The effective radium content and radon concentrations in coffee samples

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ABSTRACT

Background: Many people in Iraq and indeed the world choose to drink coffee on a daily basis; accordingly, we planned to find the radium and uranium concentrations in addition to the Radon emission rate to determine if coffee shows the levels of radioactivity that may affect people’s lives.

Material and Methods: A nuclear track detector (CR-39) was used to study the natural radioactivity of ten samples of coffee collected from local Iraqi markets. Results: The effective radium content varied from 0.13 to 0.66 Bq/kg with a mean 0.32 Bq/kg. The mass exhalation rates of radon vary from 0.024 – 0.121/kg .h, with a mean 0.059 Bq/kg .h, while the surface exhalation rates of radon vary from 0.47 – 2.39/m2.h, with a mean 1.16 Bq/m2. h. Uranium content varied from 0.10 to 0.54 ppm with a mean value of 0.26 ppm. Conclusion: Insofar as health effects are concerned, the results have shown that all the samples were entirely safe.

Keywords: Coffee, CR-39, radon exhalation rates, radium.

INTRODUCTION

Radon is a heavier than air gas and is referred to as radium emission. Since radon gas is colourless, odourless, tasteless and invisible, appropriate detectors should be used to detect its presence. Radon-222 is the radioactive natural gas that arises from the decay of radium-226 as part of the uranium-238 decay series. Radium is distributed in soil rocks, ocean water and sediments. As radon atoms decay, they emit alpha molecules with an energy of 5.486 MeV, producing polonium isotopes (Po-218 and Po-214) (1–3).

Radium is a naturally radioactive element found in trace amounts throughout the earth's crust. Radium decomposition leads to radon in the environment (internal and external), soil, groundwater, oil and gas oil. It is estimated that radon, to a large extent in households, accounts for more than 50% of the equivalent dose received by the general population from all sources of radiation, both natural and man-made (4–6).

The alpha particles emitted from this radon and other forms of radiation emitted from its products increase the absorbed dose in the respiratory and digestive systems (7,8).

Exposure occurs when airborne radon is inhaled while using water: bathing, washing dishes, cooking, and drinking water, and other daily objects, all contain radon (9).

226Ra is widespread in the environment, with different (though trace) concentrations in water, soil, sediment and rock (10,11).

When radium is ingested, most substances that contain it are rapidly excreted. However, since the mechanism of chemical uptake of radium is identical to that of calcium, radium will be absorbed into the blood from the gastrointestinal tract or lungs follows the calcium conduit and is deposited in the bone (12).

Radium is a common radionuclide within the environment and is the parent species of radon in the uranium decay series. 226Ra is amongst the most lethal of radionuclides because it produces alpha particles and has a very long half-life (1600 y) (13). The measurement of the natural
radiation activity of the alpha particles emitted from food, water and beverages of all kinds is one of the subjects related to the health effects of radioactivity on human health and the potential risks that could affect people's lives, especially cancerous diseases. Many people in Iraq, and indeed the world, like to drink coffee on a daily basis; accordingly, this study was conducted to determine the potential importance to human health in terms of whether the limited radioactivity in coffee might nevertheless that may affect people's lives.

This study aims to measure the concentrations of uranium and radium in and the rate of radon exhalation that can be achieved from, coffee samples collected from a local Iraqi market. This study has potential implications for public health across the world because the coffee plant is grown in different soil types and consumed by millions of people daily. These plants are often sustained by soils that, relatively speaking, contain a lot of radioactive nuclei.

**MATERIALS AND METHODS**

The concentration of uranium, the effective radium content and the rate of radon exhalation of the radon were determined by a passive technique, namely solid-state nuclear track detectors using the so-called "sealed can technique" (14-18).

**Sampling and samples preparing**

Ten samples of coffee were collected from local Iraqi markets. They were then roasted and dried in an oven to drive off any residual moisture. They were then ground to a very fine powder and sieved using a 2 mm pore sieve. The samples were thus produced in a soft powder form after extracting any remaining large granules using the sieve. 19 g of each sample of coffee were taken and placed in a plastic cylinder container facing the CR-39 nuclear track detector used the sealed cup technique shown in figure 1. In this technique, the same weight of coffee was placed and left in a sealed cup for four weeks in order to reach the equilibrium between radium and radon.

19 g and a 3.5 cm height of each coffee sample were placed separately to each other in the bottom of a closed cup with a height of 6.5 cm and a diameter of 3.5 cm, with a fixed, 3 cm distance between the sample surface and the CR-39 detector (thickness, 500 μm and area, 1.5×1.5 cm²) installed in the upper part of the lid of the cup, as shown in figure 1.

**Etching and scanning process**

During the 62-day sample storage, the CR-39 nuclear track detector recorded the pathways taken by the alpha particles emitted from the radon generated by the radium decomposition in the samples. The effects on the detector surface can only be detected and observed through the process of electrochemical drilling of the detector using 6.25 M sodium hydroxide solution at 70 ± 1°C in a fixed temperature water bath to detect the pathways. After finishing with the reagents, the CR-39 nuclear track detector was washed with water and dried for a period of 30 minutes. The tracks on the detector were counted using a kruss-mbl 2000 optical microscope at a ×100 magnification.

**Theoretical considerations**

**Radon concentration**

After determining the total number of tracks (N_i) on the detector surface for all samples, the density of the tracks (ρ) formed on the detector surface can be calculated using the following relationship (19):

$$\rho = \frac{\sum N_i}{nA}$$

Figure 1. A test tube technique used in the study.
Where \( A \) and \( n \) are the area of the field of view and the total number of tracks in the field of view, respectively.

Radon activity concentrations \( (C_{Rn}) \) in the volume above the samples are correlated with the density of the nuclear pathways, the time of exposure, \( T \), and the calibration factor, \( K \), of the CR-39 plastic track detector via the following mathematical formula (20):

\[
P = KC_{Rn}T
\]

where the value of \( K \) is equal to 0.223 track.cm\(^{-2}\)/Bq.d.m\(^{-3}\) (21).

Alpha decomposition of radon can be used to determine the concentration of radium in all samples stored in the sealed cups. An effective radon-radium balance (about 98%) can be found in the decay chain in about four weeks because the half-life of \(^{226}\)Ra is 1600 years, whilst that of \(^{222}\)Rn is 3.82 days. Once the radioactive balance is established, radon activity increases with time \( T \) according to the following relationship (22):

\[
C_{Rn} = C_{Ra} \left(1 - e^{-\lambda_{Rn}T}\right)
\]

Equation 3 shows the relationship between the effective radium content \( (C_{Ra}) \) of the sample and the concentration of radon \( (C_{Rn}) \) in the box air used in this study. \( \lambda_{Rn} \) here is the decay constant of \(^{222}\)Rn. The plastic track effect detector measures the total number of alpha disintegration instances per unit size of the cup during the exposure period via the calibration factor. The track density of the alpha particles monitored are given in the following relationship (23):

\[
P = KC_{Ra}T_e
\]

The effective exposure time \( (T_e) \) associated with the real exposure time \( (T) \) is as follows (24):

\[
T_e = \left[T - \frac{1}{\lambda_{Rn}}(1 - e^{-\lambda_{Rn}T})\right]
\]

Equation 6 can then be used to calculate the dissolved radon concentration in coffee samples (25):

\[
C_s = C_{Rn} \frac{\lambda h T}{L}
\]

This equation connects the radon concentrations in the ambient air \( (C_{Rn}) \) from the sample, the dissolved radon concentration of the sample \( (C_s) \), the decomposition of the radon \( (\lambda) \), the sample height \( (L) \), the distance between the sample surface and the detector installed in the upper part of the lid of the box \( (h) \), and the exposure time \( (T) \), also known as the sample storage period, with the nuclear impact detector.

### Effective radium content

The effective radium content \( [C_{Ra} \text{ (Bq.kg}^{-1})] \) can be calculated by taking into account the mass \( (M) \) of the coffee sample, the surface area \( (A) \) of the sample placed in the closed cups and the distance \( (h) \) between the sample surface and the detector surface installed in the upper inner surface of the lid of the box with equation 4, as per equation 7 (16,17,23,26):

\[
C_{Ra} \text{ (Bq.kg}^{-1}) = \left(\frac{P}{K T_e}\right) \left(\frac{h A}{M}\right)
\]

### Radon exhalation rates

The radon exhalation rate in terms of mass is given by equation 8 (6):

\[
E_M \text{ (Bq.kg}^{-1}.d}^{-1} = \frac{CV \lambda}{M[T + \lambda^{-1}(e^{-\lambda T} - 1)]}
\]

Where \( E_M \) is the radon exhalation rate in terms of mass expressed in Bq.kg\(^{-1}.d}^{-1} \), and \( M \) is the mass of the sample (kg).

The radon exhalation rate in terms of area was obtained from the expression shown as equation 9 (27,28):

\[
E_A \text{ (Bq.m}^{-2}.d^{-1} = \frac{CV \lambda}{A[T + \lambda^{-1}(e^{-\lambda T} - 1)]}
\]

Where \( E_A \) is the radon exhalation rate in terms of area expressed in Bq.m\(^{-2}.d}^{-1} \), \( C \) is the integrated radon exposure expressed in Bq.m\(^{-3}.d \), \( V \) is the effective volume of the cup in m\(^3 \), \( T \) is the exposure time in hours, \( \lambda \) is the decay constant for \(^{222}\)Rn radon (d\(^{-1}) \), and \( A \) is the area of the cup (m\(^2\)).
Uranium concentration

The ratio of the weight of the uranium in the sample ($W_U$) to the weight of the coffee sample ($W_S$) itself enables us to determine the concentrations of uranium ($C_U$) in the coffee samples in units of parts per million (ppm) according to the following equation (10) (27,28):

$$C_U^\text{ppm} = \frac{W_U}{W_S} \quad (10)$$

RESULTS AND DISCUSSION

In this study, a sealed can technique, which contained the CR-39 nuclear track detector, was used to study the natural radioactivity of alpha particles emitted from ten different samples of coffee imported into local Iraqi markets. Table 1 reports various properties of the samples including uranium concentrations, effective radium content, mass and the surface area for radon exhalation rate. In these results, radium concentrations were found to range from 0.13 to 0.66 Bq/kg with a mean value of 0.32 Bq/kg. Results showed that the highest concentration of uranium was 0.54 ppm in the House of Brazilian, while the lowest concentration of uranium was 0.10 ppm in Coffee prince with an average of 0.26 ppm for those samples whose radium concentrations were measured.

The effective radium content for coffee samples calculated in this paper are lower than those reported by Ibrahim et al. (2007) for garden rocket, and are similar to the values typically found for cucumber, carrot, spinach, green beans and green haricots (29).

The results for the uranium concentrations in coffee samples were well below the allowed limit (11.7 ppm) (30). The concentrations of uranium and radium in the coffee samples in this study were very close to those found in a previous study on a variety of vegetables typically found in Iraqi markets (31).

The mass exhalation rates varied from 0.024 to 0.121 Bq/kg.h with a mean value of 0.059 Bq/kg.h, while the surface exhalation rates varied from 0.47 to 2.39 Bq/m².h with a mean value of 1.16 Bq/m².h.

Figure 2 shows the distribution of radium content for the 10 samples of coffee available from the local Iraqi markets. Figure 3 shows the positive linear relationship between the radon exhalation rate per unit mass and the effective radium content, where the correlation coefficient between them is 0.9999 ($R^2 = 0.9999$). Figures 4 and 5 show the excellent positive linear relationship between the effective radium content and the radon exhalation rate for the surface unit and the uranium concentration. The values of the radon exhalation rate for the mass and surface units were found to vary depending on the changes in the uranium and radium concentrations in the samples.

Table 1. Track density, concentrations of radon in air tube and dissolved samples, uranium concentrations, surface and mass exhalation rates and effective radium concentrations for ten coffee samples.

<table>
<thead>
<tr>
<th>Code</th>
<th>Sample Name</th>
<th>$p \times 10^3$ Trak/cm²</th>
<th>$C_{\text{Ra}}$ Bq/m²</th>
<th>$C_{U} \times 10^2$ Bq/m²</th>
<th>$C_{\text{Ra}}$ Bq/kg</th>
<th>$E_{\text{m}}$ mBq/kg.h</th>
<th>$E_{\text{a}}$ mBq/m².h</th>
<th>CU ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>Coffee stop</td>
<td>3.88</td>
<td>280.63</td>
<td>3.68</td>
<td>0.54</td>
<td>0.099</td>
<td>1.95</td>
<td>0.44</td>
</tr>
<tr>
<td>C2</td>
<td>Mass café</td>
<td>2.29</td>
<td>165.99</td>
<td>2.17</td>
<td>0.32</td>
<td>0.059</td>
<td>1.15</td>
<td>0.26</td>
</tr>
<tr>
<td>C3</td>
<td>Mahmood coffee</td>
<td>1.89</td>
<td>136.69</td>
<td>1.79</td>
<td>0.26</td>
<td>0.048</td>
<td>0.95</td>
<td>0.21</td>
</tr>
<tr>
<td>C4</td>
<td>Nescafe matinal</td>
<td>1.00</td>
<td>72.327</td>
<td>0.94</td>
<td>0.14</td>
<td>0.026</td>
<td>0.50</td>
<td>0.11</td>
</tr>
<tr>
<td>C5</td>
<td>Coffee prince</td>
<td>0.93</td>
<td>67.380</td>
<td>0.88</td>
<td>0.13</td>
<td>0.024</td>
<td>0.47</td>
<td>0.10</td>
</tr>
<tr>
<td>C6</td>
<td>Brazilian of club</td>
<td>1.36</td>
<td>98.365</td>
<td>1.29</td>
<td>0.19</td>
<td>0.035</td>
<td>0.68</td>
<td>0.15</td>
</tr>
<tr>
<td>C7</td>
<td>House of Brazilian</td>
<td>4.74</td>
<td>343.19</td>
<td>4.50</td>
<td>0.66</td>
<td>0.121</td>
<td>2.39</td>
<td>0.54</td>
</tr>
<tr>
<td>C8</td>
<td>Ben shire</td>
<td>2.10</td>
<td>151.88</td>
<td>1.99</td>
<td>0.29</td>
<td>0.054</td>
<td>1.05</td>
<td>0.24</td>
</tr>
<tr>
<td>C9</td>
<td>Coffee belle</td>
<td>2.73</td>
<td>197.45</td>
<td>2.59</td>
<td>0.38</td>
<td>0.070</td>
<td>1.37</td>
<td>0.31</td>
</tr>
<tr>
<td>C10</td>
<td>Coffee break</td>
<td>2.21</td>
<td>159.84</td>
<td>2.09</td>
<td>0.31</td>
<td>0.056</td>
<td>1.11</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td>Minimum</td>
<td>0.93</td>
<td>67.38</td>
<td>0.88</td>
<td>0.13</td>
<td>0.024</td>
<td>0.47</td>
<td>0.10</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>4.74</td>
<td>343.19</td>
<td>3.68</td>
<td>0.66</td>
<td>0.121</td>
<td>2.39</td>
<td>0.54</td>
</tr>
<tr>
<td></td>
<td>Mean</td>
<td>2.31</td>
<td>167.33</td>
<td>1.93</td>
<td>0.32</td>
<td>0.059</td>
<td>1.16</td>
<td>0.26</td>
</tr>
</tbody>
</table>
CONCLUSION

In this study, CR-39 detectors are widely used for the determination of effective radium content, radon exhalation rate and uranium concentration measurements in coffee samples. The results for the uranium concentrations and effective radium content in coffee samples were found to range between 0.10 - 0.54 ppm and 0.13 - 0.66 Bq/kg, respectively. *House of Brazilian* (C1) was found to have the highest concentration, while *Coffee prince* (C5) the lowest. The mean radium and uranium concentrations were found as 0.32 Bq/kg and 0.26 ppm, respectively.

Concentrations of radium and uranium in coffee vary widely because of the differing background levels, climates, and agricultural conditions that prevail where they are cultivated.

Finally, the results gained from the samples imply that they are safe for use as far as effects on human health are concerned, being well below what are considered to be acceptable limits. There was a positive correlation between uranium concentration and radium content in all coffee samples.

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REFERENCES

1. Jagadeesha BG and Narayana Y (2017) Radon exhalation rate measurement in the environment of Hassan district of...
southern India, Radiochemistry, 59(1): 104-108.


