pm3.83\times10^{-7}$), which reflects the closeness of the values and the absence of significant variation between the samples.

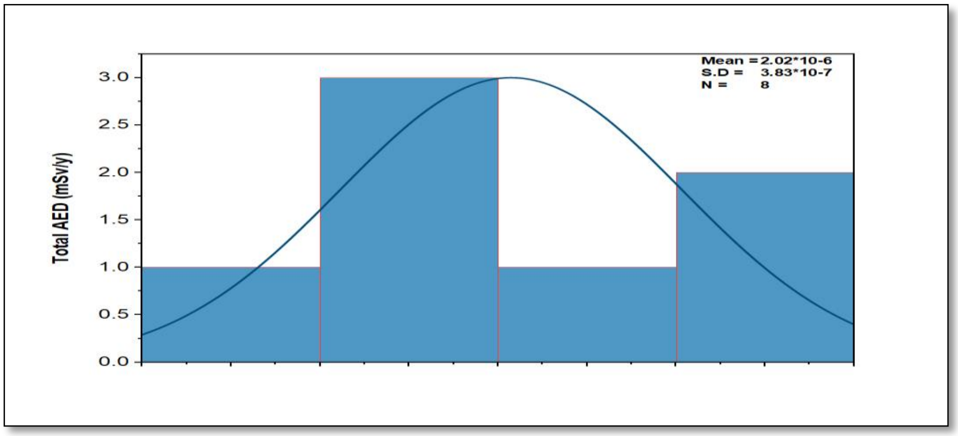

Figure 5. Distribution curve of total AED for different supplement samples

For the (RECFPMP) was ranged from (9.157×10^{-6}) in sample (6) to (5.062×10^{-6}) in sample (8), as show in **Figure 6**:

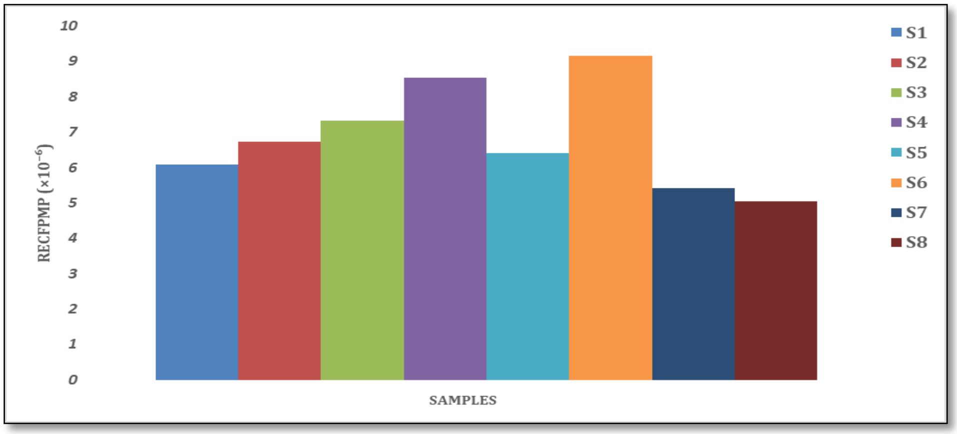

Figure 6. (RECFPMP) values for different supplement samples

A histogram distribution with a normal curve was performed the risk index data (RECFPMP) **Figure7**, shows the statistical distribution, which shows that the values are distributed close to the normal curve, with the mean being ($6.84\times10^{-6}\pm1.437\times10^{-6}$), This indicates that the data are close to normal distribution.

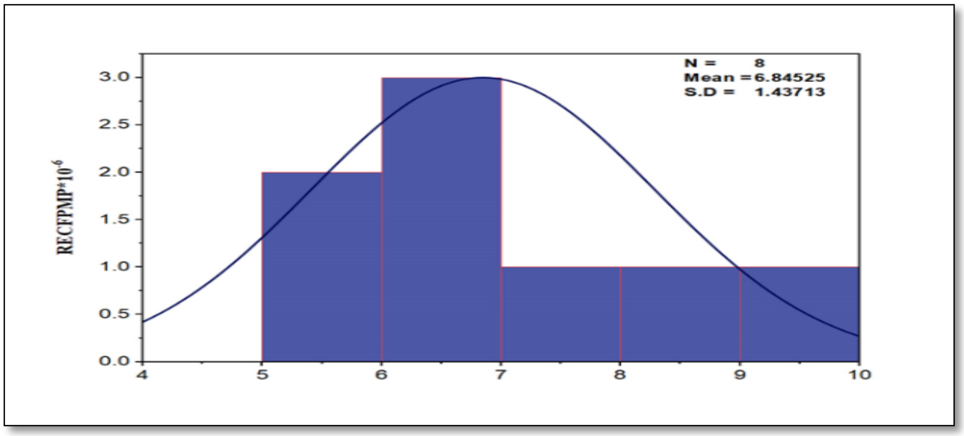

Figure 7. Distribution curve of (RECFPMP) for different supplement samples

3.2.Radon concentration and risk factors in nutritional supplements

Radon concentrations and risk factors were determined for 8 dietary supplements over a (90 day), when using the CR-39 nuclear trace detector with exposure method. radon concentrations in air above samples are ranged from (2.143 Bq/L) in Sample 2 to (43.592Bq/L) in Sample 6, In **Figure8** shows the different values of radon concentration in air with the acceptable limit:

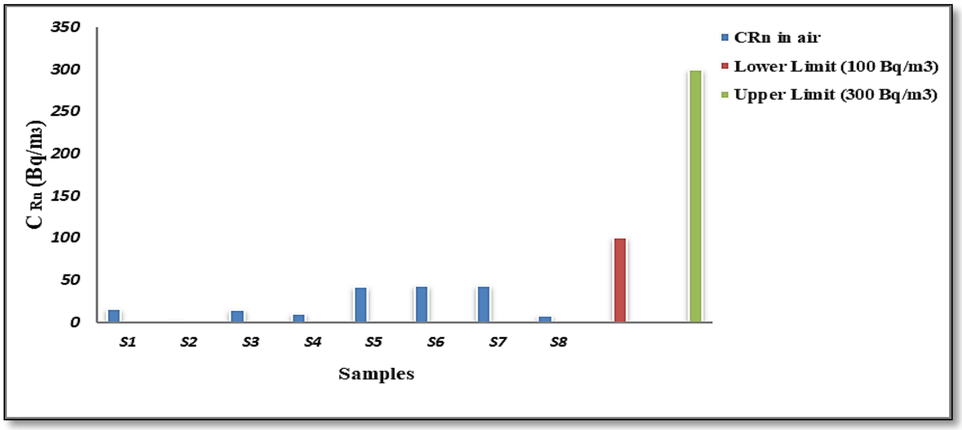

Figure 8. Radon concentration values for supplement samples with acceptable limit

A statistical distribution of radon gas concentration was conducted, and as shown in **Figure 9**, the average concentration was (22.421±17.389 Bq/L), indicating that most values are close to the average with relatively limited variation. The consistency of the data with the normal curve reflects the reliability of the results and their potential for use in assessing the radiation risks associated with radon exposure.

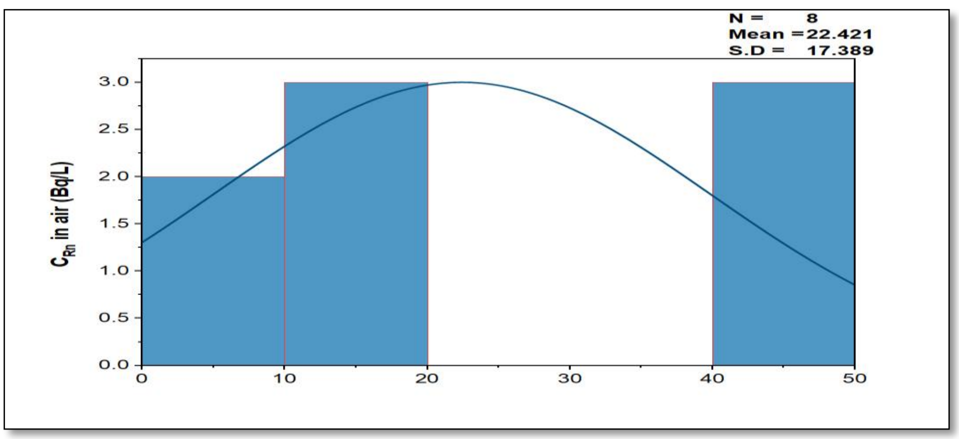

Figure 9. Distribution curve of radon concentration in air

The annual effective doses (AED) were expected to vary between (0.054mSv/y) for Sample 2 and (1.099mSv/y) for Sample 6, with an average value of (0.565 ±0.155mSv/y), as shown in **Figure 10:**

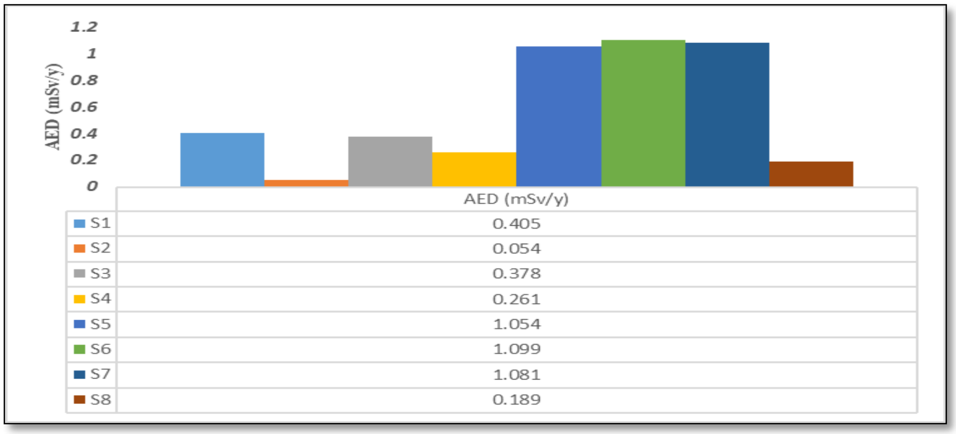

Figure 10. AED values for different supplement samples

The incidence of lung cancer per million population (CPPP) also were change from ranged between (0.973) per million population in sample (2) to (19.796) per million population in sample (6), showed in **Figure 11:**

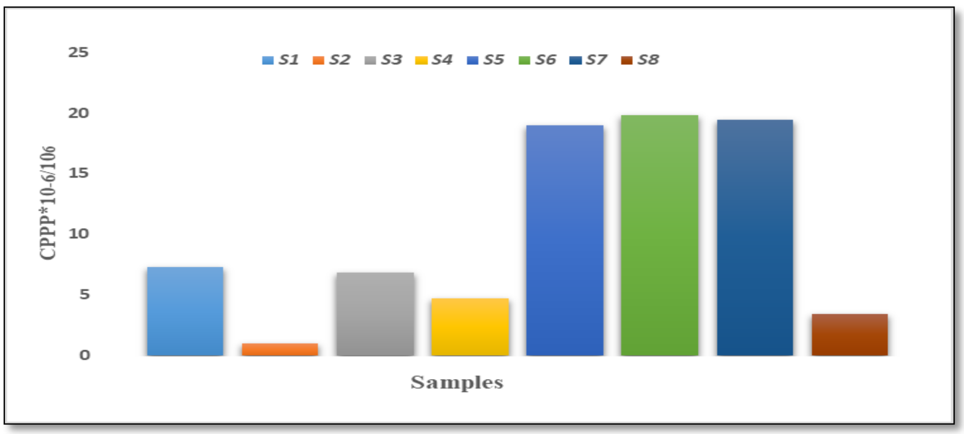

Figure 11. (CPPP) values for different supplement samples

The statistical distribution of the cancer risk factor (CPPP) values was performed, as shown in **Figure 12** and the results showed that the average was (10.181±7.896), indicating that most of the values are clustered around the average with relatively limited dispersion. The conformity to the normal curve enhances the reliability of the data and reflects the possibility of adopting it in assessing the health risks associated with radiation exposure.

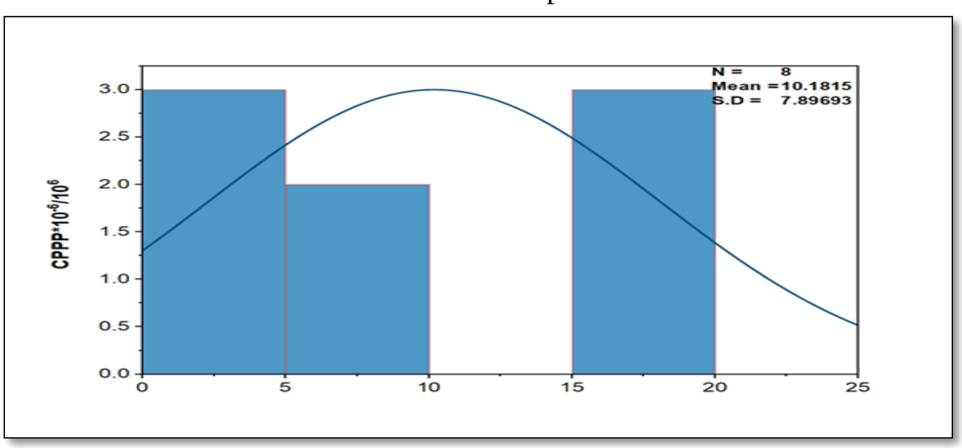

Figure 12. Distribution curve of (CPPP) for different supplement samples

A statistical analysis was conducted to find the relationship between the concentration of radon in the air and (AED). **Figure 13** shows that (AED) depend directly on the radon concentration. Therefore, the higher the radon concentration, the higher (AED). The figure below confirms the presence of a strong positive correlation between (AED) and radon concentration, where the correlation coefficient was (R^2=1).

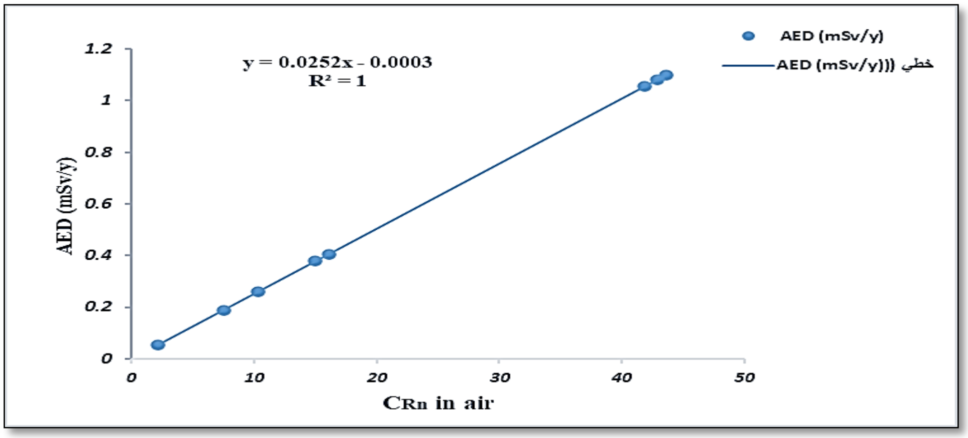

Figure 13. Distribution between the Radon concentration and (AED)

3.3.Heavy Metal Concentrations and Risk Factors in Dietary Supplements

Heavy metal concentrations and some risk factors were measured in 8 samples of dietary supplements, **Figure 14** shows the values of heavy metal concentrations (Cu, Fe, Zn, Cr, and Cd), Cd concentration values with the average was (0.307±0.020ppm). for Cr concentration, values with an average were (0.028±0.005ppm). also, Cu concentration in supplements, value with an average were (0.714±0.048ppm). and for Zn concentration has average value was (2.714±0.188ppm), finally concentration of Fe in supplements with have an average value was (0.145±0.035ppm).

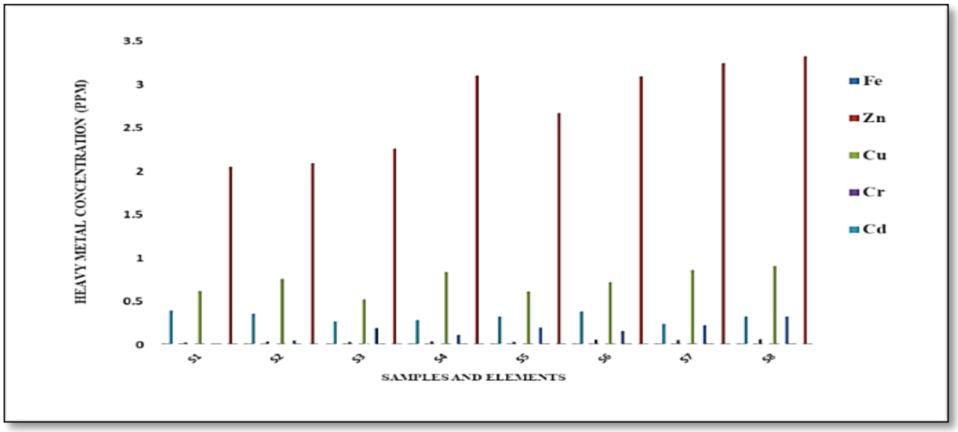

Figure 14. Heavy metal concentrations of nutritional supplement samples

The daily intake of carcinogens (CDIca), the values fluctuate. The highest value was for Zn, ranging between (1.08789×10^{-8} -6.6904×10^{-9} mg/kg/day) with the average value equal ($8.92\times 10^{-9} \pm 6.19\times10^{-10}$), and also the lowest value was for Cr, ranging between (1.578×10^{-10} - 3.616×10^{-11} mg/kg/day) with average value ($9.33\times10^{-11}\pm1.57\times10^{-11}$) **Figure 15**, shows the values of Chronic Daily Intake for Carcinogens (CDI_{ca}) for supplement samples.

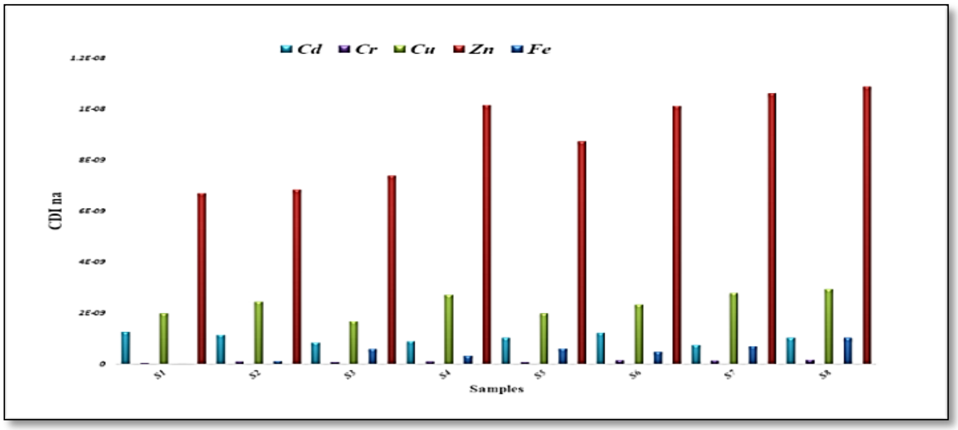

Figure 15. the values of Chronic Daily Intake for Carcinogens (CDI_{ca}) for supplement samples

The risk of cancer (CR) was calculated for both Cr and Cd only, the cancer risk values for taking a nutritional supplement containing Cr ranged between (1.81×10^{-11} - 7.89×10^{-11}) with an average of ($4.66\times10^{-11}\pm7.86\times10^{-12}$), As for Cd, its value ranged between (4.66×10^{-9} - 7.87×10^{-9}), with an average of (6.36×10^{-9} $\pm4.07\times10^{-10}$) **Figure 16** shows the values of the risk cancer for supplement samples:

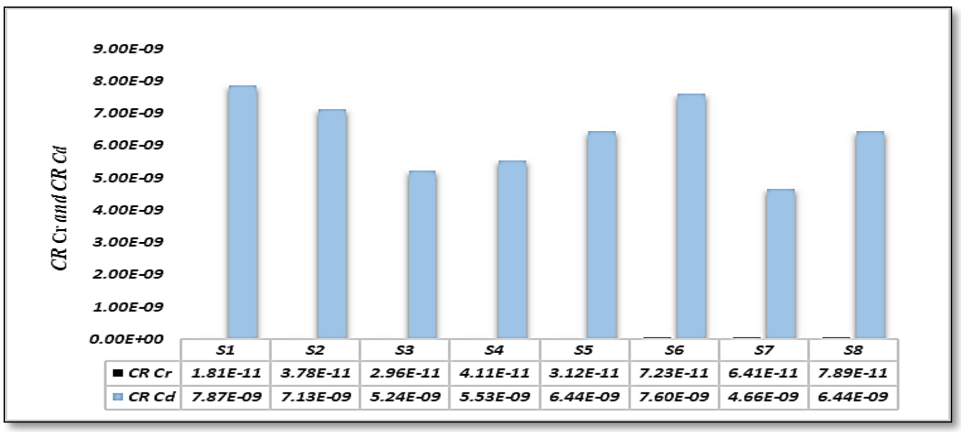

Figure 16. the values of the risk cancer for supplement samples

As for the total risk of infection, the values varied between (4.72×10^{-9}) in sample (7) and (7.89×10^{-9}) in sample (1), as shown in **Figure17**:

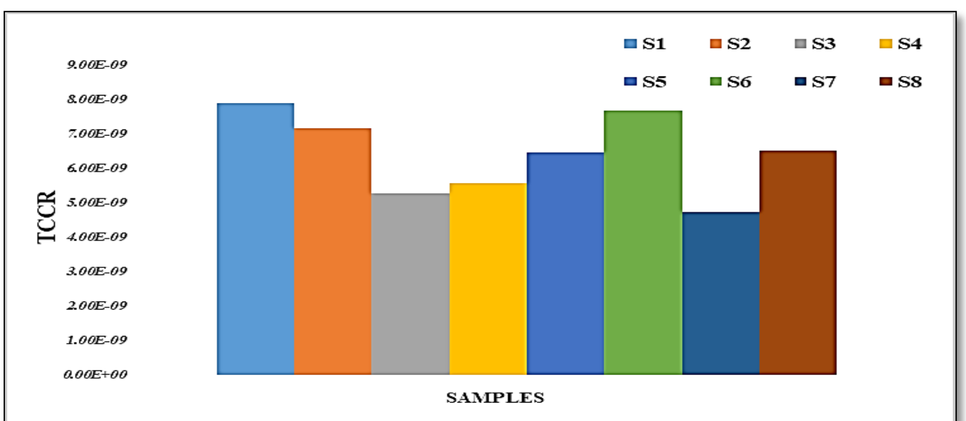

Figure 17. the values of the Total Cumulative Cancer Risk for supplement samples

A statistical analysis was conducted using **Figure 18**, which represents the normal distribution curve and shows the results of the total cumulative cancer risk factor. It was found that the ($6.41\times10^{-9} \pm 1.147\times10^{-9}$).

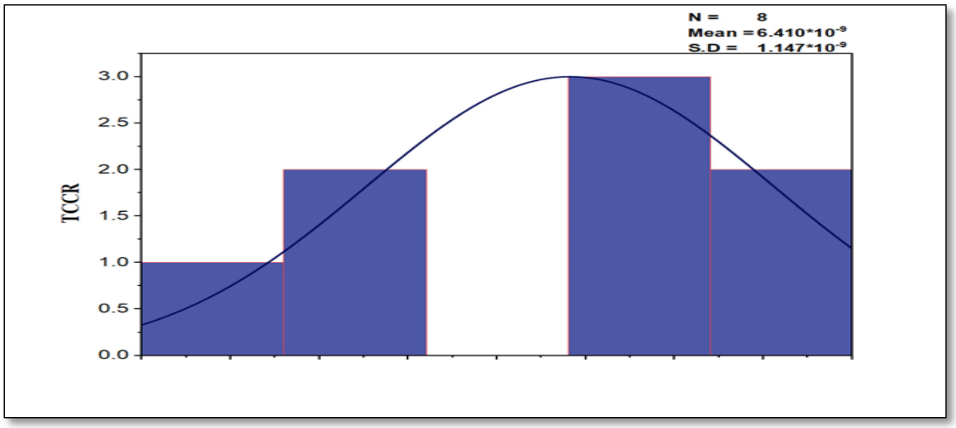

Figure 18. Distribution curve of the Total Cumulative Cancer Risk for supplement samples

4.Discussion

From the results, the uranium concentrations and comparing with the Uranium isotope activity values obtained with the 2005 World Health Organization report, and based on research, the total activity of the three uranium isotopes is equal to the internationally accepted limit of (10 Bq/L) [17], In addition in all the obtained annual effective dose values are less than (0.18 mSv/y) [24], which is less than the limits permitted by international (ICRP and WHO) for the annual effective dose resulting from radioactive materials.

Also, after conducting the statistical analysis, it was found that the rate was much lower than the permissible value that had been set 24 all recorded values fall within the global limits set by (ICRP and UNSCEAR), and therefore the level of radiation exposure resulting from these samples is considered negligible and does not affect health.

The result shows that the values obtained and the average were significantly lower than the allowed values set by the International Commission on Radiological Protection (1×10^{-3}) [25], This means that the radioactivity of the dietary supplements does not have a harmful effect on the consumer, Also, regarding the statistical analysis in result, the obtained rate was significantly lower than the permissible limit according to (USEPA & ICRP) recommendations ($10^{-6} – 10^{-4}$), indicating that the expected radiation health effects from these samples are very limited and do not pose a significant risk to consumers, the result indicates that radon concentrations in all samples were below the recommended range (100-300 Bq/m^3), as indicated in the study [26], and did not exceed the permissible limit This indicates that radon concentrations in these samples are very low and safe for human health, Furthermore, the result shows that the statistical analysis of radon concentration in the air revealed that the average was significantly lower than the permissible value, considering that radon gas is a light element that evaporates quickly and does not have a direct effect on the consumer, As for the annual effective dose, the result shows that, when comparing the values and the average results, These results are lower than the acceptable values (3-10 mSv/year) [25]. Regarding, the results obtained compared with the acceptable values set by the International Commission on Cancer Prevention (170-230 cases/million/year)[25], also Statistical analysis was conducted to determine the relationship between radon concentration in the air and AED. The finding shows a strong correlation between them. It is noted that radon can enter the human body through ingestion, although low concentrations do not pose a significant risk. However, continuous monitoring is necessary, given that radon is a significant source of natural radiation in our daily lives[26,27]. The results indicating a difference in the concentrations of heavy metals obtained. This difference is attributed to their association with the source, the manufacturing method, the type of components, quality standards, and storage conditions[28], In addition, this is lower than the acceptable limit adopted by most studies, Studies indicated that the concentration values of Fe (15 ppm), Zn (15 ppm), Cu (2 ppm), Cr (0.5 ppm), and Cd (0.3 ppm) are considered permissible and safe limits [29,30], Also,the results were lower than the allowed values, also the finding shows the cancer risk (CR) calculated for only two elements: Cr and Cd. Other elements were considered non-carcinogenic based on scientific studies[31,32], and all results obtained were much lower than the permissible limits, the results related to total risk of infection, which were found to be low compared to the acceptable values. Regarding the statistical analysis in he result, the total cumulative cancer risk was significantly lower than the expected rate This is a very small number and indicates that the risk level is well below the internationally accepted limits (usually, the acceptable risk is $\leq 10^{-6} - 10^{-4}$ according to the EPA and health agencies).

5.Conclusion

This study shows the dietary supplements examined used by adults, are not associated with radiological risks to health. All activity of uranium isotopes and radon concentrations were determined to be under internationally recognized standards. The detected amounts of heavy metals were within acceptable limits. Despite the overall risk factors being under international standards, careful monitoring is necessary as long-term exposure can cause toxicity and harm physiological systems. Therefore, although these supplements may be deemed radiologically safe, consistent monitoring of heavy metal levels is very important for ensuring consumer safety.

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Conflict of Interest

The authors declare that they have no conflicts of interest.

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Ethical Clearance

The study did not involve human participants, animal subjects, or any form of personal data collection. Therefore, ethical approval and informed consent were not required.

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