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Measurement of Radon Levels in the Groundwater of Al-Rusaifah City in Zarqa Governorate Using Liquid Scintillation Counter

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Abstract

Groundwater wells in Al- Rusaifah in central Jordan were investigated for radon (^{222}Rn) levels during the winter season using a liquid scintillation counter (LSC). ^{222}Rn was detected in eighteen groundwater samples from six different wells. The concentrations of radon varied from 151.9 to 253.1 Bq/L. The upper concentration limit of ^{222}Rn for drinking water is 11 Bq/L based on the Jordanian Standards for Drinking Purposes. This indicates that ^{222}Rn levels in all groundwater samples exceed the permissible threshold. The health impact assessment of ^{222}Rn showed that for all samples, the total population-weighted average annual effective dose was 3.147mSv/y, which is greater than the maximum value of the general annual effective dose from radon of 0.1 mSv/y, according to the WHO and the European Council.

Keywords: Radon, Al- Rusaifah, Liquid Scintillation Counter, Annual Effective Dose, Zarqa Governorate, Jordan

1. Introduction

Water for human consumption should be free of chemical, microbiological, and radiological contamination (Garba et al., 2008).

Groundwater has more radioactive contents than surface water, since it passes through rocks and soil formations, dissolving many compounds, minerals, and radioactive materials (Mohsen et al., 2016).

^{222}Rn is a naturally-occurring radioactive inert gas, which is a descendent of the uranium decay series and represents the important radon isotopes. It has no taste, smell, or color and its melting point is 202 k (-71C°), boiling point 211.3 k (-61.7 C°), and density at 293 k is 9.73 g/cm^3. Chemically, Radon is unreactive with most materials, so it travels and moves easily through very small spaces (Al-Zubaidy and Mohammad, 2012).

In natural water, Radon originates from the dissolution of radium (^{226}Ra) in groundwater from bed rocks. It doesn't react with any minerals.

Most people do not know that the greatest radiation exposure is caused by natural radiation sources. It is well-known that more than half of the radiation dose is caused by radon and its progeny (^{218}poand ^{214}po) (UNSCEAR, 1993).

Radon and its daughters, especially polonium-218 and polonium-214, are radioactive nuclei that emit alpha particles with energies of 6.003 and 7.678 MeV, respectively (polonium-214, are radioactive nuclei that emit alpha particles).

Radon exists in three natural isotopes, (^{219}Rn, Actinon) and (^{220}Rn, Thoron) and (^{222}Rn, Radon) which are formed by the decay of different isotopes of radium found in the decay chains of ^{235}U, ^{232}Th and ^{238}U, respectively. Actinon and Thoron have very short half-lives (3.96 and 55.6 seconds, respectively).

Radon gas (^{222}Rn) comes from the decay of Radium- 226 in the Uranium-238 decay chain, and has a half-life of 3.82 days. It is an emitter of alpha particles with the energy of 5.4 MeV.

Generally, the Earth's crust is the production site of Radon element that is later dispersed within soil particles in a gaseous state which will be released finally to the atmospheric layers. Radon can tolerate this long journey due to its noble chemical properties. It has been estimated that the radon soil content ranges from 10000 Bq/m^3 to 50000 Bq/m^3(WHO, 2009).

The presence of Radon in natural water and air is due to the ingrowths of ^{226}Ra occurring in the geological materials and water. This process, where radon escapes from the solid materials, is called Emanation process. Because radon is a noble gas, it is freed from any chemical bonds associated with it, and can move over a long distance enough to reach the groundwater or the air.

Jordan is rich in uranium deposits found in phosphate rocks being processed into fertilizers. In its decaying to Lead (^{206}Pb), Uranium generates Radon which is a moderately short-lived element with a life-span of approximately 3.825 days. Phosphate mines in Jordan, are located in relatively highly populated areas (e.g. Ruseifa town, a suburb of Zarqa city). They form potential threats to the public health of those residents living in close proximity to phosphate mines. Additionally, there exist other sources of natural and industrial radioactivity scattered throughout Jordan. (Ragheb, 2010).

The residents of Al-Rusaifa city usually depend on groundwater sources that are pumped through the water distribution system for drinking, cooking, and other uses. Furthermore, the area is rich in phosphate ores.

Radon may cause cancer, and may be found in drinking

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water and indoor air. People who are exposed to radon through drinking water may have an increased risk of cancer over the course of their lifetime, especially lung cancer (EPA, 2005). Radon in the soils under homes is the biggest source of indoor radon, and presents a greater risk of lung cancer than the radon in drinking water. As required by the Safe Drinking Water Act, the EPA has developed proposed regulations to reduce radon in drinking water that has a multimedia mitigation option to reduce radon in indoor air (EPA, 2005).

The International Agency for Research on Cancer (IARC) had classified radon (^{222}Rn) as a mutagenic and carcinogenic substance to human beings. It has also been identified as a public health concern when present in drinking water. The World Health Organization (WHO) suggests that radon causes up to 15% of lung cancers worldwide (EPA, 2005)

The Maximum Contaminant Level (MCL) for the concentrations of radon is 11.1 Bq/L (about 300 pCi/L) in drinking water and in dwellings of 150 Bq/m^3 (about 4.05 pCi/L) (EPA, 1986).

2. Material and Methods

2.1. Study Area

The study area is Al-Rusaifah City at Zarqa governorate in central Jordan. It is one of the biggest cities in terms of population density, with more than half a million people living in a 38 km2 area (15,000 people per square kilometer), which makes it the fourth largest city in Jordan (Al-Ruseifah Manucipality, 2015).

Al-Rusaifah is part of the Amman–Zarqa Basin, one of the most important groundwater basins in Jordan. The renewable groundwater averages 88 million cubic meters per year in this basin (Salameh and Bannayan, 1993). The following are the most outcropping lithostratigraphic units in the study area:

1- ***Amman Silicified Limestone Formation (Santonian–Campanian):***

This formation is of varying thickness in different locations of Jordan. It is distinguished by hard, massive chert beds that form a steep cliff above the pale weathering chalks of the Umm Ghudran formation in central and north Jordan (Powell, 1989; Abed, 2009; Diabat, 2015).

2- ***Al-Hasa Phosphorite Formation (Late Campanian–Early Maastrichtian):***

The top of the Amman formation is usually distinguished by the first appearance of the thick phosphorite beds of the Al-Hasa phosphorite formation (Diabat, 2015). This formation consists of a heterogeneous lithology of medium-thick beds of phosphorite, which are intercalated with thin-medium bedded chert, marl, chalky marl, microcrystalline limestone, and oyster-coquina grain stones (Powell, 1989; Abed, 2009; Diabat, 2015).

The thickness of the phosphate beds ranges from 1.25 m to 3 m. The Upper Cretaceous sediments in Al-Rusaifah form a large basin within the generally eastward dipping sequence. Minor asymmetric folds occur, and faults are rare, consisting mainly of step faults (Jallad et al., 1989).

Many factors affect the formation and movement of radon in the ground: uranium content, grain size, and permeability of the host rock and the nature and extent of fracturing in the host rock. These factors also affect the amount of radon in groundwater (Bodansky et al., 1989). Figure (1) shows the study area.

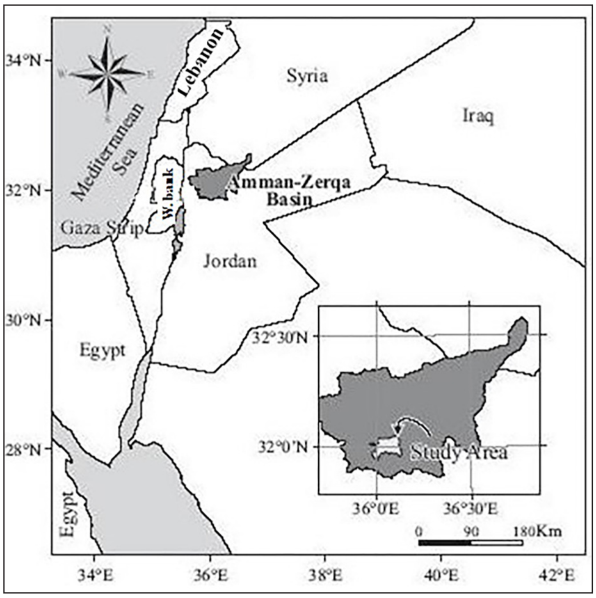

Figure 1. Study area locations.

2.2. Liquid Scintillation Counting

Liquid scintillation counting (LSC) is the most sensitive and widely used technique for detecting and quantifying radioactivity. It is a simple method for determining ^{222}Rn in natural water. Cocktail is first added to the vial, then another 10 ml of water sample was taken with a disposable syringe, ensuring that there is no aeration of the sample, and the water sample is injected at the bottom of the vial, below the immiscible scintillation solution with tightly secured vial's cap. Then the vial was shaken rapidly to ensure that the radon is fully dissolved in the organic solvents, such as toluene. Radon-222 will be extracted from the water phase to the organic scintillate solution. The time and date of mixing the water samples with the liquid scintillation solution will be recorded, and then the mixture will be stored for three hours so that the short-term decay products of ^{222}Rn can grow, attaining a secular equilibrium.

The LSC technique measured the activity of radon and decay products from the rate of photons emitted from the scintillation fluid. In this paper, the Tri-Carb Liquid Scintillation Counter is utilized to determine the counting efficiency of each sample, which then utilized to calculate the disintegration per unit minute (DPM). The main operation principle of the counter is that it depends on the interaction between the emitted beta from the radionuclide and a scintillator, which results into the production of light photons. The generated photon intensity during the scintillation is proportional to the initial energy of the beta particle (PerkinElmer Life and Analytical Sciences, 2008). The scintillation counter can measure photon intensity when the vial containing the radionuclide and the scintillation is placed into the instrument detector.

After that, the light emitted from the sample vials will be amplified via the photosensitive device. The output amplified signal is then converted to pulses of electrical energy, which are recorded as counts. The accumulated counts are then sorted into individual channels (keV) into which the counts are sorted. The sample spectrum is finally produced from these collected and sorted counts. For further details and operation method of the counter, one may refer to (PerkinElmer Life and Analytical Sciences, 2008). From this spectrum, the system is capable to execute distinct count correction calculations and determine counts per minute (CPM) for every sample.

The instrument also determines the counting efficiency of each sample. Also, it compares the quenching index to the quench index for the samples from the so-called quenching curve, in order to obtain the sample counting efficiency and afterwards calculate DPM for the unknown samples (PerkinElmer Life and Analytical Sciences, 2008).

Using this spectrum, the system can perform various count correction calculations and determine counts per minute (CPM) for each sample. To calculate disintegrations per minute (DPM), the instrument will find the counting efficiency of every sample. By utilizing the quench curve, the instrument is capable to compare the quench index for the samples to the quench index for the quench standards to determine sample counting efficiency and subsequently calculate DPM for the unknown samples https://www.proz.com/profile/970169,https://researchpark.spbu.ru/en/equipment-geomodel-eng/2266-geomodel-experimentalnoe-modelirovanie-eng).

2.3. Annual Effective Dose

The obtained annual effective doses (AED) received by the human body for all drinking water samples (in units of mSv/y) are divided into two parts as follows:

1. The annual dose of radiation resulting from ingestion of radon in drinking water (D_{eff}) in terms of mSv/y, measured by using the following formula (Kozlowska et al., 2010):

   $$D_{eff}(mSv/y) = 1000\ A_c W_d\ V$$

   where A_c is the concentration of radon in water in Bq/l; W_d is the radiation dose (conversion factor), which is equal to 1×10^{-8} mSv/Bq; and V is the amount of drinking water consumed by the individual per year, which is equal to 182.5 l/y.
2. The annual dose of radiation resulting from radon which has escaped from drinking water supply, inhalation (H_{eff}) (Kozlowska et al., 2010):

   $$H_{eff} = A\ F\ \gamma f\ d\ Q\ C_w$$

   where A is a constant representing the number of working levels (WL); **F** is the equilibrium factor between radon and its daughters and equal to (0.3); **Y** is the equivalent occupational working months per year for a member of the population allowing for residency time and breathing rate, equal to 18 M/y; **f** is the transfer factor of radon gas from water into the air of the dwelling, which is equal to 10^{-4} in air per 1 Bq/l in water; **d** is the dose conversion coefficient factor of radon progeny in the respiratory region, which is equal to 5 mSv/WLM; **Q** is the quality factor for α-particles, which is equal to 20; and C_w is the radon concentration in water in Bq/L.

To calculate the total annual dose resulting from both the ingestion and inhalation of radon gas from consuming water for drinking and washing purposes is given by $H_{eff}+D_{eff}$.

2.4. Estimating the lung cancer risk factor

Radon gas can diffuse and escape from water during showering or washing into indoor air at a rate of 10^{-4} per 1 Bq/l in water as proposed by Gesell and Prichard (1975). The lung cancer risk factor from natural radiation is assumed to be 36×10^{-4} per 1 pCi/L as proposed by Cross (1992).

3. Results and Discussion

Table 1 presents the overall radon concentration levels and their annual effective dose exposure for each well. The spatial variations in radon concentration could be a function of the geological structure of the area, depth of the water source, and differences in climate and geohydrological processes occurring in the area (PerkinElmer Life and Analytical Sciences, 2008).

Table 1. Minimum and maximum average radon concentrations, average effective dose, and increase of cancer for water samples in Al-Rusaifah City.

Well	No. of samples	C. min (Bq/L)	C. max (Bq/L)	C. average (Bq/L)	AED mSv/y	Increase of cancer
BASATEEN(A1)	3	165.8±17.6	204.6±22.9	191.1±20.8	3.135±0.363	0.0019
BASATEEN(1)	3	171.8±19.1	188.9±20.9	182.4±19.9	2.992±0.290	0.0018
WELL(8)	3	181.8±21.7	188.9±20.9	186.6±21.3	3.062±0.311	0.0018
MUNICIPAL(4)	3	151.9±16.4	167.6±18.0	159.7±17.2	2.619±0.251	0.0015
OFFICE(2)	3	240.3±25.9	253.1±26.9	248.0±27.4	4.069±0.399	0.0024
WELL(9A)	3	194.3±23.9	209.6±22.6	203.7±22.8	3.007±0.322	0.0020
TOTAL	18			195.3±27.4	3.147±0.399	0.0019

The overall average of radon gas concentration in the drinking water wells sample is 195.3±27.4 Bq/l, where the minimum average radon concentration is 159.7±17.2 Bq/l for the Municipal 4 well water samples, and the maximum average radon concentration is 248.0±27.4 Bq/l for the Office 2 well water samples. The residents of this area are forty times more exposed to radon with higher probabilities of developing cancer or other health-related impacts. This is about 1900 per million inhabitants, which is more than the WHO (2012) estimate of 1000 per million inhabitants. These results are shown in Figure 2.

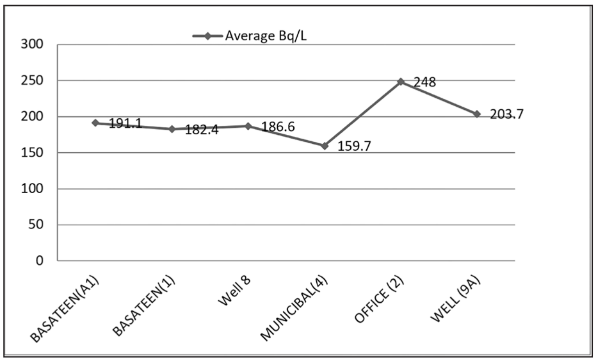

Figure 2. Average radon concentration in Al-Rusaifah City.

Table 2 shows the results of radon concentrations in the present study compared with others. Several national and international health organizations assigned the acceptable action limits for radon concentrations as follows: The EPA (1999) defined a value of 11.1 Bq /l for radon concentration in water; the UNSCEAR (2012) set it at 40 Bq/l; and the WHO (2012) set a value of 100 Bq/l as an action limit.

It seems that our results are much greater than the results obtained by Al-Bataina et al. (2003), Ismail et al. (2008), Aleissa et al. (2012), Rafat (2013), and Mohsen et al. (2016).The high radon concentration values in our study could be attributed to the presence of phosphate mines, which plays a major role in providing an enormous source of radon in the study area. Another plausible source of difference is the use of various detectors in each study.

Table 2. Comparison of 222Rn levels in spring water with results from other workers.

Sample number	Country	Year	Average radon concentration Bq/l	Detection method	Reference
1	Jordan (Al-Karak)	2003	1.854±0.17	CR-39	AL-Bataina et al. (2003)
2	KSA (Riyad)	2012	1.01±0.10	LSC	Aleissa et al. (2013)
3	Iraq (Irbil)	2007	4.693±2.213	CR-39	Ismail et al. (2007)
4	Libya (South Region)	2013	3.46±1.76	CR-39	Rafat (2013)
5	Saudi Arabia (Buraydah City)	2016	4.73±0.84	ϒ -ray spectrometry	Mohsen et al. (2016)
6	Our study	2016	195.3±27.4	LSC	Current study

4. Conclusions and Recommendations

Eighteen groundwater samples from wells collected from Rusaifah City were investigated for ^{222}Rn concentrations through LCS. It is found that these samples exhibited higher radon concentrations in water than the maximum contamination level of 11.1 Bq/l, as recommended by the EPA (2005).

The highest concentration of radon gas in groundwater in Rusaifah was found in Office 2 (253.1 Bq\l), and the lowest concentration level was in Municipal 4 (151.9 Bq/l). For all samples, the total average of the annual effective doses was found to be larger than the maximum value of the general annual effective dose (0.1 mSv/y) according to the WHO (2004) and the European Council (EC, 2001).

Based on the results, the groundwater in the study area poses a risk to residents, as they are exposed to radon radiation via drinking water (ingestion and inhalation). We recommend that the water of the Rusaifah wells, which contains radon gas with free water, be diluted before distributing it to the people. In addition, we recommend the aeration and filtration of water after pumping from the well and before it is distributed to the public network and the use of treatment plants such as reverse osmosis and/or other electrodialysis processes along with aeration. Finally, more epidemiological public health studies in the area are recommended to find any potential linkage between radon concentrations and incidences of cancer among the inhabitants.

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