Radon and Thoron Concentrations Measurement in Tea Samples Consumed in Iraqi Markets Using CR-39 Detector

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Abstract. Tea is one of the most food used in the form of cold and hot drinks in Iraq, so 12 samples of the most frequently traded teas have been collected on the market, where Radon and thoron levels were measured using CR-39 nuclear track detector, and exhalation rates of radon and thoron gas were calculated. The results show that the highest concentration of radon is in sample T-4 and the highest concentration of thoron is in sample T-5 and were 34.725 ± 13.2 Bq/m³ and 10.164 Bq/m³ respectively. The obtained radon levels is within the natural limits of the activity concentrations of the naturally occurring radionuclides in foods, and this will not form any risk on human health.

Keywords. CR-39, Radon, Thoron, Tea, Activity Concentration

1. Introduction
There has been an increasing concern about exposure to Radon (²²²Rn), Thoron (²²⁰Rn) and their short lived decay products are recognized as the most important contributors to committed effective dose received by population due to the natural sources [1]. The environmental radon concentration is a function of time and climate conditions. Owing to this fact, radon is the most popular subject of studies on environmental radioactivity [2]. The presence of high level of radon in indoor environment constitutes a major health hazard for man [3]. The radon and thoron progeny are well established as causative agents of lung cancer and other types of cancers. Radon’s unique properties as a naturally radioactive gas have led to its use as a geophysical tracer for locating buried faults and geological structures, in exploring for uranium, and for predicting earthquakes [4].

The radon arising in the soil partly reaches the surface and is released into the atmosphere. The exhalation rate strongly depends on the permeability of the soil and weather conditions (temperature, humidity, air pressure and soil permeability). Radon can also be dissolved in ground water and migrates in bedrock. The techniques for the collection and measurement of ²²²Rn in air can be subdivided into active (requiring power to collect a sample) and passive (requiring no power) methods. Active methods are usually used for short-term measurements of radon. Passive methods are more
suitable for the assessment of radon exposure over long time scales and can be used for large scale surveys at moderate cost. Solid state nuclear track detectors, known as passive method, are widely used for radon measurements [5, 6 and 7].

2. Materials and Methods

2.1. Collection and preparation of samples
A total of 12 samples of tea were collected from local Iraqi markets which are mostly consumed by people. The samples were dried, grinded and sifted with 650-micron slot diameter sieve to get a homogeneous powder. The samples were placed in cans and left 7 cm from the top of the can. As well as the track detectors (CR-39) were pasted to the inner surface of the lids. The cans were sealed and encoded, and then stored for 60 days to attain radioactive secular equilibrium. Table 1 shows the sample codes and country of origin.

| Code | Name of the Product | Packing | Origin | Production date | Expiration date |
|------|---------------------|---------|--------|-----------------|-----------------|
| T-1  | Ahmed               | Emirates| UK     | 2018            | 2021            |
| T-2  | Alwazah             | Sri Lanka| South of Sri Lanka | 2018 | 2021 |
| T-3  | Al-Ghazaleen        | Sri Lanka| Ceylon Mountains | 2016 | 2019 |
| T-4  | Cihan               | Sri Lanka| India  | 2018            | 2021            |
| T-5  | Mahmood             | Sri Lanka| India  | 2018            | 2021            |
| T-6  | Budgerigar          | Sri Lanka| India  | 2016            | 2019            |
| T-7  | Apple               | Sri Lanka| India  | 2017            | 2020            |
| T-8  | Lipton              | Sri Lanka| India-Kenya | 2018 | 2021 |
| T-9  | 272                 | Sri Lanka| Sri Lanka | 2018 | 2021 |
| T-10 | Ahmed Green tea     | Emirates| China  | 2018            | 2021            |
| T-11 | Al-Wazah Green tea  | Sri Lanka| South of Sri Lanka | 2018 | 2021 |
| T-12 | Mahmood Green tea   | Sri Lanka| India  | 2017            | 2020            |

2.2. Radioactivity measurements
The CR-39 nuclear track detector was used to detect the natural radioactivity of $^{222}$Rn and $^{220}$Rn in the tea samples. The CR-39 detector records the path of alpha particles. The chemical engraving process to the CR-39 detectors has been done to show the fission tracks. In this process, NaOH solution (sodium hydroxide) is used as an etchant solution, in normal mode 6.25N and temperature 80°C. After 5 hours, the detector was removed and washed with distilled water, then calculated the number of alpha traces using microscopic viewing process with a camera of 5 MPixels to estimate the track density.

2.3. Calculation of Radon Exposure

2.3.1. Radon concentration measurement
Radon gas ($^{222}\text{Rn}$) concentration in the samples has been measured by comparison between track densities register on the detector around the unknown sample and that of the standard calibration source. It can be calculated from the following relation [8]:

$$C_{\text{Rn}} = \frac{\rho_{Rn}}{k} \times T$$  \hspace{1cm} (1)

$$k = \frac{\rho_s}{E_s}$$  \hspace{1cm} (2)

Where; $C_{\text{Rn}}$ is the radon gas concentration in the unknown sample; $\rho_{Rn}$ is the track density of the unknown sample (track/mm$^2$); and $k$ is the slope of calibration curve. $\rho_s$ is the track density of standard source (tracks/mm$^2$); $E_s$ is the Radon exposure (Bq.day/m$^3$); and $T$ is the exposure time (day).

### 2.3.2. Determination of radon exhalation rate in samples

The amount of radon gas flow from the surface of a sample is defined as the exhalation rate of radon from the sample. The surface and mass exhalation rate ($E_s$, $E_m$) in (Bqm$^{-2}$h$^{-1}$) and (Bqkg$^{-1}$h$^{-1}$) units respectively can be calculated as follows [9]:

$$E_s = \frac{CV\lambda}{AT + 1/\lambda(e^{-\lambda T} - 1)}$$  \hspace{1cm} (3)

$$E_m = \frac{CV\lambda}{mT + 1/\lambda(e^{-\lambda T} - 1)}$$  \hspace{1cm} (4)

Where $C$ is the integrated Radon exposure (Bq.h.m$^{-3}$); $V$ is the volume of air in can (m$^3$); $\lambda$ is the decay constant for $^{222}\text{Rn}$ (h$^{-1}$). $A$ is the surface area of the sample (m$^2$); $m$ is mass of sample. $T$ is the exposure time (h).

### 2.3.3. Thoron Concentrations and Exhalation Rates

For the purpose of measuring the alpha emissions of both radon and thoron, a CR-39 detector was placed under each sample in the same cans. It has also been exposed to the same period of time as the detector hanging in the lid of each can. Then the processes of chemical etching and calculation have been proceeding in the same way and the same conditions.

With this technique of measurements, the concentration of thoron was calculated by subtracting the top track concentration from the bottom track for each of the samples studied.

The results showed that there was a difference in the concentration of the thoron from one sample to another, but in all cases, the concentration of thoron in all samples is less than the concentration of radon, no doubt that it has a very short half-life.

### 2.3.4. Dissolve Radon Concentration

The dissolve radon concentration in tea was calculated using the following equation [10]:

$$C_{\text{diss}} = \frac{A \lambda t}{L}$$  \hspace{1cm} (5)

Where, $C_{\text{diss}}$ is radon concentration in ambient air (Bq/m$^3$), $\lambda$ is the decay constant for radon = 0.1814 day$^{-1}$ [11], $h$ is the distance from the surface of tea sample to the detector, $T$ is the time of exposing, and $L$ is the depth of the sample.

### 2.4. Calibration of CR-39 Detector

The standard source $^{226}\text{Ra}$ with radioactivity ($A = 1.3$ μCi = 48100 Bq) manufactured in 1989 was used to calibrate the CR-39 detector. The CR-39 detector and the standard source of Radium $^{226}\text{Ra}$ were placed at the special can with same specification of the can used for samples.

The activity of Radon gas ($A_{\text{R}}$) inside the can at any time can be calculated by using the following equation [8]:

$$A_{\text{R}} = A_{Ra} \times (1 - \text{Exp} (-\lambda_{Ra} \times t))$$  \hspace{1cm} (6)

Where $t_{1/2}$ ($^{222}\text{Rn}$) = 3.8253 day; $A_{Ra} = 47333$Bq is the activity of standard source $^{226}\text{Ra}$ manufactured in 2019; $\lambda_{Ra}$ is the decay constant of $^{222}\text{Rn}$ = 0.1812 day$^{-1}$; and $t$ is the build-up time in day.
Hence, the Radon activity may be estimated inside the container after 1, 2, 3, 4 and 5 days. The radon exposure was also calculated for these different times which can be determined by the following equation [8]:

\[ E_s \text{ (Bq.day/m}^3\text{)} = \left( \frac{A_{Rn} \text{ (Bq)}}{V \text{ (m}^3\text{)}} \right) \times t \text{ (day)} \]  

(7)

Where \( E_s \) is the Radon gas exposure (i.e. concentration) of standard source (Bq/m\(^3\)) \( \times \text{days} = \text{(Bq/m}^3\text{)} \) by multiplying with (1, 2, 3, 4 and 5 days); \( A_{Rn} \) is the radioactivity of \(^{222}\text{Rn} \) calculated by equation (6); \( V \) is the can volume in m\(^3\); and \( t \) is the exposure time in day.

Figure (1) shows the relation between the track density (\( \rho \)) and the Radon exposure (\( E_s \)). The slope is \( k = \rho/E_s = 0.1488x+245.53 \text{ (track/mm}^2\text{)/(Bq.day/m}^3\text{)} \).

![Figure 1. Calibration curves for Radon exposure.](image)

3. Results

3.1. Radon Concentrations and Exhalation Rates in Tea Samples

Radon concentrations were calculated for the tea samples and the results were disclosed in Table (2) and Figure (2). The radon concentration found in tea samples ranged from 18.131±3.1 Bq/m\(^3\) in the sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest concentration 34.725±13.2 Bq/m\(^3\) in the sample T-4 (China tea, Indian origin), with an average value 27.983±8.1 Bq/m\(^3\).

If the black tea samples were compared, the lowest concentration of radon was 20.205±7.5 Bq/m\(^3\) recorded in T-5 (Mahmood tea, Indian origin) sample in comparison with high concentration in T-4 sample. If the green tea samples were compared, the highest concentration was 31.614±13.3 Bq/m\(^3\) in sample T-10 (Ahmed-Green tea, China origin) compared to the lowest concentration of radon in T-11. It can be noticed that all types of tea under study had a concentration of radon in the allowable global limit 200-300 Bq/m\(^3\) which is recommended by ICRP, 2009 [12].

Also, Radon Exhalation Rate was calculated for both Surface exhalation rate and Mass exhalation rate for the studied tea samples. Table (2) and Figures (3) and (4) shows the behavior of the surface exhalation rate was higher than the mass exhalation rate. The surface exhalation rate varies from 0.020 \( \pm \) 0.007 Bq/m\(^2\).h in sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest value 0.039 \( \pm \) 0.014 Bq/m\(^2\).h in a sample T-4 (China tea, India origin). While the mass exhalation rate varies from 3.9\( \times \)10\(^{-5}\)\( \pm \)1.6\( \times \)10\(^{-5}\) Bq/kg.h in sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest value7.5\( \times \)10\(^{-5}\)\( \pm \)2.8\( \times \)10\(^{-5}\) Bq/kg.h in a sample T-4 (China tea, India origin). With an average values 0.031 \( \pm \) 0.009 Bq/m\(^2\).h, and 6.1\( \times \)10\(^{-5}\)\( \pm \)1.7\( \times \)10\(^{-5}\) Bq/kg.h respectively.
Table 2. Track Density, Radon gas concentration ($C_{Rn}$), surface exhalation rate ($E_s$), mass exhalation rate ($E_m$), and dissolved radon concentration $C_{diss}$ in tea samples.

| Code | Track Density (track/mm²) | $C_{Rn}$ (Bq/m³) | $E_s$ (Bq/m².h) | $E_m$ (Bq/kg.h) | $C_{diss}$ (Bq/m³) |
|------|---------------------------|-------------------|-----------------|-----------------|-------------------|
| T-1  | 546.296 ± 332.6           | 33.687 ± 9.7      | 0.037 ± 0.012   | 7.3×10⁻⁵ ± 2.1×10⁻⁵ | 641.6363 |
| T-2  | 501.851 ± 297.4           | 28.709 ± 5.8      | 0.032 ± 0.005   | 6.2×10⁻⁵ ± 1.3×10⁻⁵ | 546.8203 |
| T-3  | 481.430 ± 279.9           | 26.422 ± 3.8      | 0.029 ± 0.004   | 5.7×10⁻⁵ ± 1.2×10⁻⁵ | 503.2598 |
| T-4  | 555.556 ± 363.1           | 34.725 ± 13.2     | 0.039 ± 0.014   | 7.5×10⁻⁵ ± 2.8×10⁻⁵ | 661.4071 |
| T-5  | 425.925 ± 382.8           | 20.205 ± 7.5      | 0.023 ± 0.008   | 4.4×10⁻⁵ ± 1.6×10⁻⁵ | 384.8446 |
| T-6  | 507.407 ± 345.1           | 29.332 ± 11.1     | 0.032 ± 0.012   | 6.3×10⁻⁵ ± 2.4×10⁻⁵ | 558.6866 |
| T-7  | 518.532 ± 305.9           | 30.578 ± 6.7      | 0.034 ± 0.007   | 6.6×10⁻⁵ ± 1.5×10⁻⁵ | 582.4192 |
| T-8  | 490.740 ± 288.7           | 27.465 ± 4.8      | 0.031 ± 0.005   | 5.9×10⁻⁵ ± 1.1×10⁻⁵ | 523.1259 |
| T-9  | 537.037 ± 335.4           | 32.651 ± 10.1     | 0.036 ± 0.011   | 7.1×10⁻⁵ ± 2.2×10⁻⁵ | 621.9036 |
| T-10 | 527.778 ± 364.2           | 31.614 ± 13.3     | 0.035 ± 0.010   | 6.8×10⁻⁵ ± 2.9×10⁻⁵ | 602.1519 |
| T-11 | 407.408 ± 272.8           | 18.131 ± 3.1      | 0.020 ± 0.007   | 3.9×10⁻⁵ ± 1.6×10⁻⁵ | 345.3412 |
| T-12 | 444.524 ± 318.7           | 22.288 ± 8.2      | 0.025 ± 0.009   | 4.8×10⁻⁵ ± 2.3×10⁻⁵ | 424.5195 |
| Average | 495.373 ± 323.9 | 27.983 ± 8.1  | 0.031 ± 0.009 | 6.1×10⁻⁵ ± 1.7×10⁻⁵ | 533.0097 |

Limit

ICRP, 2009 [11]

200-300

Figure 2. Radon gas concentrations in tea samples.

Figure 3. Surface exhalation rate in tea samples, for radon.
3.2. Thoron concentrations and Exhalation Rates in Tea samples

Thoron concentrations were calculated for the tea samples and the results were explained in Table (3) and Figure (5). The thoron concentration found in tea samples ranged from 1.244 Bq/m$^3$ in the sample T-8 (Lipton tea, SriLanka-India origin) to the highest concentration 10.164 Bq/m$^3$ in the sample T-5 (Mahmood tea, SriLanka-India origin), with an average value 4.143 Bq/m$^3$.

If the black tea samples were compared, the comparison is just as above. If the green tea samples were compared, the highest thoron concentration was 6.223 Bq/m$^3$ in sample T-11 (Al-Wazah-Green tea, South of Sri Lanka origin) compared to the lowest concentration of thoron in T-10 (3.339Bq/m$^3$).

Also, Thoron Exhalation Rate was calculated for both Surface exhalation rate and Mass exhalation rate for the studied tea samples. Table (3) and Figures (6) and (7) show the behavior of the surface exhalation rate was higher than the mass exhalation rate. The surface exhalation rate varies from 0.001 Bq/m$^2$.h in sample T-8 (Lipton tea, SriLanka-India origin) to the highest value 0.011 Bq/m$^2$.h was in sample T-5 (Mahmood tea, SriLanka-India origin). While the mass exhalation rate varies from $2.7\times10^{-5}$ Bq/kg.h in sample T-8 to the highest value $21.9\times10^{-5}$ Bq/kg.h in a sample T-5. With an average values $0.005$ Bq/m$^2$.h and $9.1\times10^{-5}$ Bq/kg.h respectively.

In all cases, there is no need to worry, since all tea samples under study contain radon concentrations far below the universally permitted limits recommended by the ICRP, 2009 [12].

Table 3. Radon gas concentration at the bottom of the can (CRnB), Radon gas concentration at the top of the can (CRnT), Thoron gas concentration (CTh), surface exhalation rate (EA), and mass exhalation rate (Em).

| Code | CRnB (Bq/m$^3$) | CRnT (Bq/m$^3$) | CTh (Bq/m$^3$) | EA (Bq/m$^2$.h) | Em (Bq/kg.h) |
|------|----------------|----------------|----------------|----------------|--------------|
| T-1  | 35.139         | 33.687         | 1.452          | 0.002          | 3.1$\times10^{-5}$ |
| T-2  | 34.311         | 28.709         | 5.602          | 0.006          | 12.2$\times10^{-5}$ |
| T-3  | 29.124         | 26.422         | 2.702          | 0.003          | 5.8$\times10^{-5}$ |
| T-4  | 37.836         | 34.725         | 3.111          | 0.003          | 6.7$\times10^{-5}$ |
| T-5  | 30.369         | 20.205         | 10.164         | 0.011          | 21.9$\times10^{-5}$ |
| T-6  | 33.687         | 29.332         | 4.355          | 0.005          | 9.4$\times10^{-5}$ |
| T-7  | 32.984         | 30.578         | 2.406          | 0.002          | 5.2$\times10^{-5}$ |
| T-8  | 28.709         | 27.465         | 1.244          | 0.001          | 2.7$\times10^{-5}$ |
| T-9  | 36.799         | 32.651         | 4.148          | 0.004          | 8.9$\times10^{-5}$ |
| T-10 | 34.953         | 31.614         | 3.339          | 0.003          | 7.2$\times10^{-5}$ |
| T-11 | 24.354         | 18.131         | 6.223          | 0.007          | 13.4$\times10^{-5}$ |
| T-12 | 27.257         | 22.288         | 4.969          | 0.006          | 10.7$\times10^{-5}$ |
Average 35.139  27.98392  4.143  0.005  9.1×10^{-5}

In any case, the concentration of thoron has a very short half-life. All milk under study is suitable for use because the radon concentrations in all samples were lower than the recommended limit recorded by ICRP, 2009 [12].

Despite occupational exposure to the thoron, there is no data available for reference in estimating its health damage.

As explained earlier when discussing the damage of radon-222 through the concept of working level, the energy of the alpha particles absorbed by the lung tissue needs to be calculated.
In the case of radon, the primary exposure is due to the emission of alpha particles from isotopes Po-218 and Po-214 and their stability in lung tissue because the half-life of these isotopes is very short (3 minutes for Po-218 and 26.8 minutes for Po-214 [13]). In the case of the thoron, the primary isotope of its daughters is Pb-212, which decay to Bi-212 and then to Po-212, which releases an alpha particle of 8.78 MeV into a radioactive stable isotope of Pb-208. However, the half-life of lead-212 of 10.6 hours [13] is very long compared to the half-life of polonium-218 and polonium-214 measured in minutes. Hence, the bulk of lead-212 will be removed from the lungs prior to its dissolution, and therefore the role of the thoron can be neglected compared to that of radon in general.

3.3. Dissolved Radon Concentration
Dissolved radon concentrations were calculated for the tea samples and the results were disclosed in Table (2) and Figure (8). The dissolve radon concentration found in tea samples ranged from 345.3412 Bq/m³ in the sample T-11 (Al-Wazah Green tea, South of Sri Lanka) to the highest concentration 661.4071 Bq/m³ in the sample T-4 (China tea, India origin), with an average value 533.0097 Bq/m³.

![Figure 8. Dissolved radon concentration in tea samples.](image)

4. Conclusions
Radon concentration results of the best black tea are Mahmood tea and the best green tea is Al-Wazah Green Tea, because they had the lowest radon concentration compared to the rest of the samples under study.

Thoron concentrations results although, Lipton tea has lower concentration of thoron, but we cannot consider it the best black tea because the concentration of radon is high compared to the concentrations of the rest of the tea samples. Therefore, our consideration on the adoption of Mahmoud tea is the best black tea. For the same explanation, Alwazah Green tea remains the best compared to other green tea samples for study.

The value of radon content in different tea samples is due to the naturally occurring of uranium isotopes in soil. It can be noted that the obtained values which range between 18.131 ± 3.1 and 34.725 ± 13.2 Bq/m³ are in general lower than the ICRP, 2009 values cited 200-300 Bq/m³ for activity concentrations of the naturally occurring radionuclides in foods.

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