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

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Abstract. Tea is one 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 cods and country of origin.

Table 1. Sample code, origin, packing and date of tea samples.

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 80C° . 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}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]:

$$C_{Rn} = \rho_{Rn} / k \times T \quad (1)$$

$$k = \rho_s / E_s \quad (2)$$

Where; C_{Rn} is the radon gas concentration in the unknown sample; ρ_{Rn} is the track density of the unknown sample (track/mm²); and k is the slope of calibration curve. ρ_s is the track density of standard source (tracks/mm²); E_s is the Radon exposure (Bq.day/m³); 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_A , E_m) in (Bqm⁻²h⁻¹) and (Bqkg⁻¹h⁻¹) units respectively can be calculated as follows [9]:

$$E_A = \frac{CV\lambda}{A[T + 1/\lambda(e^{-\lambda T} - 1)]} \quad (3)$$

$$E_m = \frac{CV\lambda}{m[T + 1/\lambda(e^{-\lambda T} - 1)]} \quad (4)$$

Where C is the integrated Radon exposure (Bq.h.m⁻³); V is the volume of air in can (m³); λ is the decay constant for ^{222}Rn (h⁻¹). A is the surface area of the sample (m²); 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]:

$$C_{diss} = C_{Rn} \frac{\lambda h T}{L} \quad (5)$$

Where, C_{Rn} is radon concentration in ambient air (Bq/m³), λ 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}Ra with radioactivity ($A = 1.3 \mu\text{Ci} = 48100 \text{ Bq}$) manufactured in 1989 was used to calibrate the CR-39 detector. The CR-39 detector and the standard source of Radium ^{226}Ra were placed at the special can with same specification of the can used for samples.

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

$$A_{Rn} = A_{Ra} \times (1 - \text{Exp}(-\lambda_{Rn} \times t)) \quad (6)$$

Where $t_{1/2} (^{222}\text{Rn}) = 3.8253 \text{ day}$; $A_{Ra} = 47333 \text{ Bq}$ is the activity of standard source ^{226}Ra manufactured in 2019; λ_{Rn} is the decay constant of $^{222}\text{Rn} = 0.1812 \text{ day}^{-1}$; 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]:

$$E_s (\text{Bq.day/m}^3) = [A_{\text{Rn}} (\text{Bq}) / V (\text{m}^3)] \times t (\text{day}) \quad (7)$$

Where E_s is the Radon gas exposure (i.e. concentration) of standard source (Bq/m^3).days = (Bq/m^3) by multiplying with (1, 2, 3, 4 and 5 days); A_{Rn} is the radioactivity of ^{222}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 (ρ_s) and the Radon exposure (E_s). The slope is $k = \rho_s/E_s = 0.1488x + 245.53$ (track/mm^2)/(Bq.day/m^3).

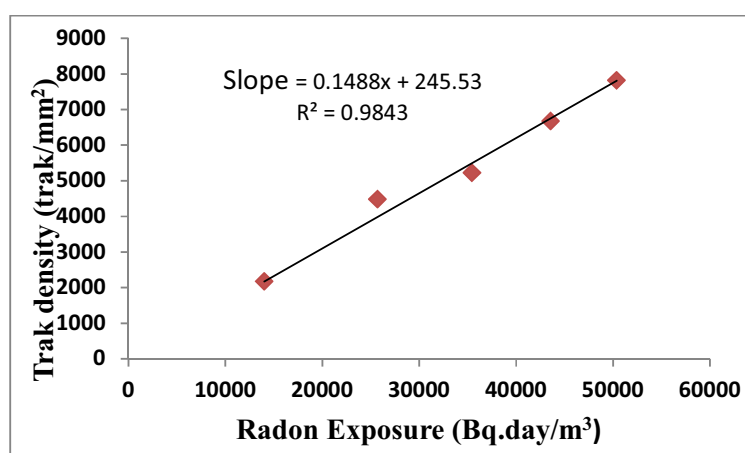


Figure 1. Calibration curves for Radon exposure.

3. Results

3.1. Radon Concentrations and Exhalation Rates in Tea Samples

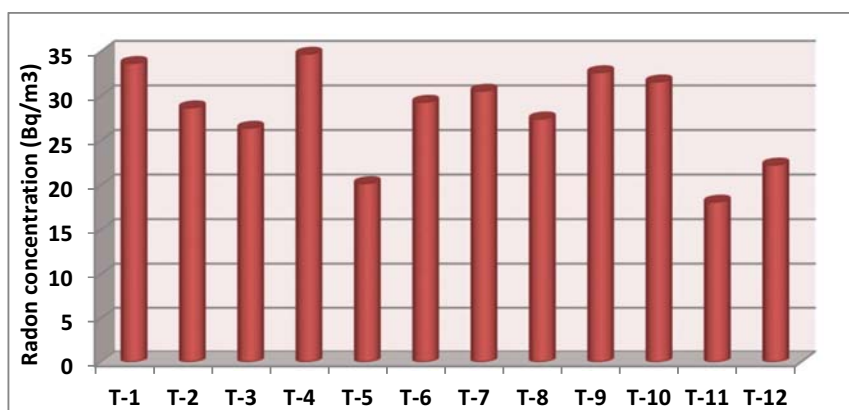
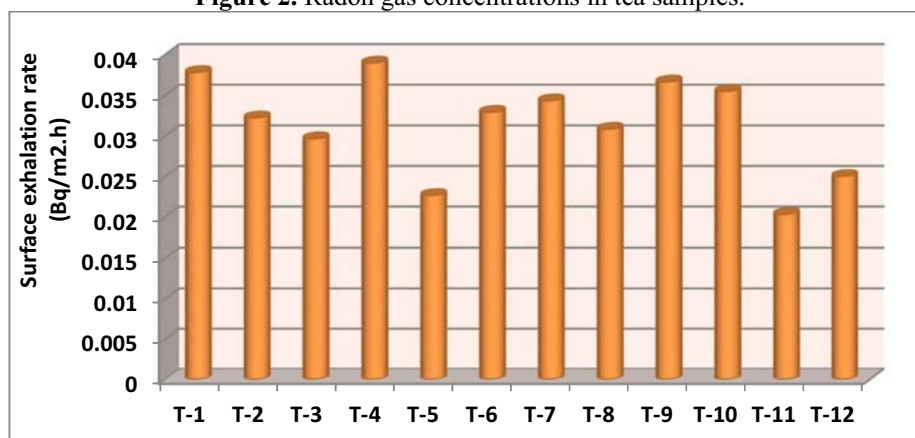
Radon concentrations were calculated for the tea samples and the results were disclosed in Table (2) and Figure (2). The radon concentration found in tea samples ranged from $18.131 \pm 3.1 \text{ Bq/m}^3$ in the sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest concentration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

If the black tea samples were compared, the lowest concentration of radon was $20.205 \pm 7.5 \text{ Bq/m}^3$ 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 $31.614 \pm 13.3 \text{ Bq/m}^3$ 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 $200\text{-}300 \text{ Bq/m}^3$ 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 $0.020 \pm 0.007 \text{ Bq/m}^2.\text{h}$ in sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest value $0.039 \pm 0.014 \text{ Bq/m}^2.\text{h}$ in a sample T-4 (China tea, India origin). While the mass exhalation rate varies from $3.9 \times 10^{-5} \pm 1.6 \times 10^{-5} \text{ Bq/kg.h}$ in sample T-11 (Al-Wazah- Green tea, South of Sri Lanka origin) to the highest value $7.5 \times 10^{-5} \pm 2.8 \times 10^{-5} \text{ Bq/kg.h}$ in a sample T-4 (China tea, India origin). With an average values $0.031 \pm 0.009 \text{ Bq/m}^2.\text{h}$, and $6.1 \times 10^{-5} \pm 1.7 \times 10^{-5} \text{ Bq/kg.h}$ respectively.

Table 2. Track Density, Radon gas concentration (C_{Rn}), surface exhalation rate (E_A), mass exhalation rate (E_m), and dissolved radon concentration C_{diss} in tea samples.

Code	Track Density (track/mm ²)	CRn (Bq/m ³)	E_A (Bq/m ² .h)	E_m (Bq/kg.h)	C_{diss} (Bq/m ³)
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.

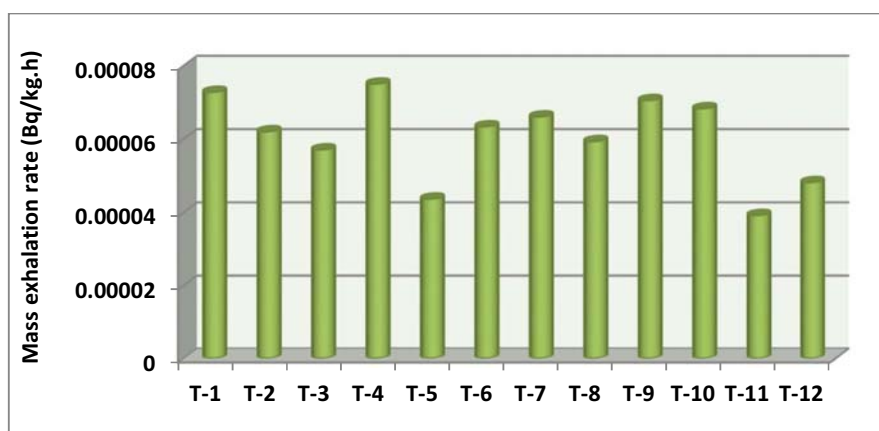


Figure 4. Mass exhalation rate in tea samples, for radon.

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/m³ in the sample T-8 (Lipton tea, SriLanka-India origin) to the highest concentration 10.164 Bq/m³ in the sample T-5 (Mahmood tea, SriLanka-India origin), with an average value 4.143 Bq/m³.

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/m³ 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/m³).

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/m².h in sample T-8 (Lipton tea, SriLanka-India origin) to the highest value 0.011 Bq/m².h was in sample T-5 (Mahmood tea, SriLanka-India origin). While the mass exhalation rate varies from 2.7×10^{-5} Bq/kg.h in sample T-8 to the highest value 21.9×10^{-5} Bq/kg.h in a sample T-5. With an average values 0.005 Bq/m².h and 9.1×10^{-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 (C_{RnB}), Radon gas concentration at the top of the can (C_{RnT}), Thoron gas concentration (C_{Th}), surface exhalation rate (E_A), and mass exhalation rate (E_m).

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

Average	35.139	27.98392	4.143	0.005	9.1×10^{-5}
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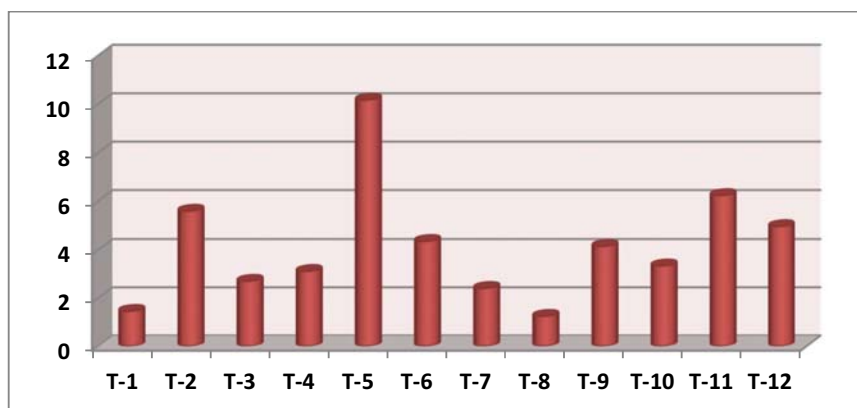


Figure 5. Thoron gas concentration in tea samples.

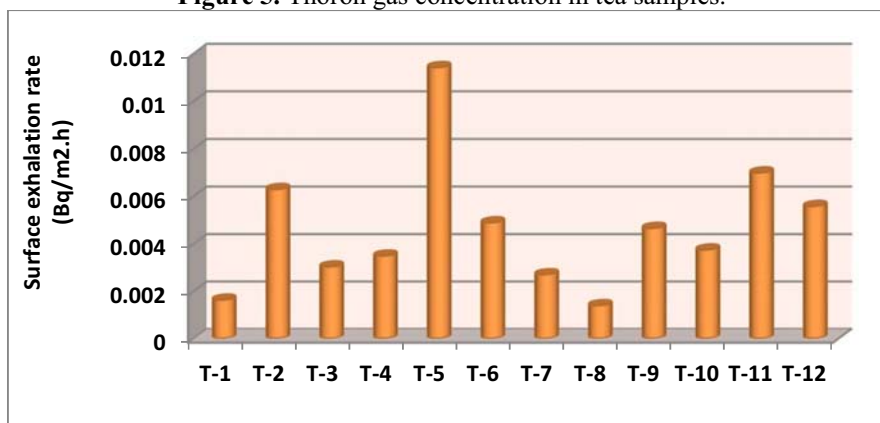


Figure 6. Surface exhalation rate in tea samples, for thoron.

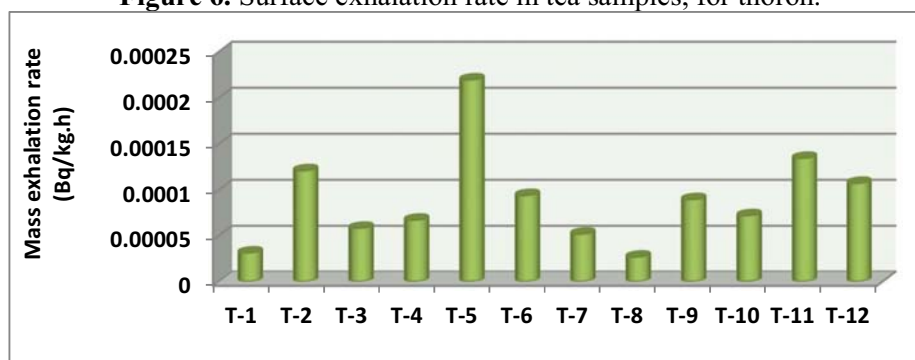


Figure 7. Mass exhalation rate in tea samples, for thoron.

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 8.78MeV 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.

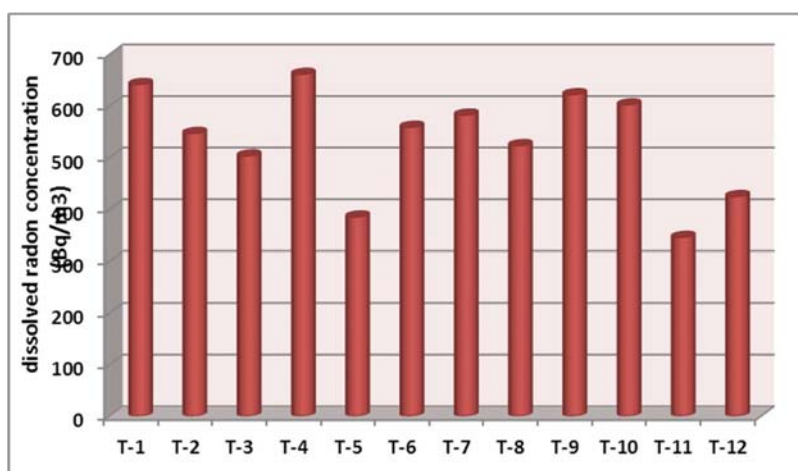


Figure 8. Dissolved radon concentration in tea samples.

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 Mahmood 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 or 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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