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## APPLICATION OF NATURAL RADIOACTIVITY FOR HYDROGEOLOGICAL AND ENVIRONMENTAL ASSESSMENT OF GROUNDWATER IN RAS AL-KHAIMAH, UAE

Mohammed Nasser Abdullah Al-Saqri



March 2022

## United Arab Emirates University

College of Science

Department of Biology

## APPLICATION OF NATURAL RADIOACTIVITY FOR HYDROGEOLOGICAL AND ENVIRONMENTAL ASSESSMENT OF GROUNDWATER IN RAS AL-KHAIMAH, UAE

Mohammed Nasser Abdullah Al-Saqri

This thesis is submitted in partial fulfilment of the requirements for the degree of Master of Science in Environmental Sciences

March 2022

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Cover: Collecting groundwater samples from an irrigation well.

(Photo: By Ibrahimullah Mohd Azim Khan)

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### **Declaration of Original Work**

I, Mohammed Nasser Abdullah Al-Saqri, the undersigned, a graduate student at the United Arab Emirates University (UAEU), and the author of this thesis entitled "*Application of Natural Radioactivity for Hydrogeological and Environmental Assessment of Groundwater in Ras Al-Khaimah, UAE*", hereby, solemnly declare that this thesis is my own original research work that has been done and prepared by me under the supervision of Dr. Dalal Matar AlShamsi, in the College of Science at UAEU. This work has not previously formed the basis for the award of any academic degree, diploma, or a similar title at this or any other university. Any materials borrowed from other sources (whether published or unpublished) and relied upon or included in my thesis have been properly cited and acknowledged in accordance with appropriate academic conventions. I further declare that there is no potential conflict of interest with respect to the research, data collection, authorship, presentation and/or publication of this thesis.

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#### Abstract

Groundwater includes a certain concentration of natural radioactive isotopes of uranium (U) and its decay products (daughters) like radon (Rn). Defining the concentration levels, spatial distribution, and possible environmental impact of these isotopes in groundwater is vital for sustainable groundwater resources in the United Arab Emirates (UAE). This dissertation focuses on documenting the distribution and determining the probable environmental impact and sources of Uranium-235 (<sup>235</sup>U), Uranium-238 (<sup>238</sup>U), and Radon-222 (<sup>222</sup>Rn) in groundwater in the Northern part of the UAE and specifically in the Wadi Al Bih aguifer in Ras Al Khaimah Emirate. The sampled wells occur at different distances from the coast, where some are very close to the coastal line, and some exist in the center of Wadi Al Bih. A variety of techniques including ICP-MS, ICP-OES, and RAD7, were used for the analyses. The results reveal comparable activity concentration in the measured radioactivity in terms of spatial and local variability. All the <sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn concentrations in the measured groundwater samples are below the World Health Organization permissible limit for drinking water. The occurrence of <sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn in the measured samples suggest a geochemical interaction between the aquifer's lithology and water. In some wells, seawater intrusion is expected to be an additional source of uranium and elevated Total Dissolved Solids (TDS). The calculated radioactivity annual effective doses of inhalation and ingestion were below the maximum permissible annual dose limits defined by the WHO. The probable -uraniumrelated- cancer mortality and morbidity were also calculated and found to be not hazardous following the permissible limits determined by the United States Environmental Protection Agency (EPA) and International Commission on Radiological Protection (ICRP).

Keywords: Radioactive isotopes, carbonate aquifer, uranium, radon, UAE.

### **Title and Abstract (in Arabic)**

## استخدام النظائر المشعة الطبيعية في التقييم الهيدروجيولوجي و البيئي للمياه الجوفية في إمارة رأس الخيمة، دولة الإمارات العربية المتحدة.

#### الملخص

تحتوي المياه الجوفية على تركيز معين من النظائر المشعة الطبيعية لليورانيوم ونواتج إشعاعه مثل الرادون. يعد تحديد مستويات التركيز والتوزيع المكاني والتأثير البيئي المحتمل لهذه النظائر في المياه الجوفية أمرًا مهمًا لاستدامة موارد المياه الجوفية في دولة الإمار ات العربية المتحدة. تركز هذه الرسالة البحثية على تحديد التوزيع المكانى والتأثير البيئي المحتمل ومصادر اليورانيوم والرادون في المياه الجوفية في الجزء الشمالي من دولة الإمارات العربية المتحدة وتحديداً في خزان وادى البيح الجوفي في إمارة رأس الخيمة. تتوزع الآبار التي تم اختبار عيناتٍ مائية منها على مسافات مختلفة من الساحل، حيث يقع بعضها قريبًا جدًا من الخط الساحلي ويقع البعض الآخر في وسط وادى البيح. وقد تم استخدام مجموعة متنوعة من التقنيات بما في ذلك ICP-OES و ICP-OES و RAD7 للتحاليل الكيميائية، وكشفت النتائج عن تراكيز منخفضة نسبيًا في النشاط الإشعاعي، حيث كانت جميع تراكيز اليورانيوم-235 واليورانيوم-238 والرادون-222 في عينات المياه الجوفية المقاسة أقل من الحد الأعلى المسموح به الذي قررته منظمة الصحة العالمية. ويشير وجود اليورانيوم-235 واليورانيوم-238 والرادون-222 في العينات المقاسة إلى تفاعلات جيوكيميائية بين صخور الخزان الجوفي والمياه التي يحتويها. كما يتوقع أن يكون تداخل مياه البحر في الخزانات الجوفية مصدرًا إضافيًا لليور إنيوم والأملاح المذابة في المياه الجوفية. وقد تم حساب الجرعات السنوية من النشاط الإشعاعي والتي يسببها الاستنشاق والابتلاع وكانت كمية الجرعات المحسوبة أقل بكثير من الحد الأقصى للجرعة السنوية المسموح بها التي حددتها منظمة الصحة العالمية، وتم أيضًا حساب احتمالات الوفيات واحتمالات الإصابة بمرض السرطان نتيجة التعرض لليورانيوم، ووجدت أنها بعيدة عن الحدود الخطرة التي حددتها وكالة حماية البيئة الأمريكية واللجنة الدولية للحماية الإشعاعية.

مفاهيم البحث الرئيسية: النظائر المشعة ، الخزان المائي الجيري ، اليورانيوم ، الرادون ، الإمارات العربية المتحدة.

#### **Author Profile**



Mohammed Al-Saqri is currently a master's student in the Environmental Sciences program at the College of Science at the UAE University. He was a Research Assistant at the Department of Geosciences for two years (2019-2021). He has a membership in the Club of Sustainability at the College of Science. He received his bachelor's degree in Geology of Petroleum and Water from the same University. He worked as an International Drilling and Measurements Field Engineer in the leading oilfield service company (Schlumberger) for one year in Tanzania and Ethiopia. Mohammed is living in Abu Dhabi and is a senior volunteer in the UAE Volunteers Platform. He accomplished more than 300 volunteering hours in different medical, social, and sports events across Abu Dhabi. He has many personal interests that include reading, nature photography, biking, swimming, and playing football.

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أود أن أتوجه بجزيل الشكر والامتنان والعرفان لكل من ساندني وساعدني للوصول إلى هذه المرحلة من مسيرتي التعليمية. وأخص بالذكر والدي العزيز ناصر ووالدتي العزيزة سلوى حفظهما الله وأدام عليهما موفور الصحة والعافية، وإخوتي وأخواتي وجميع أفراد عائلتي وكافة آل الصقري.

أساتذتي المشرفين، الدكتورة دلال مطر الشامسي التي استمرت بمتابعتي في جميع التفاصيل، بالتوجيه والدعم في مجال البحث العلمي وطرق البحث الحديثة والتقييم العلمي لأفكار البحث ونتائجه، والأستاذ الدكتور علاء الدهان الذي أنار بصيرتي نحو آفاق علمية وبحثية عالية المستوى، أخذتني نحو الاستمتاع بالعمل البحثي وفترة الدراسات العليا.

أصدقائي الأعزاء، كان لهم الكثير من الفضل في تشجيعي على مواصلة مسيرتي التعليمية، لقد قدموا لي كافة أشكال الدعم المعنوي. أتوجه بجزيل الشكر لصديقي الدكتور أحمد بن حسين الحارثي على نصائحه وتوجيهاته القيمة في مجال البحث العلمي. أيضاً أشكر كل من حسين بن طلان الحارثي و عبد العزيز بن عمير الحارثي على دعمهما اللامحدود لي. شكر خاص لصديق طفولتي مالك بن جريبة الحارثي على وقوفه معي في أصعب اللحظات التي مررت بها. كذلك شكر وامتنان خاص لأصدقائي تشجيعهم ودعمهم لي وأذكر منهم -على سبيل المثال لا الحصر - محمد بن عوض وصالح بن عوض وحسين بن علي وحسين بن سالم ومتعب بن حيدر وناجي بن مساعد وصالح بن حيدر وحسين بن صالح ومنصور بن حيدر وحسن بن صالح.

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## Dedication

To my beloved Parents, sisters, brothers, family, and friends To my special ones Awadh, Dada, Membes, Heidi, Salhuf, and Sulhufa

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## List of Abbreviations

U	Uranium		
<sup>238</sup> U	Uranium-238		
<sup>235</sup> U	Uranium-235		
Rn	Radon		
<sup>222</sup> Rn	Radon-222		
Bq L <sup>-1</sup>	Becquerel per Liter		
TDS	Total Dissolved Solids		
$Na^+$	Sodium		
$K^+$	Potassium		
Cl-	Chloride		
<b>SO</b> <sub>4</sub> <sup>2-</sup>	Sulfate		
$E_{w.Ig} \\$	Effective Ingestion Dose		
$E_{w.Ih} \\$	Effective Inhalation Dose		
UAE	United Arab Emirates		
ICP-MS	Inductively Coupled Plasma Mass Spectrometry		
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry		
WHO	World Health Organization		
EPA	The United States Environmental Protection Agency		
ICRP	International Commission on Radiological Protection		

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# **Chapter 1**

### **Chapter 1: Introduction**

#### 1.1 Radioactive Isotopes Significance in Hydrogeology

Radioactive elements occur almost everywhere in the environment. Their origin might be either natural or man-made (anthropogenic). They are chemically unstable and releasing different radiation particles upon decay. Uranium-238 (<sup>238</sup>U) decay chain -for instance-, starts with the parent isotope (<sup>238</sup>U) and continues to produce daughters and different modes of energies until it reaches the stable isotope of Lead-206 (<sup>206</sup>Pb) which marks the end of this decay chain (Figure 1). The natural radioactive elements are produced through different pathways such as primordial which are linked to the creation of Earth, cosmogenic that are generated in the atmosphere, and those that are forming due to chain-series decay. The decay of natural daughters' nuclides brings more than 80% of the entire effective radiation dose to the environment and is considered the main source of radiation hazards (Dinh Chau et al., 2011). Some of the anthropogenic radioactive isotopes are produced through human activities such as nuclear bomb tests, reprocessing of nuclear fuel, medical uses, and the excess use of fertilizers. Anthropogenic short-lived radionuclides are of special concern due to their fast decay and high emitted energy in a short time which might expose the ecosystem to extreme pollution (EPA, 2022). Natural and anthropogenic radioactive isotopes in the environment might cross the living organisms' bodies through inhalation and ingestion (WHO, 2011). It is, therefore, significant to study these radioactive isotopes in the ecosystem to preserve the balance of sustainability and to assess the possible negative environmental impacts.



Figure 1: Uranium-238 decay chain (https://www.nachi.org/gallery/radon/uranium-238-decay-chain)

#### **1.2 Natural Radioactive Isotopes and Possible Environmental Impacts**

There are three major decay chains producing the natural radionuclides isotopes, which are  ${}^{238}$ U,  ${}^{235}$ U, and  ${}^{232}$ Th decay chains. The focus of this study is on the parent  ${}^{238}$ U and its daughter  ${}^{222}$ Rn (Figure 1). Uranium has three natural isotopes which are  ${}^{238}$ U (T<sub>1/2</sub> = 4.468 × 10<sup>9</sup> years, abundance: 99.27%),  ${}^{235}$ U (T<sub>1/2</sub> = 7.04 × 10<sup>8</sup> years, abundance: 0.72%) and  ${}^{234}$ U (T<sub>1/2</sub> = 245500 years, abundance: 0.005%) (Rogers & Adams, 1969). Radon-222 ( ${}^{222}$ Rn, T<sub>1/2</sub> = 3.8 days) is produced from the decay chain of  ${}^{238}$ U (Baskaran, 2016). The gas form of  ${}^{222}$ Rn makes the isotope occurrence in air, soil, rocks, and water. A high concentration of these isotopes might affect human health if the quantity exceeds the permissible limits of ingestion or inhalation (Table 1).

Table 1: Permissible limits for radioactivity level in groundwater recommended by the World Health Organization (WHO, 2017; WHO, 2011)

Component	WHO permissible limit of radionuclides concentration in drinking water	WHO permissible annual effective dose of radionuclides (mSv/y)
<sup>235</sup> U	1 Bq L <sup>-1</sup>	0.1
<sup>238</sup> U	10 Bq L <sup>-1</sup>	0.1
TotalU	60 μg L <sup>-1</sup> (60000 ng L <sup>-1</sup> )	-
<sup>232</sup> Th	1 Bq L <sup>-1</sup>	0.1
<sup>222</sup> Rn	100 Bq L <sup>-1</sup>	0.1
<sup>226</sup> Ra	1 Bq L <sup>-1</sup>	0.1

*Note:* Bq L<sup>-1</sup>: Becquerel per Liter,  $\mu$ g L<sup>-1</sup>: Microgram per Liter, ng L<sup>-1</sup>: Nanogram per Liter, mSv/y: Millisievert per year

The radioactive isotopes possible impacts might appear either in the short or long-term depending on the consumed dose. For example, in the case of a nuclear accident, the adverse impacts on the environment might be observed relatively faster due to the higher dose transferred to the ecosystem, and this negative impact might affect wide areas depending on the radioactive isotope's ability to stay in the atmosphere or hydrosphere and this would negatively affect the biodiversity, ecosystem natural balance, pollution state, and human health. Thus, the radiological environmental impact becomes a global concern. Radioactive isotopes are used in tracing the groundwater quality in aquifers. The isotopes are selected depending on their half-life, probability of occurrence in certain aquifer lithology, and significant environmental impact. These isotopes can also be used to trace the groundwater sources and recharge conditions (Ji et al., 2020; Xiong et al., 2020). In addition, a relatively high concentration of some radioactive isotopes in groundwater may cause health hazards, where they might enter the human body through inhalation or ingestion. Therefore, categorizing the activity of these isotopes in groundwater will be significant in defining the environmental effect and its relation to aquifer type, sea-water intrusion, and farming activities effect (Alshamsi, Hussien, Aldahan, & Murad, 2020).

#### 1.3 <sup>238</sup>U and <sup>222</sup>Rn Occurrence in Groundwater

The major sources of <sup>238</sup>U and <sup>222</sup>Rn in groundwater are through natural processes (due to rock-water interaction) or anthropogenic such as fertilizers infiltration in soil or industrial by-products wastes. Additional factors that may affect the <sup>238</sup>U and <sup>222</sup>Rn occurrence in groundwater are aquifer lithologic type, water residence age, aquifer depth, and annual recharge conditions. Uranium concentrations vary in different rocks: in shale (3.7 ppm), granite (3.0 ppm), and carbonate rocks (2.2 ppm) (Faure, 1998; Dinh Chau et al., 2011). Also, the annual rainfall might influence the concentration of uranium, particular in shallow aquifers, and this is observed more clearly when comparing these concentrations between arid and humid regions (Alshamsi, Murad, Aldahan, & Hou, 2013), where the rainfall could dilute the concentration of radioactive isotopes. The radioactive isotopes concentration in water depends also on their half-life and the contact time of the source rocks to the water which is called residence time in the aquifer. In general, as half-life and contact time increases the concentration of radioactive isotopes increases, and as the radioactive decay proceeds more decay products (daughters) of the original radioactive isotope will be found in the groundwater particularly if soluble in the existing conditions

(Adithya, Chidambaram, Keesari, Mohokar, & Prasanna, 2019). The pH (hydrogen ions activity) of groundwater may change due to the aquifer's lithology, vadose zone soil composition, and organic matter content (Jardine, McCarthy, and Weber, 1989). The interplay between pH and solubility of uranium is indicated by the formation of uranyl ions in natural alkaline waters (pH> 7), resulting in elevated uranium solubility, especially in oxidizing conditions where uranium exists in the hexavalent state U(VI) (Guerrero et al., 2016; Qiao, Hansen, Hou, Aldahan, & Possnert, 2012). The uranyl-carbonate complexes generally stay in the liquid state rather than adsorbed to rock grains surfaces. Therefore, these uranyl-carbonate complexes might raise the concentration of dissolved uranium in groundwater (Elless & Lee, 1998).

#### **1.4 Research Objectives**

There is no doubt that isotope technology represents a vital tool for the tracing of natural and anthropogenic processes in the Earth's surface archives (sediment, soils, and water). Among these isotopes, the naturally occurring radioactive isotopes such as <sup>238</sup>U and <sup>222</sup>Rn provide significant tools for information related to the rock-groundwater interaction and the consequent impact on the environment. The applications of these isotopes to explore recharge sources, environmental impact and effects of climate have been used in the UAE (Alshamsi et al., 2013; Murad et al., 2014; Zheng et al., 2016). New investigations are needed to expand the understanding of processes with respect to the UAE's different aquifers and recharge pathways. Among regions that are depending on groundwater as a natural water resource is the Ras Al Khaimah Emirate. Evaluation of natural radioactivity in groundwater of this region is lacking and the data from this study will provide new information to decision-makers. Consequently, in the thesis presented here, analyses of natural radioactivity and other

parameters of the groundwater were performed with the following objectives:

- Acquisition of data on the major sources of radioactivity including <sup>238</sup>U and <sup>222</sup>Rn.
- 2- Identifying spatial variability of the radionuclides and their distribution patterns.
- 3- Establishing factors that control the interplay between physiochemical and geological parameters and their effects on the radionuclide's distribution.
- 4- Estimating possible environmental-ecological effects of the radionuclides in different uses of groundwater.

# Chapter 2

## **Chapter 2: Sampling Sites and Analytical Techniques**

#### 2.1 Study Area

The study area is in Ras Al Khaimah Emirate in the northern part of the UAE. Most of the groundwater samples have been collected from the Wadi Al Bih area. While only two samples have been collected from the Sham area in the northern part of Ras Al Khaimah Emirate nearby the coastal line (Figure 2). The Wadi Al Bih Aquifer in Ras Al Khaimah is considered the main source of groundwater for drinking water, agricultural activities, and domestic uses. Overexploitation, fewer rainfalls, climatic conditions, and seawater intrusion, all these factors led to the decline of the water table and the quality of the groundwater in terms of salinity.



Figure 2: The sampled wells in the study area

The Wadi Al Bih Aquifer consists of two main units. The upper unit is a gravel layer that contains loose, coarse, and high permeable alluvial of carbonate origin. While the lower unit is mainly fractured limestone (Murad, 2014). The age of limestone ranges from Permian to Triassic. The area of recharge catchment of the limestone aquifer is 475 km<sup>2</sup> at an elevation of about 1,050 m above sea level (Abu Al Enien, 1996).



Figure 3: The groundwater flow direction (white arrows) showing the south-east (recharge zone) towards the north-west ends in Arabian Gulf (discharge zone).

The study area in the Wadi Al Bih is part of Ru'us Al Jibal Massif of the Oman Mountains, which are located on the southeast edge of the Arabian Peninsula. According to Glennie et al. (1974), the Ru'us Al Jibal Group (the age from Permian to Middle Triassic) is located on the bottom of the Hajar Supergroup, overlaid by the Elphinstone Group (the age from Middle to Upper Triassic) in middle, and Musandam Group (the age from Early Jurassic to Lower Cretaceous) on the top. The thickness of these three groups is about 3500 m and forms the surface and subsurface of the Wadi Al Bih drainage basin. The Ru'us Al Jibal Group consists of Bih, Hagil, and Ghail formations (Figure 3). The structural nature of the Wadi Al Bih contains thrust faults and folds that allow surface water to move horizontally or percolate to subsurface layers (Figures 2-3).

The area of the Wadi Al Bih covers 483 km<sup>2</sup> which gives credit to this wadi being the largest drainage basin in the United Arab Emirates (UAE). Thin soil and little plants are covering the surface of the Wadi Al Bih. The topography is rugged with sharp slopes, so the risk of flash flood hazards is high (Al Assam, Al Matari, Garamoon, & Suwaid, 2005).


Figure 4: Geological map of the study area

### 2.2.1 Sampling and Field Measurements

Groundwater was sampled from thirty wells in the study area (Figure 2). Each well was left to pump for at least 20 minutes before sampling to ensure representation of the aquifer water. The amount of water sampled for each analytical procedure is described in the measurement of the entities. Temperature, pH, and Total Dissolved Solids (TDS) were measured directly in the field using WTW-COND-3301 (Figure 5). The error was not exceeding 5% and the accuracy was  $\pm$  one digit.



Figure 5: WTW-COND-3301 portable instrument to measure physiochemical parameters in-situ

# 2.2.2 <sup>222</sup>Rn Measurements

The radon-222 activity was measured in-situ for the collected water samples from 30 wells in Wadi Bih and Sham areas in Ras Al Khaimah, using Rad7 (an electronic portable radon detector, from Durridge Co., USA) (Figure 6). The samples were filled in a 40mL vial and then analyzed at the same time by connecting the vial by a Rad7 H<sub>2</sub>O accessory. The Rad7 was placed in the car bag where air conditioning is turned on, to keep the humidity in the permissible range of analysis which must be less than 10%. The measurements were performed in four cycles besides one cycle for purging. After that, the average value of radon concertation was recorded for the four cycles in Bq/m<sup>3</sup> and then converted into Bq L<sup>-1</sup> in order to be compared with World Health Organization (WHO) standards. Also, decay correction was applied based on the period between the time of sampling and actual measurements. The analytical accuracy of the measurement was calculated using three executive measurements and estimated at 5%.



Figure 6: Configuration of the <sup>222</sup>Rn measurement in-situ cabin and the Rad7 instrument



# 2.2.3 <sup>235</sup>U and <sup>238</sup>U Measurements

The measurement of <sup>235</sup>U, <sup>238</sup>U, and Cl<sup>-</sup> was performed at the Center for Nuclear Technologies of the Technical University of Denmark. The procedure was described in Alshamsi et al. (2013) and includes the addition of 0.20 mL of 100 mg m L<sup>-1</sup> in (III) (as InCl<sub>3</sub>) as internal standard and 10 times dilution with 3% HNO<sub>3</sub> (super pure). The preparation of standards was done by using a similar method as samples by dilution of uranium and chloride standard solutions (purchased from National Institute of Standard Technology, USA) with 3% HNO<sub>3</sub> (super pure). Indium solution, as an internal standard, was also added to the standard solution. The concentrations of requested analytes (e.g. <sup>238</sup>U, <sup>235</sup>U, and Cl<sup>-</sup>) and internal standard (<sup>115</sup>In) in the samples and standards were measured using an inductively coupled plasma mass spectrometry (ICP-MS) system (X Series<sup>II</sup>), Thermo Fisher Scientific, Waltham, MA) equipped with an Xtskimmer cone and a concentric nebulizer under hot plasma conditions. The concentrations of <sup>235</sup>U, <sup>238</sup>U, and Cl<sup>-</sup> in the water samples were computed by correlating with standard and then corrected for introduction efficiency using indium internal standard. The detection limits calculated as three times the standard deviation  $(3\sigma)$  of the processing blank are 0.21 mg L<sup>-1</sup> for Cl, 0.37mBq L<sup>-1</sup>, for <sup>235</sup>U, and 0.95 mBq L<sup>-1</sup> for <sup>238</sup>U. A 0.5 mol/L HNO<sub>3</sub> solution was used as a washing solution among consecutive assays. The accuracy estimate is  $\pm 2.5\%$ , and the precision is around 0.5%.

## 2.2.4 $Na^+$ and $K^+$ Measurements

The analyses of Na<sup>+</sup> and K<sup>+</sup> were performed using inductively coupled plasma optical emission spectroscopy (ICP-OES-Varian 715 instrument) in the laboratories of Geosciences at UAE University. Each water sample was injected into the nebulizer and spectral analysis of every element was standardized by a multi-element standard solution GSC-CAL-8 offered by Inorganic Ventures. The analytical error of the measured samples is <5%, and the detection limit ranges between 0.001 and 0.017 mg  $L^{-1}$ .

#### 2.2.5 Cl<sup>-</sup> Measurements

The Cl<sup>-</sup> was determined using ICP-MS at the Center for Nuclear Technologies of the Technical University of Denmark, where it was injected into the plasma, at a temperature of a few thousand Kelvin, and the chloride was atomized and ionized to cation, and at last measure. The detection limits of the ICP-MS range at (0.01 - 0.6) ng L<sup>-1</sup> (Becker, 2003), The accuracy estimate is  $\pm 2.5\%$ .

# 2.2.6 $SO_4^{2-}$ Measurements

The anion of sulfate was measured by high-performance ion liquid chromatography (HPILC) at the Center for Nuclear Technologies of the Technical University of Denmark. Measurement's error calculated at 1 standard deviation is <5%, and the detection limit ranges at (0.01 to 0.2) mg L<sup>-1</sup>.

## 2.2.7 Statistical and Mapping Techniques

Pearson linear correlation and factor analysis statistical techniques were used for finding relationships between the different parameters. The ArcGIS software has been used for mapping of data distribution.

# Chapter 3

# **Chapter 3: Results**

### **3.1 General Properties of Groundwater**

The pH, temperature, TDS, Na, K, Cl, and  $SO_4^{2-}$  for the analyzed water samples are presented in Table 3.1. The pH shows a range covering neutral to slightly basic values (7.1 - 8.5). The temperature of the water varies from about 29.9°C to 40°C (average 34.7°C), while the TDS values span between 143 mg L<sup>-1</sup> and 10040 mg L<sup>-1</sup> (average 2320.5 mg L<sup>-1</sup>). The distribution of groundwater TDS in the investigated area is illustrated in Figure 7. Variability in Na<sup>+</sup> spans between 20.9 mg L<sup>-1</sup> and 938.4 mg L<sup>-1</sup> (average 329.9 mg L<sup>-1</sup>), while the K<sup>+</sup> range at (2.88 – 31.09) mg L<sup>-1</sup> with averages of 10.49 mg L<sup>-1</sup>. The Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup> range at (33.1 - 3834) mg L<sup>-1</sup> and (0.5 – 1862.4) mg L<sup>-1</sup> with averages of 953.09 mg L<sup>-1</sup> 581.21 mg L<sup>-1</sup>, respectively.



Figure 7: The TDS distribution in the investigated area

16	15	14	13	12	11	10	6	8	7	6	S	4	3	2	1	Sample #
R-KH17	R-KH16	R-KH15	R-KH14	R-KH13	R-KH12	R-KH11	R-KH10	R-KH08	R-KH07	R-KH06	R-KH05	R-KH04	R-KH03	R-KH02	R-KH01	Sample ID
407252	406153	406728	405789	405233	405935	403196	403377	408561	405098	404555	404577	403887	402635	403589	403711	UTM (E)
2852160	2853900	2853143	2852053	2851739	2850976	2851429	2850866	2853735	2851144	2851588	2851081	2851103	2849717	2849530	2850942	UTM (N)
7.8	7.5	8.1	7.7	7.3	7.1	7.8	7.7	8.0	7.6	7.2	7.5	7.6	7.1	7.8	8.4	pН
35.80	33.60	35.20	36.50	35.20	36.20	35.60	36.40	33.40	36.80	35.00	36.20	35.70	34.60	35.70	35.40	Temp. °C
414	900	310	2200	2730	1099	1564	1268	238	1596	1690	1800	1330	6400	6600	1510	TDS (mg L <sup>-1</sup> )
133.49	162.24	107.18	595.66	654.02	347.84	475.97	398.49	38.00	522.75	558.49	453.86	416.50	46.88	170.67	413.60	Na <sup>+</sup> (mg L <sup>-1</sup> )
5.45	10.33	4.94	13.01	18.00	9.00	11.53	9.76	3.45	11.92	13.31	11.22	10.54	31.09	5.71	10.07	$\mathbf{K}^+$ (mg $\mathbf{L}^{-1}$ )
156.2	319.5	106.5	1420	1491	717.1	986.9	880.4	46.15	1278	1242	994	887.5	3834	170.4	834.3	Cl <sup>-</sup> (mg L <sup>-1</sup> )
197.80	1065.60	104.60	86.40	878.40	460.80	1555.20	497.30	76.80	950.40	12.00	518.40	345.60	163.20	88.30	1862.40	$SO_4^{2-}$ (mg L <sup>-1</sup> )

Table 2: Sampling location and groundwater features including pH, temperature, TDS, Na<sup>+</sup>, K<sup>+</sup>, Cl<sup>-</sup> and  $SO_4^{2^-}$ .

NM:	
not	
measured	

30	29	28	27	26	25	24	23	22	21	20	19	18	17
R117	RBih	R95	R94	R93	R76	R65	R72	R70	R63	R-KH21	R-KH20	R-KH19	R-KH18
400011	410041	405333	409157	408506	411850	408736	406319	405990	404728	402764	411668	411196	409690
2832455	2863022	2863166	2879677	2880284	2858004	2853509	2853673	2851987	2851372	2854872	2858249	2855958	2854120
8.12	7.74	7.69	7.87	7.75	8.5	8.4	8.2	8.3	7.9	7.3	7.9	7.9	7.5
40	31	30.50	29.93	30.03	35.00	34.20	33.00	36.90	35.20	32.40	33.90	38.40	34.10
1395	900	9105	5714	10040	438	295	1049	1707	2840	3955	229	155	143
MM	NM	983.42	77.73	20.90	22.08								
MM	NM	18.88	5.03	3.74	2.88								
MN	NM	2591	120.7	33.1	NM								
MN	NM	1094.40	1084.80	0.50	NM								

Table 2: Sampling location and groundwater features including pH, temperature, TDS, Na<sup>+</sup>, K<sup>+</sup>, Cl<sup>-</sup> and  $SO_4^{2-}$  (continued)

# 3.2 <sup>222</sup>Rn, <sup>235</sup>U, and <sup>238</sup>U Measurements

The results of radon and uranium isotopes as an activity are presented in Table 3.2. The <sup>222</sup>Rn activity values in groundwater samples in the investigated area show high variability with ranges of 2.65 - 66.12 Bq L<sup>-1</sup> (average: 23.25 Bq L<sup>-1</sup>). The distribution of <sup>222</sup>Rn concentration in the groundwater of the investigated area is illustrated in Figure 7. The activity values of <sup>235</sup>U are slightly low between 0.37 mBq L<sup>-1</sup> and 6.94 mBq L<sup>-1</sup> with an average of 2.17 mBq L<sup>-1</sup>, while the activity values of <sup>238</sup>U show high variability with ranges of 7.79 – 147.75 mBq L<sup>-1</sup> (average: 46.09 mBq L<sup>-The Figures 8 and 9 show the distribution of <sup>235</sup>U and <sup>238</sup>U respectively in the investigated area. The calculated Pearson correlation coefficient (R) between <sup>235</sup>U and <sup>238</sup>U is almost equal to one indicating the natural abundance of uranium.</sup>



Figure 8: The distribution of <sup>222</sup>Rn concentration in groundwater of the investigated area



Figure 9: The distribution of <sup>235</sup>U concentration in groundwater of the investigated area



Figure 10: The distribution of <sup>238</sup>U concentration in groundwater of the investigated area

SMP #	Sample ID	UTM (E)	UTM (N)	<sup>222</sup> Rn (Bq L <sup>-1</sup> )	<sup>235</sup> U (mBq L <sup>-1</sup> )	<sup>238</sup> U (mBq L <sup>-1</sup> )
1	R-KH01	403711	2850942	40.96	1.47	31.10
2	R-KH02	403589	2849530	53.56	1.63	34.31
3	R-KH03	402635	2849717	24.35	1.71	35.85
4	R-KH04	403887	2851103	38.77	1.59	34.01
5	R-KH05	404577	2851081	17.11	1.60	33.84
6	R-KH06	404555	2851588	24.21	3.07	65.64
7	R-KH07	405098	2851144	29.33	1.78	37.82
8	R-KH08	408561	2853735	20.76	0.86	18.26
9	R-KH10	403377	2850866	13.38	1.53	32.26
10	R-KH11	403196	2851429	25.69	1.50	31.79
11	R-KH12	405935	2850976	66.12	1.61	34.27
12	R-KH13	405233	2851739	44.31	6.88	146.30
13	R-KH14	405789	2852053	13.48	6.94	147.75
14	R-KH15	406728	2853143	43.35	1.55	32.78
15	R-KH16	406153	2853900	29.48	2.28	48.33
16	R-KH17	407252	2852160	8.89	1.68	35.73
17	R-KH18	409690	2854120	13.25	0.37	7.80
18	R-KH19	411196	2855958	24.61	0.53	11.09
19	R-KH20	411668	2858249	40.92	1.19	25.30

Table 3: Radon and Uranium concentrations in groundwater samples

20	R-KH21	402764	2854872	10.44	3.66	77.68
21	R63	404728	2851372	9.91	NM	NM
22	R70	405990	2851987	5.72	NM	NM
23	R72	406319	2853673	20.05	NM	NM
24	R65	408736	2853509	13.60	NM	NM
25	R76	411850	2858004	31.33	NM	NM
26	R93	408506	2880284	2.65	NM	NM
27	R94	409157	2879677	5.90	NM	NM
28	R95	405333	2863166	12.52	NM	NM
29	RBih	410041	2863022	7.51	NM	NM
30	R117	400011	2832455	5.27	NM	NM

Table 3: Radon and Uranium concentrations in groundwater samples (Continued)

NM: not measured.

# **Chapter 4**

## **Chapter 4: Discussion**

#### 4.1 Correlations and Factor Analyses

#### 4.1.1 Radon-222, Uranium-235 and Uranium-238

In order to quantify the level of the relationships between all measured parameters, a correlation analysis was made using Minitab software (Figures 11-12). The correlation was calculated twice; the first round includes 20 samples and measures the correlation coefficient between <sup>222</sup>Rn, <sup>235</sup>U, <sup>238</sup>U, pH, Temperature (Temp.), Total Dissolved Solids (TDS), sodium  $(Na^+)$ , potassium  $(K^+)$ , chloride  $(Cl^-)$ , and sulfate  $(SO_4^{2-})$ . Whereas in the second round all the 30 samples were included and tested for correlation coefficients between <sup>222</sup>Rn, pH, Temperature (Temp.), and TDS only due to the lack of measurements. Thus, the second round represents a wider range of samples and might point at different values of correlation coefficients. In both correlation rounds, <sup>222</sup>Rn shows no correlation with any of the parameters, because radon behaves independently (Figure 12-13). The correlation coefficients between radon and other parameters were very low, mostly less than  $\pm 0.15$ , except for Cl<sup>-</sup> in the first round (R= -0.26), and temperature in the second round (R=0.26). In general, this pattern may relate to the nature of radon (gas form) that creates independent behavior with respect to the (soluble) measured parameters in groundwater. Such behavior was also described in other studies (e.g. Cho & Choo, 2019; Grolander, 2009).

The factor analysis, which provides a measure of multi-correlation behavior between variables, shows no significant loading of  $^{222}$ Rn, where the highest loading reaches only -0.208. This feature confirms that  $^{222}$ Rn is behaving independently in the groundwater system, most likely due to its gaseous form. The  $^{235}$ U and  $^{238}$ U show a good correlation with sodium (R= 0.62) where and potassium (R= 0.41). The factor analysis (Figure 14) shows

also that uranium and sodium might be traced from a common source. The factor analysis suggests that around 41 % of the variation in the data is explained by factor 1, and 16 % is explained by factor 2 (Table 4).

In factor1, the large positive loadings are found in uranium isotopes ( $^{235}$ U,  $^{238}$ U), sodium (Na<sup>+</sup>) and potassium (K<sup>+</sup>), where  $^{235}$ U and  $^{238}$ U fit 0.74 and 0.74, respectively, while sodium (Na<sup>+</sup>) and potassium (K<sup>+</sup>) fit 0.69 and 0.88, correspondingly. So, factor 1 might describe in general the paragenesis of uranium, (Na<sup>+</sup>) and potassium (K<sup>+</sup>). They might be sourced partially from the chemically weathered feldspars in the Northeast. In factor2,  $^{235}$ U and  $^{238}$ U fit 0.58 and 0.58, successfully, which shows large positive loadings on factor 2. Thus, factor 2 is probably describing a source of uranium isotopes, which might be dissolution from carbonate rocks in the aquifer body to form uranyl complexes. The sulphate (SO<sub>4</sub><sup>2-</sup>) has no correlation with uranium, however, it has large positive loading on factor 3 where it fits (0.87), so factor 3 may describe gypsum occurrence and dissolution in the aquifer body.



Figure 11: Correlation coefficients between the measured parameters in the collected water samples (from sample #1 to sample #20), calculated using Minitab 19

Correla	tions								
	<sup>222</sup> Rn	<sup>235</sup> U	<sup>238</sup> U	рН	Temp	TDS	Na ⁺	<b>K</b> +	Cl-
<sup>235</sup> U	-0.048								
<sup>238</sup> U	-0.048	1.000							
рН	0.021	-0.312	-0.312						
Temp	0.124	-0.031	-0.032	0.154					
TDS	0.134	0.268	0.265	-0.374	-0.121				
Na⁺	-0.118	0.642	0.642	-0.326	-0.064	0.232			
K⁺	-0.104	0.450	0.447	-0.604	-0.178	0.668	0.445		
Cl-	-0.257	0.362	0.359	-0.617	-0.238	0.651	0.448	0.973	
<b>SO</b> <sub>4</sub> <sup>2-</sup>	0.107	0.001	-0.000	0.210	-0.270	-0.091	0.366	0.119	0.084
SO <sub>4</sub> <sup>2-</sup>	0.107	0.001	-0.000	0.210	-0.270	-0.091	0.366	0.119	0.084

Figure 12: Correlation matrix between the groundwater parameters and good correlation coefficients values (in bold) were considered in the outcome







Figure 14: The factor analysis loading plot, including samples #1 to #20

Variable	Factor1	Factor2	Factor3	Factor4	Factor5	Factor6	Factor7	Factor8	Factor9
Rn-222	-0.208	-0.106	0.031	0.959	0.035	0.088	0.048	0.121	0.000
U-235	0.737	0.576	-0.239	0.118	0.161	-0.127	0.109	-0.034	0.006
U-238	0.735	0.578	-0.240	0.118	0.162	-0.125	0.109	-0.033	0.006
pН	-0.697	0.304	0.245	-0.098	-0.041	-0.565	0.026	0.174	-0.012
Temp	-0.286	0.123	-0.608	0.088	-0.720	-0.061	0.052	-0.043	0.002
TDS	0.618	-0.514	-0.053	0.241	0.003	-0.440	-0.288	-0.132	0.001
Na <sup>+</sup>	0.692	0.466	0.206	-0.017	-0.245	0.184	-0.387	0.133	-0.017
$K^+$	0.878	-0.367	0.093	-0.053	-0.135	-0.028	0.238	0.052	-0.072
Cl-	0.863	-0.383	0.096	-0.143	-0.174	-0.043	0.125	0.165	0.065
(SO <sub>4</sub> ) <sup>2-</sup>	0.092	0.261	0.871	0.160	-0.311	-0.018	0.123	-0.165	0.013
Variance	4.0768	1.6147	1.3654	1.0716	0.7780	0.5929	0.3496	0.1409	0.0102
% Var	0.408	0.161	0.137	0.107	0.078	0.059	0.035	0.014	0.001

Table 4: Unrotated factor loadings and communalities (Samples #1 to #20), bold values represent the significantly loading parameters in that factor

## 4.1.2 TDS

For samples, #1 to #20; TDS and the other measured physiochemical parameters and major ions were tested for correlation. The variables most strongly correlated with TDS were potassium (K<sup>+</sup>) and chloride (Cl<sup>-</sup>), where correlation coefficients of both are R= 0.65. It is worth mentioning the correlation between (K<sup>+</sup>) and chloride (Cl<sup>-</sup>) where R=0.97, which is the most significant value in the whole correlation between all parameters tested in this study. This feature might indicate that the TDS is sourced mainly from potassium chloride. The TDS is not correlated to Na<sup>+</sup> (R= 0.23), and Na<sup>+</sup> is also weakly correlated to Cl<sup>-</sup> (R=0.45), which indicates lower concentrations of sodium chloride but a higher concentration of potassium chloride. To determine possible sources of lower sodium and higher potassium, the Na<sup>+</sup>/Cl<sup>-</sup> ionic ratio and molar ratios in groundwater were calculated in 19 samples, and it ranges at (0.01- 1.01) for the ionic ratio and at (0.019- 1.55) for the molar ratio, where 13 samples have Na<sup>+</sup>/Cl<sup>-</sup> molar

ratio less than 0.86, 4 samples have  $Na^+/Cl^-$  molar ratio >1 and two samples are between 0.86 and 1. It is expected that when this molar ratio is less than 0.86, then the groundwater is probably mixed with seawater (Naily, 2018), whereas fresh water will have a molar ratio >1, and the values between 0.86 and 1 will represent the transition zone (Vengosh & Rosenthal, 1994). The mixing with seawater might be before the aquifer recharge, especially if the aquifer area was close to the coastal area, and this will also cause a similar effect of sea intrusion, and the Na<sup>+</sup>/Cl<sup>-</sup> the ratio will be less than unity (Srivastava, 2019). Relatively lower sodium and higher potassium and chloride concentrations in Wadi Bih aquifers were also previously reported in the literature and assigned to seawater mixing (Rizk, Alsharhan, & Wood, 2007). Potassium and chloride in general might be sourced from groundwater interaction with the aquifer body and specifically feldspars and clay minerals whose surface commonly includes  $K^+$  and  $Cl^-$  (Khan, 2018; Murad, Mahgoub, and Hussein, 2012). Sodium (Na<sup>+</sup>), chloride (Cl<sup>-</sup>), and potassium  $(K^+)$  were selected to be focused on when analyzing the correlative parameters with TDS as they are significant in identifying the source of salinity in the groundwater. By looking at factor analysis, the TDS fit 0.62, sodium (Na<sup>+</sup>) fit 0.69, potassium (K<sup>+</sup>) fit 0.88, and chloride (Cl<sup>-</sup>) fit 0.86, that indicates TDS, sodium (Na<sup>+</sup>), potassium ( $K^{+}$ ), and chloride (Cl<sup>-</sup>) together have large positive loadings on factor1. This loading effect is attributed to the salinity sources like evaporites. For samples #21 to#30, the TDS was mostly correlated to the temperature (Figure 13) and the largest positive loading on factor 1 (0.83) occurs in the temperature, and the largest negative loading on factor 1 (-0.84) occurs in the TDS. The largest negative loading on factor 2 exists in the pH (-0.827), and the largest positive loading occurs in  $^{222}$ Rn (0.67). So, factor 1 might describe the effect of temperature (degassing) and factor 2 probably points at the source of <sup>222</sup>Rn.

# 4.1.3 pH

TDS, Sodium (Na<sup>+</sup>), chloride (Cl<sup>-</sup>), and potassium (K<sup>+</sup>) were negatively correlated with pH, indicating that as pH increases, the TDS, Na<sup>+</sup>, Cl<sup>-</sup> and K<sup>+</sup> will likely decrease. The most highly correlated parameters with pH were K<sup>+</sup> (R= -0.66) and Cl<sup>-</sup> (R= -0.64). The pH seems to be controlled by a thermodynamic equilibrium which is related to CaCO<sub>3</sub> saturation (Banks, Frengstad, Midtgård, Krog, & Strand, 1998). The factor analysis shows negative loadings of pH with measured uranium isotopes (<sup>235</sup>U, <sup>238</sup>U), sodium (Na<sup>+</sup>) and potassium (K<sup>+</sup>), and chloride (Cl<sup>-</sup>). This might be attributed to the dissolution of feldspars and clay minerals from which these elements are partially sourced.

Table 5: Unrotated factor loadings and communalities (Samples #21 to #30); bold values represent the significantly loaded parameters in that factor

Variable	Factor1	Factor2	Factor3	Factor4
Rn-222	0.460	0.696	-0.551	0.027
рН	0.282	-0.827	-0.454	0.173
Temp	0.831	0.081	0.347	0.427
TDS	-0.843	0.183	-0.110	0.493
Variance	1.6931	1.2081	0.6428	0.4560
% Var	0.423	0.302	0.161	0.114



Figure 15: The factor analysis loading plot, including samples #21 to #30

#### 4.2 Environmental Impact

#### 4.2.1 Radon-222

The <sup>222</sup>Rn concentration in all tested water samples is below the maximum permissible limit of WHO, which is 100 Bq L<sup>-1</sup>. To reach a more accurate view, the annual mean effective doses of <sup>222</sup>Rn groundwater samples due to ingestion and inhalation have been calculated by using the parameters established in the reference (Mittal, Rani, & Mehra, 2016), as shown below:

1- Ingestion could be calculated by the following equation:

 $E_{w.Ig} (\mu Sv/y) = CRn_w \times C_W \times (EDC) / 1000....(Eq.1)$ 

Where  $E_{w.Ig}$  is the effective ingestion dose,  $CRn_w$  is the activity concentration of Radon in water (Bq L<sup>-1</sup>),  $C_W$  is the weighted estimated of water consumption (730 L y<sup>-1</sup>), and EDC is the ingestion Effective Dose Coefficient 3.5 nSv/Bq

2- Inhalation could be calculated by the following equation:

 $E_{w.Ih} (\mu Sv/y) = CRn_w \times R_{a.w} \times F \times O \times (DCF) \dots (Eq.2)$ 

Where  $E_{w.lh}$  is the effective inhalation dose,  $CRn_w$  is the activity concentration of Radon in water (Bq L<sup>-1</sup>),  $R_{a.w}$  is the ratio of radon in the air to radon in tap water (10<sup>-4</sup>), F is the equilibrium factor between radon and its decay products (0.4), O is the average indoor occupancy time per person (7000 h/y), and (DCF) is the Dose Conversion Factor for radon exposure 9 nSv/h /(Bq/m<sup>3</sup>).

3- Eq. 1. + Eq. 2. = Annual mean effective dose ( $\mu$ Sv/y)

The calculated total annual effective dose of all sampled water is found to be less than the permissible limit which is 0.1 mSv/y (Table 1), recommended by WHO (2004).

## 4.2.2 Uranium-235 and Uranium-238

The equations in this section were sourced from (Pintilie-Nicolov, Georgescu, Iticescu, Moraru, & Pintilie, 2021; Ajay et al., 2016; Singh et al., 2014; the United States Environmental Protection Agency, 2000; ICRP, 1995).

The <sup>235</sup>U ranges at (0.37- 6.94) mBq L<sup>-1</sup>and this is far below the permissible limit recommended by WHO which is (1 Bq L<sup>-1</sup> = 1000 mBq L<sup>-1</sup>). Also, the <sup>238</sup>U in all tested water samples falls below the limit (10 Bq L = 10000 mBq L<sup>-1</sup>), where it ranges at (7.80- 147.75) mBq L<sup>-1</sup>. For further radiological risk estimations, the annual radioactivity dose to humans due to uranium isotopes was calculated using the following equation:

Annual radioactivity dose (mSv/year) =  $U_a \times W_{in} \times (DCF) \times 10^3$ .....(Eq.3)

Where,  $U_a$  is Uranium activity in water (Bq L<sup>-1</sup>),  $W_{in}$  is Annual Consumption of water (L), and (DCF) is Radioactivity Dose Conversion Factor (Sv Bq<sup>-1</sup>). The (DCF) is (4.7 × 10<sup>-8</sup> Sv Bq<sup>-1</sup>) for <sup>235</sup>U and (4.5 × 10<sup>-8</sup> Sv Bq<sup>-1</sup>) for <sup>238</sup>U. We assumed an intake of 2 liters per capital daily, which is equal to 730 liters per year. The multiplication by 10<sup>3</sup> is added to convert Sv/year to mSv/year. It is also worth mentioning that the activity units of uranium isotopes shall be converted to Bq L<sup>-1</sup> before inserting in the above equation to avoid mistakes. In our table, the uranium activity concentration in the water samples was in mBq L<sup>-1</sup>, so we didn't need to multiply by 10<sup>3</sup> for the conversion from Sv/year to mSv/year. The calculated annual radioactivity dose for <sup>235</sup>U and <sup>238</sup>U ranges at (1.27 × 10<sup>-5</sup> – 2.38 × 10<sup>-4</sup>) mSv/year and  $(2.56 \times 10^{-4} - 4.85 \times 10^{-3})$  mSv/year, respectively. So, all the water samples fall below 0.1 mSv/year in terms of the uranium isotopes annual radioactivity does and are below the maximum permissible dose limit recommended by WHO.

To expand our view on the possible radiological risk from uranium isotopes occurrence in groundwater, the cancer risk was quantified using the equation below:

Cancer Risk =  $U_a \times RF$ ..... (Eq.4)

Where,  $U_a$  is Uranium activity in water (Bq L<sup>-1</sup>), and RF is a Risk Factor (per Bq L<sup>-1</sup>). The RF is calculated as the following:

 $RF = R_{coeffIn} \times IR_{w} \times ET.....(Eq.5)$ 

Where  $R_{coeffIn}$  is the risk coefficient for ingestion,  $IR_w$  is the ingestion rate (730 liters per year), and ET is the exposure time which is assumed to be 65 years for adults in this study. The coefficients for cancer mortality due to ingestion are  $(6.2 \times 10^{-11} \text{ Bq}^{-1})$  and  $(7.5 \times 10^{-11} \text{ Bq}^{-1})$  for <sup>235</sup>U and <sup>238</sup>U, respectively. The coefficients for cancer morbidity due to ingestion are  $(9.8 \times 10^{-11} \text{ Bq}^{-1})$  and  $(1.2 \times 10^{-10} \text{ Bq}^{-1})$  for <sup>235</sup>U and <sup>238</sup>U, respectively. The calculated cancer risk of mortality of <sup>235</sup>U and <sup>238</sup>U in the collected samples ranges at  $(1.09 \times 10^{-9} - 2.04 \times 10^{-8})$  and  $(2.77 \times 10^{-8} - 5.26 \times 10^{-7})$ , respectively. Whereas the cancer risk of morbidity <sup>235</sup>U and <sup>238</sup>U in the collected samples ranges at  $(1.72 \times 10^{-9} - 3.23 \times 10^{-8})$  and  $(4.44 \times 10^{-8} - 8.41 \times 10^{-7})$ , successively. Hence the calculated cancer risk of mortality and morbidity in both <sup>235</sup>U and <sup>238</sup>U are considered very low compared with the maximum radiological risk acceptable guideline level which is  $10^{-3}$  (Shin et al., 2002).

SMP #	Sample ID	Ew.Ig (µSv/y)	Ew.Ih (µSv/y)	<sup>222</sup> Rn Annual effective dose (μSv/y)	Na <sup>+</sup> /Cl <sup>-</sup> ionic ratio	Na <sup>+</sup> /Cl <sup>-</sup> molar ratio
1	R-KH01	104	103	207	0.49	0.76
2	R-KH02	137	135	272	1.00	1.54
3	R-KH03	62	61	124	0.01	0.02
4	R-KH04	99	98	197	0.47	0.72
5	R-KH05	44	43	87	0.46	0.70
6	R-KH06	62	61	123	0.45	0.69
7	R-KH07	75	74	149	0.41	0.63
8	R-KH08	53	52	105	0.82	1.27
9	R-KH10	34	34	68	0.45	0.70
10	R-KH11	66	65	130	0.48	0.74
11	R-KH12	169	167	336	0.49	0.75
12	R-KH13	113	112	225	0.44	0.68
13	R-KH14	34	34	68	0.42	0.65
14	R-KH15	111	109	220	1.01	1.55
15	R-KH16	75	74	150	0.51	0.78
16	R-KH17	23	22	45	0.85	1.32

Table 6: Calculated values of effective ingestion dose, effective inhalation dose,  $^{222}Rn$  annual effective dose, and  $Na^+\!/Cl^-$  ratio

Table 6: Calculated values of effective ingestion dose, effective inhalation
$1 \sim 2^{2/2} \mathbf{D}_{1} \sim 1 \sim$
dose, 222 Kn annual effective dose, and Na+/CI- ratio (continued)

17	R-KH18	34	33	67	NM	NM
18	R-KH19	63	62	125	0.63	0.97
19	R-KH20	105	103	208	0.64	0.99
20	R-KH21	27	26	53	0.38	0.58
21	R63	25	25	50	NM	NM
22	R70	15	14	29	NM	NM
23	R72	51	51	102	NM	NM
24	R65	35	34	69	NM	NM
25	R76	80	79	159	NM	NM
26	R93	7	7	13	NM	NM
27	R94	15	15	30	NM	NM
28	R95	32	32	64	NM	NM
29	RBih	19	19	38	NM	NM
30	R117	13	13	27	NM	NM

NM: Not measured

Table 7: Calculated possibilities of cancer mortalities and morbidities linked to Uranium consumption

S. #	Smp ID	235U Annual radioactivity dose (mSv/y)	238U Annual radioactivity dose (mSv/y)	<sup>235</sup> U Cancer Risk Mortality	<sup>238</sup> U Cancer Risk Mortality	<sup>235</sup> U Cancer Risk Morbidity	<sup>238</sup> U Cancer Risk Morbidity
1	R- KH01	5.06 × 10 <sup>-5</sup>	1 × 10 <sup>-3</sup>	4.34 × 10 <sup>-9</sup>	1.11 x 10 <sup>-7</sup>	6.87 x 10 <sup>-9</sup>	1.77 x 10 <sup>-7</sup>
2	R-	5.59 x	1.13 x	4.79 x	1.22 x	7.58 x	1.95 x
	KH02	10 <sup>5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
3	R-	5.87 x	1.18 x	5.03 x	1.28 x	7.96 x	2.04 x
	KH03	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
4	R-	5.47 x	1.12 x	4.69 x	1.21 x	7.41 x	1.94 x
	KH04	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
5	R-	5.50 x	1.11 x	4.72 x	1.20 x	7.45 x	1.93 x
	KH05	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
6	R-	1.05 x	2.16 x	9.04 x	2.34 x	1.43 x	3.74 x
	KH06	10 <sup>-4</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>
7	R-	6.11 x	1.24 x	5.24 x	1.35 x	8.29 x	2.15 x
	KH07	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
8	R-	2.95 x	6.00 x	2.53 x	6.50 x	4.00 x	1.04 x
	KH08	10 <sup>-5</sup>	10 <sup>-4</sup>	10 <sup>-9</sup>	10 <sup>-8</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
9	R-	5.26 x	1.06 x	4.51 x	1.15 x	7.13 x	1.84 x
	KH10	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
10	R-	5.14 x	1.04 x	4.40 x	1.13 x	6.96 x	1.81 x
	KH11	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
11	R-	5.54 x	1.13 x	4.75 x	1.22 x	7.51 x	1.95 x
	KH12	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
12	R-	2.36 x	4.81 x	2.02 x	5.21 x	3.20 x	8.33 x
	KH13	10 <sup>-4</sup>	10 <sup>-3</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>
13	R-	2.38 x	4.85 x	2.04 x	5.26 x	3.23 x	8.41 x
	KH14	10 <sup>-4</sup>	10 <sup>-3</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>
14	R-	5.31 x	1.08 x	4.55 x	1.17 x	7.19 x	1.87 x
	KH15	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
15	R-	7.81x	1.59 x	6.69 x	1.72 x	1.06 x	2.75 x
	KH16	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>
16	R-	5.75 x	1.17 x	4.93 x	1.27 x	7.80 x	2.03 x
	KH17	10 <sup>-5</sup>	10 <sup>-3</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>	10 <sup>-9</sup>	10 <sup>-7</sup>
Table 7: Calculated possibil	lities of cancer	mortalities and	morbidities				
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linked to Uranium consump	otion (continued	1)					

17	R- KH18	1.27 x 10 <sup>-5</sup>	2.56 x 10 <sup>-4</sup>	1.09 x 10 <sup>-9</sup>	2.77 x 10 <sup>-8</sup>	1.72 x 10 <sup>-9</sup>	4.44 x 10 <sup>-8</sup>
18	R- KH19	1.81 x 10 <sup>-5</sup>	3.64 x 10 <sup>-4</sup>	1.55 x 10 <sup>-9</sup>	3.95 x 10 <sup>-8</sup>	2.45 x 10 <sup>-9</sup>	6.31 x 10 <sup>-8</sup>
19	R- KH20	4.07 x 10 <sup>-5</sup>	8.31 x 10 <sup>-4</sup>	3.49 x 10 <sup>-9</sup>	9.00 x 10 <sup>-8</sup>	5.52 x 10 <sup>-9</sup>	1.44 x 10 <sup>-7</sup>
20	R- KH21	1.26 x 10 <sup>-4</sup>	2.55 x 10 <sup>-3</sup>	1.08 x 10 <sup>-8</sup>	2.76 x 10 <sup>-7</sup>	1.70 x 10 <sup>-8</sup>	4.42 x 10 <sup>-7</sup>
21	R63	NM	NM	NM	NM	NM	NM
22	R70	NM	NM	NM	NM	NM	NM
23	R72	NM	NM	NM	NM	NM	NM
24	R65	NM	NM	NM	NM	NM	NM
25	R76	NM	NM	NM	NM	NM	NM
26	R93	NM	NM	NM	NM	NM	NM
27	R94	NM	NM	NM	NM	NM	NM
28	R95	NM	NM	NM	NM	NM	NM
29	RBih	NM	NM	NM	NM	NM	NM
30	R117	NM	NM	NM	NM	NM	NM

NM: Not measured

### 4.2.3 TDS, pH, and Ions

Total dissolved solids (TDS) in the tested samples range at (142.5 - 10040) mg L<sup>-1</sup>, where ten samples out of thirty include TDS less than 1000 mg L<sup>-1</sup>, and eight samples of these ten include TDS less than 500 mg L<sup>-1</sup>. World Health Organization reports considered drinking water including TDS less than 600 mg L<sup>-1</sup> as preferable and those with TDS less than 1000 mg L<sup>-1</sup> as within the permissible limit of TDS (World Health Organization,

2017). Therefore, one-third of the collected samples are below the maximum permissible limits of drinking water in terms of TDS, and more than two-thirds (21 samples out of 30) contain less than 2000 mg L<sup>-1</sup> which is the maximum permissible limit for irrigation water for some crops (Ayers & Westcot, 1985). The pH has no harmful effect on health in drinking water, however, it might affect the general taste and increase corrosion possibilities as it rises. In the collected water sample the pH ranges at (7.1- 8.5) which is acceptable in terms of taste and corrosive issues during water distribution. Note that the optimum values of pH in water to avoid corrosive effect in pipes during water distribution falls between 6.5 and 8.5 (World Health Organization, 2017). The sodium (Na<sup>+</sup>) ranges at (20.9- 983.42) mg L<sup>-1</sup>, and eleven measurements out of twenty exceed 200 mg L<sup>-1</sup>, which is the limit of sodium taste detection as proposed by WHO.

The chloride (Cl<sup>-</sup>) ranges at (33.1- 3834) mg L<sup>-1</sup>, where thirteen samples out of nineteen contain chloride of more than 300 mg L<sup>-1</sup> which is the maximum limit to detect salty taste in drinking water as mentioned by WHO in 2017.

## 4.3 Radon-222 Concentration in Groundwater from Different Countries

The measured radon concentrations in groundwater from the study area compared with that in other countries around the world (Table 8 and Figure 16) shows that the average value of radon concentration is in a middle position compared to other countries, and this is probably due to the lithology composition in the UAE which is relative with lower content of <sup>222</sup>Rn and it is series parent <sup>238</sup>U. Also, most of the sampled wells in our study are from shallow aquifers which might be a reason behind the radon gas escaping quicker when it is in touch with the atmosphere. Table 8: Comparison of <sup>222</sup>Rn concentration in groundwater (in Bq L-1), measured in Ras Al Khaimah, UAE with those in other regions reported in the literature

Country	<sup>222</sup> Rn Bq L <sup>-1</sup> (Average)	<sup>222</sup> Rn Bq L <sup>-1</sup> (range)	Reference	
UAE	23.2	2.65- 66.1	This study	
Iraq, Erbil	7.92	4- 12.18	Qadir et al., 2021	
India (Northern Rajasthan)	4.42	0.5-22	Mittal & Mehra, 2016	
South Korea	86.6	0.1-2393.5	Cho et al., 2019	
Tunisia	867	0- 2860	Telahigue et al., 2018	
Nigeria (Southwestern part)	8.8	8.2- 12.9	Oni et al., 2019	
U.S (California)	35.39	0- 1506	Grande & Moran, 2021	
Saudi Arabia (Najran)	0.024	0.012- 0.032	Al-Naggar & Abdalla, 2017	
China (Hetao Basin)	19.8	7.67-40.0	Guo et al., 2018	
Oman	0.256	0.140- 0.363	Nasser et al., 2019	
Italy	4.98	1.4- 12.7	Kozłowska et al., 2009	





# **Chapter 5**

# **Chapter 5: Conclusion**

#### 5.1 Concluding Summary

Based on the investigation results and discussion above, the following main conclusions can be drawn:

- The <sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn concentrations in the investigated groundwater (in the Wadi Al Bih aquifer in Northern UAE) are below the WHO permissible limits for drinking water in all the groundwater analyzed here.
- 2. The <sup>222</sup>Rn concentrations in the investigated groundwater are comparable with other countries in arid regions and considered relatively low compared with other countries around the world.
- 3. The calculated annual effective dose resulting from the consumption of <sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn in the investigated groundwater here is below the WHO permissible limits which indicate that radiation in the measured samples will not add a sufficient amount to the maximum permissible annual dose.
- 4. The calculated possibilities of cancer mortalities and morbidities linked to uranium consumption were below the hazardous limits determined by the United States Environmental Protection Agency (EPA) and the International Commission on Radiological Protection (ICRP).



- <sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn in the groundwater of the UAE are mainly sourced from aquifer geochemical interaction and possible seawater intrusion in the coastal aquifers.
- 6. According to statistical analyses:
  - Na<sup>+</sup>, K<sup>+</sup>, Cl<sup>-</sup>, <sup>235</sup>U, and <sup>238</sup>U are more likely to be derived partially from a common source which might be feldspar.
  - <sup>235</sup>U, and <sup>238</sup>U might be also partially derived from the carbonate rocks in the body of the aquifer.
  - SO<sub>4</sub><sup>-2</sup> shows a unique trend which might point to sourcing from gypsum.
  - <sup>222</sup>Rn looked to behave differently from all the abovementioned geochemical parameters, however, it is believed that <sup>222</sup>Rn is a decay product of <sup>238</sup>U.

#### **5.2 Prospect for Future Research**

The results of this research propose several issues for future studies that will expand our understanding of the sources and geochemical interactions of natural radioactivity in the UAE aquifers. Among these issues, conducting regular sampling in groundwater wells for complete major ions analysis and radioactivity measurements in the different seasons. Another issue is sampling rocks from aquifers and starting empirical tests in the lab to estimate the extent of rock-water interaction in different ranges of contact time and different oxidation states. Finally, conducting complete natural radioactive isotopes measurements to understand the decay processes and the isotopes disequilibrium in the UAE's different aquifers.

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#### UAE UNIVERSITY MASTER THESIS NO. 2022: 5

This thesis presents the hydrogeological and environmental assessment of the natural radioactive isotopes (<sup>235</sup>U, <sup>238</sup>U, and <sup>222</sup>Rn) concentrations in the Carbonate Aquifer in Ras Al Khaimah, UAE. All the concentrations are below the WHO's permissible limits for drinking water. Moreover, this thesis will help the decision-makers to adopt regulations and practices that will help in the achievement of sustainable groundwater quality standards and water security.

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