Measurement the uranium, radon and radium concentrations in urine samples for diabetics in Najaf city.

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Abstract
In this study, the total number of urine samples were 88 for diabetic patients and healthy controls in Najaf city of Iraq. Uranium, radium and radon concentration levels were estimated using CR-39 detector method. The samples have been grouped in two groups of 44 individuals each. The first group of urine samples were taken as 22 healthy controls and 22 diabetic patients from different regions of Najaf city. The second group were taken from Al-Ansaar region also as 22 healthy controls and 22 diabetic patients. Results indicate the presence of uranium, radium and radon pollution in all urine samples for four groups. It was found that urine samples of Al-Ansaar area were more polluted with radiation than those of other regions in Najaf city. Also urine samples of diabetic patients were more polluted specially with radon than urine samples of healthy controls.

Keywords: Uranium, Radon, Radium, Urine, Diabetics.

1. Introduction
During the first Gulf War in 1991, and during the military operations to occupy Iraq in 2003, the America and British armed forces used depleted uranium munitions in populated areas, especially in southern Iraq. As a result, the area was contaminated with radioactive materials, where the most important source of radiation exposure was depleted uranium aerosols and oxides, in addition to other sources of radiation exposure such as destroyed armor fragments containing radionuclides of the uranium decay chain emitted from them such as radium-226 and radon-222 from the remaining contaminated soil near these destroyed targets [1]. Laboratory, clinical and pathological studies of persons and warriors in areas facing radiation doses have demonstrated that an increase in human exposure to radioactivity, even at low levels, for repeated or long periods of time leads to health damage to the body in proportion to the quantity and type of radiation dose and the general and genetic health of the person. Among these diseases: loss of cellular immunity, autoimmune diseases such as leukemia, other cancerous diseases of the lung, liver, colon, thyroid glands, diabetes, etc., as well as to chromosomal abnormalities that generate birth defects in future generations and infertility[2].
Uranium, radium, and radon belong to the group of primitive radionuclides, and these nuclides are always present on Earth. The radionuclides uranium-238 ($^{238}\text{U}$), uranium-235 ($^{235}\text{U}$) and thorium-232 ($^{232}\text{Th}$) that decay to other nuclei by emitting nuclear radiation and particles through three distinct chains of radionuclides. Likewise, those radioactive nucleons are present in the human body and nutrients in very small quantities [3].

There are three naturally occurring uranium isotopes that are of great importance in relation to the mining of this element and the nuclear industry. These include U$^{238}$, which comprises the majority of this element in the Earth’s crust, U$^{235}$, and U$^{234}$, which together comprise a much smaller portion. The half-lives of these isotopes are 4500 million years, 703 million years, and 2.46×10$^5$ years, respectively [4]. Uranium is a heavy metal with density 19.0g/cm$^3$, weakly radioactive and have great chemical toxicity [3]. The health effects of uranium, which include lung cancer, kidney damage and DNA damage. The World Health Organization (WHO) and most of the national regulatory agencies have established minimum, recommended or permissible exposure limits for soluble and insoluble uranium compounds by ingestion and inhalation in which the public’s intake of soluble uranium compounds should not exceed 0.5µg/kg body weight per day by ingestion and 1µg/m$^3$ by inhalation[5]. For healthy person the concentration uranium was limited by WHO is 0.1µg/L [6].

Half-life of Ra$^{226}$ is 1600 years, Ra$^{224}$ and Ra$^{228}$ half-lives 3.6 days and 5.8 years, respectively. In the environment where it is found in different concentrations in water and soil. The predecessor of radium is U$^{238}$. The chemical behavior of U$^{238}$ is similar to that of calcium in the human body, as radium is absorbed into the blood from the digestive system and lungs, and is similar to the behavior of calcium is mainly deposited in the bones. Microscopic amounts of radium in the environment can lead to the accumulation of some radium in bone tissue due to ingestion or the body’s exposure to radium for serious health effects including anemia, ulcers, bone cancer and other disorders [3,7], where the natural level of radium in blood from food and water is 0.003 pCi/kg[8].

Radon gas Rn$^{222}$ is a decay product of radium with a half-life of 3.8d. Humans are exposed to radon by ingesting and digesting contaminated plant and animal foods or by inhaling air polluted with radon gas. Epidemiological studies have shown that exposure to radon gas at a higher than the permissible dose increases the risk of leukemia and lung cancer, the allowed limit according to International Commission on Radiological Protection and International Atomic Energy Agency is 200Bq/m$^3$[5,9].

In this study the concentrations of uranium, radium and radon as a result of radioactive contamination in urine samples of diabetics and healthy people in the province of Najaf were measured. Recently, the number of diabetics in the Najaf city increased by proportions with percentage 4.3% according to the statistics of the Najaf Health department for year 2018, so this study aims to measure the level of radioactive contamination by uranium, radium and radon in urine samples of diabetics and the effect of this radioactive contamination on the development of diabetes in patients and healthy people, and thus increase the number of diabetics in Najaf city. Samples taken from different regions of Najaf represent controls people without radioactive contamination, while samples taken from Al-Ánsar region represent radioactive samples because Al-Ánsar region is considered a radioactive polluter region according to the environmental department of Najaf city.

2. Materials and Methods
2.1. Patients Groups and Controls
A total of 88 urine samples were collected as 44 patients them were taken from Diabetes Center in the Al-Sadr Hospital which includes all the regions and from Al-Ansaar region Health Center which includes Al-Ansaar region in Najaf city Iraq. While 44 people healthy controls they were taken from Al-Sadr Hospital which includes different regions and from Al-Ansaar region Health Center which includes Al-Ansaar region in Najaf city, Iraq. The following are the groups in all age and in both sexes of patients and controls that were involved in this study:

1-Group one G1: Included 22 healthy controls them were taken from Al-Sadr Hospital which includes different regions in Najaf city, Iraq.
2-Group two G2: Included 22 diabetic patients samples of them were taken from Diabetes Center in the Al-Sadr Hospital which includes different the regions in Najaf city, Iraq.
3-Group three G3: Included 22 healthy controls them were taken from Al-Ansaar region Health Center in Najaf city, Iraq.
4-Group four G4: Included 22 diabetic patients of them were taken from Al-Ansaar region Health Center in Najaf city, Iraq.

2.2. Collection of Urine Samples
Three milliliters of urine samples were taken from all healthy and patient’s individuals included in this study. Urine samples put in vacuumed tubes and addition drop from concentrated HCl to avoid precipitate formation. Track Detector CR-39 was used to measure uranium, radon, and radium concentrations in urine samples.

2.3. Preparation of Solution
Etching solution sodium hydroxide was prepared by dissolving 100g of NaOH pellets into 400mL of distilled water (D.W) in Pyrex Flask. This process was done carefully using Pyrex Flask since dissolving NaOH usually releases heat so this solution was prepared for at least one day before it was used.

2.4. Assay Method
In this study, internal ionizing radiation pollution with uranium, radon and radium, and there concentrations in urine samples of healthy and diabetic patients were calculated using the method of Solid State Nuclear Track Detectors CR-39. The polymer sheets CR-39 were cut into small square pieces with area 1cm×1cm and placed at the bottom and top of the tube with length 5.5cm and radius (r) 0.75cm, then placed the urine sample with height 3cm in this tube and closed tightly to prevent air intake, as shown in Fig.1. The piece of CR-39 placed at the bottom of the tube is used to record the uranium concentration, while the piece of CR-39 placed at the top of the tube is used to record radon gas and radium as shown in Fig.2. Then the samples of urine are saved for 90day.
CR-39 detector are removed from urine samples and washed well with flush water. Chemical etching was carried out using NaOH solute for 6hr at 70ºC using water bath. After etching completed, detectors CR-39 pieces were removed and washed under flush water, cleaned and dried for microscopic examination using the Microscope Lens (WF40/0.65 160/0.17) which have squares with known areas to do counting of tracks resulted.

2.5. Track Count and Uranium Concentration Calculation

After counting tracks in each etching piece of CR-39, track density $\rho$ was calculated using Eqn.1, then each $\rho$ value that obtained was used to calculated uranium concentration $U_C$ for each urine samples using Eqn.2.

\[
\rho = \frac{N}{(A \times T)}
\]

where $\rho$ is the track density in unit tracks/cm$^2$.h.

$N$ is the average number of tracks.

$A$ is the area of the field view in unit cm$^2$.

$T$ is the irradiance time in unit h.

From the calibration curve and fitting, $U_C$ can calculated from the track density using Eqn.2

\[
U_C = \frac{(\rho + 12.5)}{18.6}
\]

where $U_C$ is the uranium concentration in unit (ppb)[10].

2.6. Track Count and Radon and Radium Concentration Calculation

After counting tracks in each etching piece of CR-39 are placed in upper of tube and calculating the track density $\rho_{rn}$, the concentration of radon in unit (Tr/cm$^3$) in the air gap of the tube, $C_o$ was calculated using Eqn.3.
\[ C_o \left( \frac{Bq}{m^3} \right) = \frac{\rho_{rn} T_{rn}}{K} \]  \hspace{1cm} (3)

where

- \( T_{rn} \) is the irradiance time in unit day (T=90d).
- \( K \) represents the calibration coefficient for radon that determined using Eqn.4.

\[ K = 0.25r \left( 2 \cos \theta_c - \frac{T}{r_o} \right) \]  \hspace{1cm} (4)

where \( \theta_c \) is the critical angle of the CR-39 detector (\( \theta_c = 35^\circ \)).

\( r_o \) represents the range of alpha particles in the air (\( r_o = 4.15 \text{cm} \)). The calibration coefficient can obtained from used Eqn.4 and equal (\( K = 0.273 \text{cm} \)) in dimension of length or equal (\( K = 0.02358 \text{Track cm}^{-2}/\text{Bqm}^{-3} \text{d} \))\[11\].

The concentration of \( ^{222}\text{Rn} \) within the sample \( C_{Rn} \) can be obtained using Eqn.5.

\[ C_{Rn} \left( \frac{Bq}{m^3} \right) = \frac{C_o \lambda_{Rn} h T_{rn}}{I} \]  \hspace{1cm} (5)

where \( \lambda_{Rn} \) represents \( ^{222}\text{Rn} \) decay constant equal to 0.1814d\(^{-1}\).

\( h \) is the distance between the sample and the CR-39 detector.

\( I \) is the thickness of the sample layer in the tube.

The concentration of \( ^{228}\text{Ra} \) within the sample \( C_{Ra} \) can be obtained using Eqn.6\[9\].

\[ C_{Ra} \left( \frac{Bq}{kg} \right) = \frac{C_{Rn} h A_s}{m_s} \]  \hspace{1cm} (6)

where \( A_s \) represents the surface area of the sample.

\( m_s \) is the sample mass in unit kg.

All parameters in Eqn.5 and Eqn.6 are given from Fig.1 can be compensated when calculate concentration of \( ^{222}\text{Rn} \) and \( ^{228}\text{Ra} \) in urine samples.

2.7. Statistical Analysis

Results are reported as Mean ± STD Deviation SPSS (Graph Pad Prism.7) program. For comparisons among four groups, the unrestricted Student’s t test was performed. The previously level of significance was set at P<0.05.

3. Results and Discussion

In this study, the estimated radioactive internal pollution of uranium, radium, and radon to which the population was exposed during and after the military operations of the first Gulf War in 1991 and 2003 as a result of the use of depleted uranium shells was done. The concentrations of uranium, radon, and radium were calculated in 88 urine samples by a CR-39 detector for diabetics and health people in the city of Najaf as a result of the increasing number of diabetics in recent years in this city and compared the results with the permitted and recommended levels according to the WHO, the International Commission for Radiation Protection and the International Atomic Energy Agency.

3.1. Comparison of Uranium Concentration Levels in Urine Samples in Four Groups

The results of uranium concentrations in unit (µg/L) for the 88 urine samples for diabetics and healthy controls in the Najaf city are presented in Table1, the range for four groups (\( G_1,G_2,G_3,G_4 \)), are (0.800-2.164), (0.845-1.479), (0.841-1.543) and (0.831-1.122) µg /L, but the Mean ± STD (Standard Deviation) for four groups are (1.303±0.316), (1.107±0.153), (1.051±0.214) and (0.972±0.082) respectively. Table1 represents the minimum and maximum values of uranium concentrations which they are higher than the permissible limit for according
values by WHO for healthy person is 0.1µg /L. Figure3 shows the comparison of uranium concentration levels in urine samples among four groups (G1, G2, G3, G4).

### Table 1: The levels of uranium concentration in urine for four groups

| Variable                        | G1            | G2            | G3            | G4            |
|---------------------------------|---------------|---------------|---------------|---------------|
| **Range**                       | (0.800-2.164) | (0.845-1.479) | (0.841-1.543) | (0.831-1.122) |
| **[Mean ±STD Deviation]**       | 1.303±0.316   | 1.107±0.153   | 1.051±0.214   | 0.972±0.082   |
| **P-value**                     | 0.957         | 0.836         | 0.275         | 0.000         |
| **Total P-value**               | <0.0001 ****  |               |               |               |

Figure3: The levels of uranium concentration in urine for four groups in this study, where the phrases (⦁,⦁,⦁,⦁) represent the ((G1, G2, G3 and G4) groups one, two, three and four : healthy controls and diabetic patients samples were taken from different regions and Al-Ansaar region in Najaf city, respectively.

### 3.2. Comparison of Radium Concentration Levels in Urine Samples in Four Groups:
The results of radium concentrations in unit (Bq/kg) for the 88 urine samples for diabetics and healthy controls in the Najaf city are presented in Table 2. The range values of radium concentration in samples of four groups (G1, G2, G3, G4) are (2.054-15.519), (1.609-10.097), (3.756-24.294) and (5.095-25.644) respectively. But the Mean±STD for these groups are (5.957±3.145), (5.329±2.916), (9.268±5.609) and (10.161±5.118) respectively. Table 2 represents the minimum and maximum values of radium concentrations in these samples that are higher than the natural level of radium in urine as the ICRP Publication 23 on Reference Man, Ra226 loss in urine is 0.00296Bq/d (0.08 pCi/d) [8]. The results of the radium concentrations obtained in this study are close to the results of the radium concentration in the tissues of pasture animals, vegetables and fruits consumed by humans that were recorded in the years (1991-1996) [1]. Figure 4 and (Mean ±STD) values in Table 2 show G4 (10.161±5.118) values are higher than concentrations in G1 and G2 with p=0.0104 and p=0.0023, respectively, where G3 (9.268±5.609) are higher than concentrations in G1 with p=0.0643.

Table 2: The levels of radium concentration in urine for four groups

| Variable         | G1        | G2        | G3        | G4        |
|------------------|-----------|-----------|-----------|-----------|
| **Range**        | (2.054-15.519) | (1.609-10.097) | (3.756-24.294) | (5.095-25.644) |
| **Mean ±STD Deviation** | (5.957±3.145) | (5.329±2.916) | (9.268±5.609) | (10.161±5.118) |
| **P-value**      | 0.991     | 0.989     | 0.975     | 1         |
| **Total P-value**| 0.0005    |           |           |           |
Figure 4: The levels of radium concentration in urine for four groups in this study, Where that the phrases (⦁⦁⦁⦁) represent the (G1, G2, G3 and G4) groups one, two, three and four: healthy controls and diabetic patients samples were taken from different regions and Al-Ansaar region in Najaf city, respectively.

3.3. Comparison of Radon Concentration Levels in Urine Samples in Four Groups:

The results of radon concentrations in unit (Bq/m³) for the 88 urine samples for diabetics and healthy controls in the Najaf city are presented in Table 3. The measured radon concentration for four groups (G1, G2, G3, G4) are (110.516-834.818), (86.544-543.160), (202.074-1306.935), and (274.085-1379.531) respectively. While the Mean±STD for these groups are (320.479±169.162), (286.669±156.869), (498.593±301.754) and (546.586±275.321).

By observing Table 3 we find that the minimum and maximum values of radon concentrations are higher than that the allowed limit according to International Commission on Radiological Protection and International Atomic Energy Agency 200Bq/m³[5,9]. From Figure 5 and Table3 we notice that radon concentrations in G4 (546.586±275.321) are higher than concentrations in G1 and G2 with p=0.0104 and p=0.0023, respectively, for G3 (498.593±301.754) we notice that radon concentrations in G3 are higher than concentrations in G1 with p=0.0643.

| Variable | G1                  | G2                  | G3                  | G4                  |
|----------|---------------------|---------------------|---------------------|---------------------|
| Range    | (110.516-834.818)   | (86.544-543.160)    | (202.074-1306.935)  | (274.085-1379.531)  |
| [Mean ± STD Deviation] | (320.479±169.162) | (286.669±156.869) | (498.593±301.754) | (546.586±275.321) |
| P-value  | 1                   | 1                   | 1                   | 1                   |
| Total P-value | 0.0005              |                     |                     |                     |
Figure 5: The levels of radon concentration in urine for four groups in this study, where that the phrases (⦁⦁⦁⦁) represent the ((G1, G2, G3 and G4) groups one, two, three and four: healthy controls and diabetic patients samples were taken from different regions and Al-Ansaar region in Najaf city, respectively.

It was found through the results that urine samples of Al-Ansar area in Najaf is more polluted by radiation than the rest of Najaf and that the urine samples of people with diabetes are more polluted specially with radon. The pollution of urine is caused by environmental elements pollution such as water, soil and air, and there are previous studies confirm the contamination of environmental elements of al-Najaf city[12].

4. Conclusions
The concentrations of uranium, radium and radon have been measured using CR-39 detector. The results showed their presence of chronic exposure to uranium, radium and radon pollution of healthy controls and diabetics samples in Najaf city, where that uranium, radium and radon concentrations in urine samples were higher than the permissible limits according WHO and International Commission on Radiological Protection and International Atomic Energy Agency. Therefore, radioactive contamination helps in the development of diabetes in diabetics and causes the possibility of risk diabetes in healthy controls.
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