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Determination of natural radioactivity material concentrations consumed widely during Corona pandemic in Thi Qar province

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**Abstract**

The present study was carried out to assess the potential radiation hazards to the public, especially the persons who consumed onion and garlic plants widely during coronavirus disease in Thi Qar province South of Iraq. Nine samples collected from the market (5 samples onion and 4 sample Garlic), which classified according to their origin. Using 3"x3" NaI (Tl) gamma ray spectroscopy system, the radioactivity concentrations of the natural radionuclides radium-226, thorium-232 and potassium-40 were determined. The results obtained showed that the average concentration of radioactivity of radium-226, thorium-232 and potassium-40 is 3.398 Bqkg⁻¹, 4.667 Bqkg⁻¹ and 216.738 Bqkg⁻¹, respectively, for onion and 2.808 Bqkg⁻¹, 3.524 Bqkg⁻¹, and 172.064 Bqkg⁻¹ for garlic. The results also showed that the average annual total effective dose of the three nuclides is 122.955 μSv·y⁻¹ for onion and 97.231 μSv·y⁻¹ for garlic. Other relevant risk parameters were also calculated, such as equivalent activity concentrations, absorbed dose, excess lifetime cancer risks, and other health risk parameters. One of the most important conclusions reached by this study is that the natural radioactive elements in onions and garlic do not pose a great danger to their consumers, especially those infected with the COVID-19. Because the concentrations of these radioactive elements do not exceed the permissible limits recommended by recognized scientific organizations and agencies such as International Commission on Radiological Protection (ICRP), United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), and World Health Organization (WHO).

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1. Introduction

Onion and garlic are among the most important plants that were widely used during the Corona pandemic by people with the disease or for prevention. The use of these plants to prevent diseases is a popular tradition with a long reach in this region, due to the effectiveness of these medicinal plants in treating some diseases. This is why many people resorted to eating these plants in abundance after the outbreak of the Covid-19 disease. Natural radionuclides, which are abbreviate by the word “NORM”, are of real importance in the food that humans eat or the animals and plants they feed on. The importance of these nuclides appears when their concentrations in these substances exceed the permissible limits, which entails great risks to human health [1]. In addition, an increase in consuming these vegetables leads to increasing the concentration of these radionuclides in the human body, and this poses a danger. From this point of view, the current study came to determine the concentrations of natural radionuclides (90232Th, 88226Ra, 1940K) after the significant increase in the consumption of these vegetables by the public after the outbreak of the Corona pandemic. These vegetables are important and cannot be easily dispense with, so studying them is a necessity for their continued use by the population of the area covered by the study.
2. Assessment of radiation hazards

2.1. Minimum Detectable activity

The Minimum Detectable Activity (MDA) is very important if low-concentration radioactive elements such as NORM are detected. The sample count rate is often the same as the radiation background count. Radiation background without the sample should be measured with the same measurement conditions and preferably at the same time Measurement for sample. (MDA) depends on the detection limit level (LLD) and the counting efficiency of the detection system [2]. The LLD detection limit level of the detector system can be calculated from the following equation:

\[
LLD = (4.66x\sigma_b) + 3
\]

The minimum effectiveness of MDA detection can be calculated from the following equation:

\[
MDA = \frac{LLD}{k t}
\]

Or as follows:

\[
MDA = \frac{(4.66x\sigma_b) + 3}{k t}
\]

Where \(\sigma_b\) is the standard deviation of the radiation background and \(t\) is the measurement time of the radiation background and the sample \(k\) is a coefficient that contains both the efficiency of the detection system and the abundance of the element under measurement and the weight of the sample according to the following formula:

\[
k = \frac{\epsilon(E_i) I_x(E_i)}{W}
\]

Where \(W\) is the weight of the sample measured in Kg. Equation (3) can be redrafted as follows:

\[
MDA = \frac{(4.66x\sigma_b) + 3}{\epsilon(E_i) I_x(E_i)} W t
\]

Equation (1) and (3) are valid for use only when the sample and radiation background time is equal and otherwise the following general equations are used:

\[
LLD = 3.29 \sqrt{n_b t_b (1 + \frac{t_b}{t_b})} + 3
\]

\[
MDA = 3.29 \sqrt{n_b t_b (1 + \frac{t_b}{t_b})} + 3
\]

Where \(n_b\) is the rate of the detection of the radiation background detection for the time period \(t_b\) and \(t_i\) the total time of the sample [2,3].

2.2. Radioactivity concentration

The concentration of the specific radiation activity is defined as the activity of each unit of mass of the radioactive material and, measured in Curies per gram or Bq/Kg. The activity concentration \(A\) for each radioactive element in Bq / kg can be calculated using the following equation [3]:

\[
A(Bq/Kg) = \frac{N}{t_i(E_i) \epsilon(E_i) m}
\]

Where \(N\) is the net area under the gamma-ray peak measured for the spectrum after subtraction of the radiation background, \(t\) measurement time (sec), \(I(E_i)\) intensity of measured gamma ray energy \(E_i\), \(\epsilon(E_i)\) is the efficiency of gamma ray energy line and \(m\) is the weight of the sample Kg.

2.3. Radium equivalent activity (Ra\textsubscript{eq})

The equivalent concentration value of the radium element (Ra\textsubscript{eq}) used to estimate the hazards associated with substances containing radium-226, thorium-232 and potassium-40 radionuclides, calculated to assume a concentration of 370 Bq / kg for radium 226 in this substance or 260 Bq / kg for thorium – 232 or 4810 Bq / Kg of potassium-40 which produces the same dose for gamma rays. The equivalent radium efficiency (Ra\textsubscript{eq}) can be calculated using the following equation [4]:

\[
Ra_{eq}(Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.077A_{K}
\]

Where \(A_{Ra}\), \(A_{Th}\) and, \(A_{K}\) are the efficiencies of radium, thorium and potassium, respectively, and measured by Bq/Kg. This indicator can be circulated on both potassium and thorium according to the following equations:

\[
Th_{eq}(Bq/kg) = A_{Th} + 0.7A_{Ra} + 0.055A_{K}
\]

\[
K_{eq}(Bq/kg) = A_{K} + 18.46A_{Th} + 13.24A_{Ra}
\]

2.4. The external hazard index (\(H_{ex}\))

This term used to determine the external risk of gamma rays and to estimate the expected gamma dose that may be expose to external agents when they deal with substances containing gamma rays. The objective of this factor is to ensure that the effective dose of this radiation does not exceed permissible limits. The external risk factor can be calculate using the following equation:

\[
H_{ex} = \frac{A_{Ra} + A_{Th} + A_{K}}{370 + 259 + 4810} \leq 1
\]

Where \(H_{ex}\) is the external risk factor, \(A_{Ra}\), \(A_{Th}\) and, \(A_{K}\) are the radioactivity concentration of radium-226, thorium-232 and potassium-40, respectively, measured by Bq/Kg [5].

2.5. The internal hazard index (\(H_{in}\))

The internal risk factor determines the dose limits received by workers in fields containing normal radiation activity, which reached the workers by swallowing or inhaling. The internal risk factor is a measure of radiation dose control and, given by the following formula:

\[
H_{in} = \frac{A_{Ra} + A_{Th} + A_{K}}{185 + 259 + 4810} \leq 1
\]

Where \(H_{in}\) is the external risk factor, \(A_{Ra}\), \(A_{Th}\) and, \(A_{K}\) are the radioactivity concentrations of radium activity in (Bq / Kg) for radium-226, thorium-232 and potassium-40, respectively, where internal risk factor values should be less than one in the ideal environment to get a proper job opportunity for respiratory organs because they have dangerous respiratory effects [6].

2.6. Absorbed gamma ray dose (\(D_{g}\))

The absorbed dose is the absorbed energy in the mass unit of the body exposed to radiation. This term used for all types of radiation, energies, and all objects and materials. The rates of the absorbed doses due to gamma ray radiation of a naturally occurring radionuclide (\(^{226}Ra\), \(^{232}Th\), \(^{40}K\)) calculated based on the recommendations of ICRP [mGy / h] using the following equation[7]:

\[
D_{g} = \frac{N}{\epsilon(E_i) \times m}
\]
The conversion factors used to calculate the absorption rate of gamma rays for each radioactivity concentration (1 Bq/Kg) are for radium-226 (0.462 nGy/h) and (0.604 nGy/h) for thorium-232 and (0.0417 nGy/h) for potassium-40. In addition, the absorbed dose can be calculated using the relationship derived by Beck [8]:

$$D_{(\text{Beck})} = 0.420\text{A}_{\text{Ra}} + 0.429\text{A}_{\text{Th}} + 0.666\text{A}_{\text{K}}$$  \hspace{1cm} (9)

And according to the formula adopted by UNSCEAR [9],

$$D_{(\text{UNSCEAR})} = 0.533\text{A}_{\text{Ra}} + 0.827\text{A}_{\text{Th}} + 0.0537\text{A}_{\text{K}}$$  \hspace{1cm} (10)

2.7. Representative level index ($I_{\text{rep}}$)

It is used to estimate the level of gamma rays radiation risk associated with natural radionuclides in the measured samples, a factor representing the OECD index could be calculated from the following equation derived by the OECD [10]:

$$I_{\text{R(OECD)}} = \frac{\text{A}_{\text{Ra}}}{150} + \frac{\text{A}_{\text{Th}}}{100} + \frac{\text{A}_{\text{K}}}{1500}$$  \hspace{1cm} (11)

Where the radioactivity concentration of radium-226 ($\text{A}_{\text{Ra}}$), thorium-232($\text{A}_{\text{Th}}$) and potassium-40 ($\text{A}_{\text{K}}$), respectively are in Bq/Kg.

2.8. The annual effective dose (AED)

To calculate the annual effective dose, consider the conversion factor from the absorbed dose to the effective dose and the internal survival factor. To calculate the effective dose of the gammas emitting element, UNSCEAR2000 [9] has adopted the conversion coefficient of 0.7 Sv / Gy as a conversion factor from the absorbed dose in air to the annual effective dose received by adults. The calculations adopted that 80% of the person lifetime spent in dwelling and 20% of time spent abroad. From these data, the annual effective dose calculated as follows:

$$\text{AED}_{\text{air}}(\text{mSv}/\text{yr}) = D_{\text{a}}(\text{nGy}/\text{h}) \times 10^{-6} \times 8760 \times 0.8 \times 0.75 \text{Sv/G}$$  \hspace{1cm} (12)

$$\text{AED}_{\text{int}}(\text{mSv}/\text{yr}) = D_{\text{i}}(\text{nGy}/\text{h}) \times 10^{-6} \times 8760 \times 0.2 \times 0.75 \text{Sv/G}$$  \hspace{1cm} (13)

Where the number (8760) is the number of hours per year [11].

Table 1

| Source of Sample | Sample ID | Common Name | Scientific Name |
|-----------------|-----------|-------------|-----------------|
| Iraq            | OIQ1      | Onion       | Allium cepa     |
| Iran            | OR1       | Onion       | Allium cepa     |
| Egypt           | OEG       | Onion       | Allium cepa     |
| Turkey          | OTR       | Onion       | Allium cepa     |
| Iraq            | OIQ2      | Onion       | Allium cepa     |
| Iraq            | GQ        | Garlic      | Allium sativum  |
| Iran            | GIR       | Garlic      | Allium sativum  |
| China           | GCN       | Garlic      | Allium sativum  |
| Egypt           | GEG       | Garlic      | Allium sativum  |

2.9. Excess lifetime cancer risk (ELCR)

It is a factor used to calculate the risk of gamma ray associated to radionuclides in the studied samples. It gives the percentage of those who develop cancer because of the annual effective doses received. ELCR calculated as follows:

$$\text{ELCR} = \text{AED} \times \text{DL} \times \text{RF}$$  \hspace{1cm} (14)

Since AED is the annual effective dose, DL is the expected life expectancy of approximately 70 years and RF is the risk of fatal injury per Sievert and is equal to 0.05 for the public according to ICRP [7].

2.10. Annual effective dose and dose rate from ingested Onion and Garlic

The annual effective dose of Onion and Garlic consumption can be calculate for the possibility of collecting various radionuclides that may come from different sources of radiation. These doses can be measured by measuring the concentration of activity (Bqkg$^{-1}$) of radionuclides in Onion and Garlic, then multiplying them in mass of these Onion and Garlic consumed within a given time frame (kg / day or kg / yr.). The dose transfer factor (Sv / Bq) Given to each radionuclide as representative by the following equation: [11]:

$$\text{IAED} = \sum(A_{i}, W_{i}, DCF)$$  \hspace{1cm} (15)

$\text{IAED}$ is the ingestion annual effective dose of Onion and Garlic (Sv/yr.) and $A_{i}$ is the concentration of radionuclide activity in the sample (Bq / Kg). $W_{i}$ is the annual amount of onion and garlic consume by an adult during a year measured in (kg / yr.). The amount of adult intake in the year was 36.5 kg at 100 g per day. $DCF$ is the conversion factor of the intestinal tract through the ingestion of radionuclides (Sv / Bq), where for Radium-226 (0.28 µSv / Bq), and for Thorium-232 (0.23 µSv / Bq) and for potassium-40 (0.0062 µSv / Bq) [12].

Table 4

| Sample ID | Th$_{eq}$ | Ra$_{eq}$ | K$_{eq}$ |
|-----------|-----------|-----------|-----------|
| OIQ1      | 16.805    | 23.684    | 308.003   |
| OIR       | 19.672    | 27.745    | 360.839   |
| OEG       | 20.372    | 28.731    | 373.364   |
| OTR       | 17.963    | 25.345    | 329.835   |
| OIQ2      | 16.020    | 22.579    | 293.536   |
| Average   | 18.166    | 25.616    | 333.115   |
| STDEV     | 1.846     | 2.610     | 33.898    |
| GQ        | 12.297    | 17.336    | 225.381   |
| GIR       | 15.063    | 21.271    | 276.797   |
| GCN       | 16.644    | 23.495    | 305.421   |
| GEG       | 15.813    | 22.290    | 289.666   |
| Average   | 14.954    | 21.098    | 274.316   |
| STDEV     | 1.885     | 2.667     | 34.659    |

Table 3

Minimum detection activity (MDA) of measurement system used to determine the radioactivity concentrations of targeted Elements in Salt Samples.

| Nuclide | $E_{c}$ (KeV) | $I_{c}$ (%) | $\varepsilon(E_{c})$ | MDA (Bq/kg) |
|---------|---------------|-------------|----------------------|-------------|
| $^{90}$Th | 8120871       | 583         | 84                   | 0.1198      |
| $^{226}$Ra | 8321481      | 609.318     | 46                   | 0.1069      |
| $^{140}$K(Natural) | 1460.83 | 10.7        | 10.7                 | 11.070      |
3. Material and methods

3.1. Samples preparation

Five samples of onions and four samples of garlic grouped according to the origin. The samples collected from the market of the study area. To remove moisture, the samples dried in an electric oven at 80°C for 24 h [5,6]. After drying; the samples were standard size Marinelli beaker for each sample. The samples tightly sealed and stored for one month to obtain an acceptable radiative equilibrium for the natural radioactive elements targeted in this study. Table 1 show the collected samples according to the origin.

Table 5
The equivalent concentrations of $^{232}$Th, $^{226}$Ra and $^{40}$K in Bq kg$^{-1}$.

| Sample ID | $A$(Th-232) | $A$(Ra-226) | $A$(K-40) |
|-----------|--------------|--------------|-----------|
| OIQ1      | 3.181        | 3.076        | 208.558   |
| OIR       | 5.226        | 3.869        | 231.592   |
| OEG       | 4.969        | 3.101        | 240.586   |
| OTR       | 6.68         | 4.324        | 204.649   |
| OIQ2      | 3.279        | 2.620        | 198.309   |
| Average   | 4.667        | 3.398        | 216.738   |
| STDEV     | 1.465        | 0.684        | 18.3170   |
| GIQ       | 2.607        | 2.106        | 149.360   |
| GIR       | 3.360        | 3.761        | 161.294   |
| GCN       | 4.416        | 3.086        | 183.037   |
| GEG       | 3.515        | 2.282        | 194.567   |
| Average   | 3.524        | 2.808        | 172.064   |
| STDEV     | 0.738        | 0.764        | 20.4793   |

Table 6
Gamma ray absorption dose (D) calculated according to ICRP60 and, UNSCEAR.

| Sample ID | $D_{ICRP}$ (nGy h$^{-1}$) | $D_{UNSCEAR}$ (nGy h$^{-1}$) | $AED_{ICRP}$ (μSv y$^{-1}$) | $ELCR \times 10^{-4}$ | $I_{nu}$ Bq kg$^{-1}$ | $H_{nu}$ Bq kg$^{-1}$ | $H_{nu}$ Bq kg$^{-1}$ |
|-----------|---------------------------|-----------------------------|----------------------------|----------------------|---------------------|---------------------|---------------------|
| OIQ1      | 12.387                    | 15.469                      | 15.191                     | 5.317                | 0.191               | 0.063               | 0.072               |
| OIR       | 15.070                    | 18.820                      | 18.480                     | 6.468                | 0.232               | 0.078               | 0.086               |
| OEG       | 14.958                    | 18.681                      | 18.345                     | 6.420                | 0.232               | 0.080               | 0.092               |
| OTR       | 15.068                    | 18.818                      | 18.480                     | 6.467                | 0.232               | 0.080               | 0.092               |
| OIQ2      | 11.816                    | 14.757                      | 14.492                     | 5.072                | 0.182               | 0.060               | 0.068               |
| Average   | 13.860                    | 17.309                      | 16.998                     | 5.949                | 0.213               | 0.072               | 0.081               |
| STDEV     | 1.618                     | 2.021                       | 1.984                      | 0.694                | 0.024               | 0.009               | 0.010               |
| GIQ       | 9.047                     | 11.299                      | 11.096                     | 3.883                | 0.139               | 0.046               | 0.052               |
| GIR       | 10.898                    | 13.610                      | 13.365                     | 4.678                | 0.168               | 0.057               | 0.067               |
| GCN       | 12.111                    | 15.125                      | 14.853                     | 5.198                | 0.186               | 0.063               | 0.071               |
| GEG       | 11.667                    | 14.571                      | 14.309                     | 5.008                | 0.180               | 0.060               | 0.066               |
| Average   | 10.931                    | 13.651                      | 13.406                     | 4.692                | 0.168               | 0.056               | 0.064               |
| STDEV     | 1.352                     | 1.688                       | 1.658                      | 0.580                | 0.020               | 0.007               | 0.008               |

Table 7
The ingestion annual effective dose (IAED) and total ingestion annual Effective dose (TIAED) of onion and garlic intake in μSv y$^{-1}$.

| Sample ID | IAED (Th-232) | IAED (Ra-226) | IAED (K-40) | TIAED (Total) |
|-----------|---------------|---------------|--------------|---------------|
| OIQ1      | 26.704        | 31.436        | 47.196       | 105.337       |
| OIR       | 43.87         | 39.541        | 52.409       | 135.822       |
| OEG       | 41.714        | 31.692        | 54.444       | 127.85        |
| OTR       | 56.078        | 44.191        | 46.312       | 146.581       |
| OIQ2      | 27.527        | 26.776        | 44.877       | 99.180        |
| Average   | 39.179        | 34.727        | 49.047       | 122.955       |
| STDEV     | 12.302        | 9.699         | 4.145        | 20.145        |
| GIQ       | 21.885        | 21.523        | 33.800       | 77.209        |
| GIR       | 29.886        | 38.437        | 36.500       | 104.824       |
| GCN       | 37.072        | 31.538        | 41.421       | 110.032       |
| GEG       | 29.508        | 23.322        | 44.030       | 96.860        |
| Average   | 29.588        | 28.705        | 38.938       | 97.231        |
| STDEV     | 6.203         | 7.816         | 4.634        | 14.045        |

Table 8
Natural Radioactivity Concentrations of studied elements in the current Research compared to their concentrations in Research and global studies.

| Country     | Sample       | Activity concentration (Bq kg$^{-1}$) |
|-------------|--------------|--------------------------------------|
| Iraq/Najaf  | Onion        | 3.08                                  |
|             | Iran Onion   | 2.84                                  |
|             | Garlic       | 3.55                                  |
| Bangladesh  | Vegetables   | 83.53                                 |
|             | Onion        | –                                     |
|             | Garlic       | 0.038                                 |
|             | Vegetables   | 0.64                                  |
|             | Vegetables   | 2.37–7.2                              |
|             | Vegetables   | 57.7                                  |
|             | Vegetables   | 18.1                                  |
|             | Vegetables   | 3.38                                  |
|             | Onion        | 3.18–6.68                             |
|             | Garlic       | 2.6–4.41                              |

Note: All the values are in Bq kg$^{-1}$. The concentration values are given as range where applicable.
3.2. Samples measurement

A Teledyne isotope NaI (TI) scintillation detector with resolution 7.5% KeV at the 661.76 KeV Cs-137 source was used. The detector was shielded with a low-level background lead shield. The NaI (TI) system was calibrated using two reference materials that are Thorium oxide (ThO2-S7) from British laboratory equipment company PANAX. The certified activity of Thorium is 3570 ± 20 Bq/kg. The energy transitions of the 232Th daughters (300, 338.4, 762.5 KeV) were used to determine the efficiency calibration curve. The Minimum Detectable Activity (MDA) was calculated in the present study using Eq. (3) as shown in Table 3.

4. Results and discussion

The values of measured activity concentrations of selected radionuclides of 232Th, 226Ra and 40K in Onion and Garlic samples shown in Table 4. The average concentration of Thorium-232, Radium-226 and Potassium-40 for onion are 4.687 Bq/kg, 3.398 Bq/kg and 216.74 Bq/kg, respectively, and for garlic are 3.5245 Bq/kg, 2.808 Bq/kg and 172.06 Bq/kg respectively. The equivalent concentrations for each of Thorium-232 (Th_eq), Radium –226 (Ra_eq) and Potassium-40 (K_eq) for onion and garlic samples shown in Table 5.

The absorbed dose for these concentrations were calculated using two equations supported by ICRP and, UNSCEAR and the annual effective dose rate (AED), excess lifetime cancer risks (ELCR), external and internal hazard index are shown in Table 6. The total ingestion annual effective dose (total dose of all target elements in the study) (TIAED) resulting from the consumption of onion and garlic which calculated using equation (15) [16], are between (99.18–146.58) μSv y⁻¹ with average (122.95) μSv y⁻¹ for onion and, (77.20–110.03) μSv y⁻¹ with average (97.23) μSv y⁻¹ for garlic as shown in Table 7. The measured activity concentrations of 232Th, 226Ra and 40K were compare with worldwide reported values as shown in Table 8.

5. Conclusion

The present study has been carried out to establish baseline data regarding concentration levels of naturally occurring radionuclides of 232Th, 226Ra, and 40K in onion and garlic and the corresponding radiation doses in the province of Thi Qar. One of the most important conclusions reached by this study is that the natural radioactive elements in onions and garlic do not pose a great danger to their consumers, especially those infected with the Coronavirus pandemic. Because the concentrations of these radioactive elements do not exceed the permissible limits recommended by recognized scientific organizations and agencies such as the International Commission on Radiological Protection (ICRP), United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), and World Health Organization (WHO). Although the consumed quantity raised the value of the effective annual dose and, the value of radium equivalent in the bodies of infected consumers, they are still within the permissible limits. The main fear of consuming large quantities of these vegetables was that this consumption would cause a rise in the level of natural radiation, which would negatively affect the health of the infected person and thus increase the effectiveness of the virus. Calculated values of hazard coefficients are also lower than the world average of about 0.5 mSv per year. It has concluded that there is no potential radiological health risk associated with the onion and garlic samples investigated during this work.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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