

NATURAL RADIOACTIVITY LEVELS OF SELECTED AREAS IN JUBAN
DISTRICT YEMEN

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Dedicated to:

My inspiring father, my wonderful mother, and the memory of my beloved brother Yasir.

أهدى ثمرة سنوات من الدراسة والغربة

إلى من ادين له بكل ما انجزت في حياتي ومنه تعلمت إن أصبو الى العلياء بإيمان و
عزيمةً وصبراً
والدي: علي عبدربه القاضي

إلى من علمتني أن أكون الافضل ولا اqارن نفسي بالآخرين
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ABSTRACT

The present study aims to determine the levels of natural radioactivity and to assess the corresponding health risk in the region of elevated background radiation in Juban District, Yemen. The relationship between radionuclides concentrations and physicochemical properties in each geological formation of aquifers were estimated to determine the distributions of the radionuclides in groundwater. The mean external gamma dose rate was measured using portable survey meters, which was 374 ± 32 nGy h⁻¹. Rock samples from different geological formations were measured to identify their crystal structures and quantitative determination of radionuclides using X-ray diffraction (XRD) and hyper pure germanium gamma spectrometer, respectively. The XRD results showed that monazite was the dominant radioactive mineral in all geological formations in Juban District. The mean activity concentrations of ²³²Th, ²²⁶Ra, and ⁴⁰K were 1768 ± 918 Bq kg⁻¹, 484 ± 230 Bq kg⁻¹ and 1203 ± 186 Bq kg⁻¹, respectively. The mean specific activity of ²³²Th and ²²⁶Ra were twenty one and six times higher than the world average, thus Juban district may be characterized as an elevated background radiation area. Natural radionuclides in groundwater were estimated using different methods depending on the chemical behaviour of each radionuclides and its kind of radiation. Inductively coupled plasma mass spectrometer, inductively coupled plasma-optical emission spectrometer, and atomic absorption spectrometer were used to analyse the concentration of uranium, thorium, iron and potassium, while the activity concentrations of ²²⁶Ra, ²²⁸Ra, ⁴⁰K, ²³⁸U, ²³⁵U, ²³⁴U and ²²²Rn were measured using gamma spectrometer, alpha spectrometer and Rad 7, respectively. The measured concentration of uranium, thorium, iron, and potassium were 11.25 ± 2.65 µg L⁻¹, 0.15 ± 0.04 µg L⁻¹, 3.20 ± 0.37 mg L⁻¹ and 17.02 ± 0.61 mg L⁻¹ respectively. The mean activity concentrations of ²²⁶Ra, ²²⁸Ra, ⁴⁰K, ²²²Rn, ²³⁸U, ²³⁴U and ²³⁵U were 94 ± 21 mBq L⁻¹, 216 ± 32 mBq L⁻¹, 3306 ± 356 mBq L⁻¹, 226.4 ± 62.4 Bq L⁻¹, 138.2 ± 26.9 mBq L⁻¹, 234.0 ± 41.4 mBq L⁻¹, and 7.2 ± 0.5 mBq L⁻¹, respectively. The highest and lowest concentration of all radionuclides was found to be in the basement and sandstone aquifer, respectively. The mean value of ²³⁴U/²³⁸U activity ratios was 1.8. The relatively low ²³⁸U concentrations and high ratios of ²³⁴U/²³⁸U in the groundwater indicated the presence of younger waters with a stronger leaching of ²³⁴U from aquifer materials to the groundwater. The potential factors for high activity concentration of ²²²Rn, ²²⁶Ra and ²²⁸Ra appear to be the presence of fault and shear. In contrast, the dominant factors affecting U concentrations were salinity of water and water table flow. The annual effective dose was 2.30 mSv, which was five times the world average. The external hazard index was ten times higher than recommended value, which further epidemiological studies of health effects relative to environmental radiation in Juban District need to be conducted.

ABSTRAK

Kajian ini bertujuan untuk menentukan aras radioktiviti semula jadi dan untuk menilai risiko kesihatan yang sepadan di kawasan sinaran latar belakang yang tinggi di Daerah Juban, Yemen. Hubungan antara kepekatan radionuklid dan sifat-sifat fizikokimia dalam setiap pembentukan geologi akuifer telah dianggarkan untuk menentukan taburan radionuklid dalam air bawah tanah. Kadar dos min gama luaran diukur menggunakan meter kajian mudah alih, dengan nilai $374 \pm 32 \text{ nGy h}^{-1}$. Sampel batuan dari pembentukan geologi yang berbeza telah diukur untuk mengenal pasti struktur kristal dan penentuan kuantitatif radionuklid masing-masing dengan menggunakan pembelauan sinar-X (XRD) dan spektrometer germanium gama lampau tulen. Keputusan XRD menunjukkan bahawa monazit adalah mineral radioaktif yang dominan dalam semua formasi geologi di Daerah Juban. Kepekatan aktiviti min ^{232}Th , ^{226}Ra dan ^{40}K masing-masing ialah $1768 \pm 918 \text{ Bq kg}^{-1}$, $484 \pm 230 \text{ Bq kg}^{-1}$ dan $1203 \pm 186 \text{ Bq kg}^{-1}$. Aktiviti khusus min ^{226}Ra dan ^{232}Th ialah dua puluh satu dan enam kali lebih tinggi daripada purata dunia, maka daerah Juban boleh dicirikan sebagai kawasan sinaran latar belakang yang tinggi. Radionuklid semula jadi dalam air bawah tanah telah dianggarkan dengan menggunakan kaedah yang berbeza bergantung kepada sifat kimia setiap radionuklid dan jenis sinaran. Spektrometer jisim gandingan plasma teraruh, spektrometer pemancaran optik gandingan plasma teraruh, dan spektrometer serapan atom telah digunakan untuk menganalisis kepekatan uranium, torium, besi dan kalium, manakala kepekatan aktiviti ^{226}Ra , ^{228}Ra , ^{40}K , ^{238}U , ^{235}U , ^{234}U dan ^{222}Rn diukur dengan menggunakan spektrometer gama, spektrometer alfa dan Rad 7, masing-masing. Kepekatan yang diukur untuk uranium, torium, besi, dan kalium masing-masing ialah $11.25 \pm 2.65 \mu\text{g L}^{-1}$, $0.15 \pm 0.04 \mu\text{g L}^{-1}$, $3.20 \pm 0.37 \text{ mg L}^{-1}$ dan $17.02 \pm 0.61 \text{ mg L}^{-1}$. Kepekatan aktiviti min ^{226}Ra , ^{228}Ra , ^{40}K , ^{222}Rn , ^{238}U , ^{234}U dan ^{235}U masing-masing ialah $94 \pm 21 \text{ mBq L}^{-1}$, $216 \pm 32 \text{ mBq L}^{-1}$, dan $3306 \pm 356 \text{ mBq L}^{-1}$, $226.4 \pm 62.4 \text{ Bq L}^{-1}$, $138.2 \pm 26.9 \text{ mBq L}^{-1}$, $234.0 \pm 41.4 \text{ mBq L}^{-1}$, dan $7.2 \pm 0.5 \text{ mBq L}^{-1}$. Kepekatan tertinggi dan terendah untuk semua radionuklid masing-masing ditemui dalam ruangan bawah tanah dan batu pasir akuifer. Nilai min nisbah aktiviti $^{234}\text{U}/^{238}\text{U}$ ialah 1.8. Kepekatan ^{238}U yang relatif rendah dan nisbah $^{234}\text{U}/^{238}\text{U}$ yang tinggi dalam air bawah tanah menunjukkan kehadiran air yang lebih muda dengan larut lesapan ^{234}U yang lebih tinggi dari bahan akuifer ke dalam air bawah tanah. Faktor-faktor yang berpotensi untuk kepekatan aktiviti ^{222}Rn , ^{226}Ra dan ^{228}Ra yang tinggi kelihatannya adalah kehadiran gelinciran dan ricih. Sebaliknya, faktor-faktor dominan yang mempengaruhi kepekatan U adalah kemasinan air dan aliran air permukaan tanah. Dos berkesan tahunan ialah 2.30 mSv , iaitu lima kali ganda purata dunia. Indeks bahaya luaran ialah sepuluh kali lebih tinggi daripada nilai yang disyorkan, maka adalah perlu dijalankan kajian epidemiologi lanjut bagi kesan kesihatan yang berkaitan dengan sinaran alam sekitar di Daerah Juban.

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LIST OF ABBREVIATIONS

AAS	-	Atomic absorption spectrometer
AED	-	Annual effective dose
AR	-	The activity ratios of uranium isotopes
EC	-	Electrical conductivity
EDXRF	-	Energy dispersive x-ray fluorescence
FWHM	-	Full width at half maximum
GDR	-	Gamma dose rate
GIS	-	Geographical information system
GPS	-	Global positioning system
GSMRB	-	Yemeni Geological Survey and Mineral Resources Board
HPGe	-	High purity germanium
IAEA	-	International Atomic Energy Agency
ICP-MS	-	Inductively coupled plasma-mass spectrometer
ICP-OES	-	Inductively coupled plasma-optical emission spectrometer
ICRP	-	International commission on radiological protection
ICRU	-	International commission on radiation units and measurements
MPD	-	Ministry of Planning and Development.
NRP	-	Natural resources project
TAED	-	Total annual effective dose
TDS	-	Total dissolved solids
US-EPA	-	United States - Environmental Protection Agency

WHO	-	World Health Organization
XRF	-	X-ray
XRD	-	X-ray diffraction fluorescence

LIST OF SYMBOLS

A	-	Activity
K	-	Potassium
Fe	-	Iron
Ra	-	Radium
Rn	-	Radon
SiO ₂	-	Silicon Oxide
Al ₂ O ₃	-	Aluminum Oxide
CaO	-	Calcium Oxide
P ₂ O ₅	-	Phosphate Oxide
TiO ₂	-	Titanium Oxide
ZrO ₂	-	Zirconium Oxide
MnO	-	Magnesium Oxide
Th	-	Thorium
U	-	Uranium
α	-	Alpha particales
β	-	Beta particales
γ	-	Gamma radiation
λ	-	Decay Constant

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Natural radioactive material primarily involves materials containing ^{40}K and isotopes produced from the decay of primordial series. There are three predominant radioactive series in nature, ^{238}U series, ^{232}Th series and ^{235}U series. Each of the three series has many radionuclides in its decay chain ending with a stable isotope of lead (UNSCEAR, 2000).

Natural radioactivity in the environment and associated radiation exposure are related to the geological formations of the area and appear at different levels across the world. The specific activity concentrations of radionuclides in the environment of any area depend on its lithology, and to the ^{238}U , ^{232}Th and ^{40}K content in the rock in each area (Ramli, 2005; UNSCEAR, 2008; Shabana and Kinsara, 2014; Ravisankar et al., 2014).

There many areas with elevated background radiation across the world such as Yangjiang, in China, and Ramsar in Iran (UNSCEAR, 2000; Mohanty et al., 2004). Radiological risk assessment resulting from radionuclides present in rocks and soil has become necessary to limit exposure to ionizing radiation.

In areas that have high radioactivity in their geological formations, elevated concentrations of Th, U and their decay products might be found in the groundwater. As a result of water- rock interaction, radioactive elements could be transferred into the groundwater from the deposits of radioactive minerals in the aquifer's rocks (Kraemer and Genereux, 1998; Porcelli, 2008; Osman et al., 2008). In the elevated background areas, wells drilled in the bedrock have high concentrations of natural radionuclides in water quality tests (Isam et al., 2002).

Investigation of the concentrations and behaviour of natural radionuclides in groundwater is given baseline data for environmental and epidemiological studies (Ahmed et al, 2004). Groundwater may have high concentration of natural radionuclides, especially U, ^{228}Ra , ^{226}Ra and ^{222}Rn . Due to the chemical toxicity of U for kidney, and the radiological effects of Ra isotopes on bones, their occurrence in drinking water and associated health hazards require comprehensive monitoring. Ra is chemically similar to calcium, so grown bone takes up Ra and may cause bone cancer. (De Oliveira et al, 2001; ICRP, 1993; EPA, 1991). According to experimental studies, the ingestion of ^{222}Rn from water may cause the same fatal cancer as the fatal lung cancers due to inhalation of indoor ^{222}Rn (Correia et al., 1987). Due to their hazards, many countries have adopted standards for natural radionuclides concentrations in water (Bonotto and Bueno, 2008; Hadad et al., 2008; WHO, 2008; El-Mageed et al, 2013; Shabana and Kinsara., 2014).

1.2 Problem statement

Surveys and studies of distribution of natural radionuclides in the environment are of great importance for both the assessment of public health risks and the performance of epidemiological studies (El-mageed et al., 2011). Airborne radiometric measurements were carried out for some parts of Yemen (NRP, 1990; IAEA, 2010; Abdul-Had et al, 2011). The airborne survey identified some locations with elevated count rates (exceeding 3000 cps) in the west of Juban City and south part of Nawah Village in Juban District, compared to other locations in the survey

area (Alzeiteri, 1997; Abdurabu et al., 2016a). Juban district is one of areas with high-count rates and high population density (151.41 per km²) (CSO, 2004). The natural radionuclids could reach high levels in the anomalies rocks, as well as in the groundwater that interactives with the rocks. Consequently, the number of the population could be negatively affected by these radioactive anomalous (Pereira and Neves, 2012). As a result, an extensive survey for the district of Juban should be conducted to determine whether this area could be registered as an elevated area comparable to other areas worldwide.

Most natural radionuclides are from uranium (U) and thorium (Th) series. Natural radioactivity in groundwater is produced from rock water interactions. Groundwater may contain naturally occurring radionuclides from the ²³⁸U, ²³⁵U, and ²³²Th series and ⁴⁰K. There are many factors controlling the concentrations of these radionuclides in groundwater, such as the concentration of radioelements in the bedrock, and the chemical and physical conditions of groundwater that affect the water- rock interaction (Wanty and Schoen 1993, Kraemer and Genereux, 1998, Chau et al. 2011). The high natural radioactivity levels in groundwater are often associated with elevated concentrations of U and Th series radionuclides in rocks due to water rock interaction. Since the groundwater is the dominant of water sources at Juban District, study about radionuclides transport into groundwater is necessary.

It is very important to assess the corresponding health risk due to natural radionuclides in Juban's environment, to give a baseline for further epidemiological studies of health effects relative to exposure to natural radioactivity. Therefore, the current study being the first of its kind in Yemen, aims to evaluate natural radiation and radioactivity in the groundwater, rock, and soil for areas with high population density in Juban District and to assess the corresponding health risk in the region of an expected elevated background radiation.

1.3 Objectives

The main objectives of this study were to investigate the natural radioactivity in Juban District's environment and to describe the distribution and determine the primary factors controlling the occurrence of natural radionuclides in groundwater of Juban District, Yemen. In addition, assessment of the corresponding health risk due to natural radionuclide in Juban District's environment to provide baseline data that can be beneficial to enlighten the local radiation regulation making. To achieve this, the objectives of the study are:

- i. To measure the external gamma dose rate and the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the rock, and soil samples from Juban District.
- ii. To estimate the occurrence and distribution of natural radionuclides in groundwater of Juban District.
- iii. To investigate the relation between radionuclides concentration in water with geological formation of aquifers and physicochemical properties of water.
- iv. To evaluate the corresponding radiological health risk in the study area due to natural radioactivity occurrence in water, rock and soil samples.
- v. To produce an isodose mapping for terrestrial gamma dose rates and activity concentrations of ^{228}Ra , ^{226}Ra , ^{222}Rn and U concentrations in groundwater using Surfer 13 software.

1.4 Scope of study

Juban District is situated at $14^{\circ} 30'$ to $13^{\circ} 55'$ N latitude and $44^{\circ} 48'$ to $44^{\circ} 56'$ E longitude with an average altitude of 2070 m above sea level. It covers a total land of 178 km^2 and has a population of 26,938 inhabitants (Abdurabu et al., 2016a).

In this study, the natural radiation background of Juban District was obtained by measuring the gamma dose rate directly using Ludlum Model 3 (Model 44-88 Cylindrical (Pancake) detector operated with Ludlum Model 3 USA).

Natural radionuclides in groundwater of Juban District were estimated by different methods depending on the chemical behaviour of each radionuclide in water and its kind of radiation and energy. The physicochemical parameters of water were measured such as pH, temperature, electrical conductivity (EC) and total dissolved solids (TDS). The pH and the temperature of water were measured in the field using a 3150 pH meter (manufactured by Jenway). The electrical conductivity (EC) of water was measured in the field by HI8733 Conductivity meter (manufactured by Hanna instruments).

The concentration of U, Th, K, and Fe concentrations in water samples were measured using different methods depending on the type of element. Inductively coupled plasma mass spectrometry (ICP-MS, Elan 600) was used to determine the concentrations of U and Th in water samples, while K was measured using a flame atomic absorption spectrophotometer (AAS Analyst 800) in the analytical chemistry laboratory at Malaysian Nuclear Agency. Fe concentration was measured using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES, Agilent 700 Series) at Unit Pengurusan Makmal Universiti (UPMU), UTM.

The activity concentrations of ^{226}Ra , ^{228}Ra , ^{40}K , ^{238}U , ^{235}U , ^{234}U and ^{222}Rn were measured using different methods depending on the type of radiation and energy. Rad 7 detector (silicon-based semi-conductor detector), manufactured by DurrIDGE Company, was used to determine the concentration of ^{222}Rn since it is the best way to determine its concentration with careful sampling.

The activity concentrations of ^{238}U , ^{235}U and ^{234}U were measured using alpha spectrometry at Radiochemistry and Environment Laboratory (RAS), Malaysian Nuclear Agency, while ^{226}Ra , ^{228}Ra , and ^{40}K were analysed using hyper pure germanium (HPGe) in Nuclear Laboratory at Universiti Teknologi Malaysia.

The data of the radiometric, hydrological and geological maps of Juban were analysed to help to understand the distribution of radionuclide in Juban District's groundwater and environment.

To investigate the distribution of the natural radioactive in Juban's environment, contour maps of gamma dose rate, ^{222}Rn , ^{226}Ra , ^{228}Ra , and U concentration in groundwater were drawn by using Surfer 13 program and the Kriging method for gridding the data.

1.5 Significance of study

Natural radioactivity is everywhere on the earth. The assessment of gamma dose rate, and natural radioactivity due to ^{232}Th , ^{226}Ra and ^{40}K in environment media has become essential to identify hot spots or areas with elevated background, and to monitor the public exposure. In addition, it is important to establish a baseline data that can serve as useful information in assessing any future changes in the environment due to human activities or any other artificial activities (Othman and Yassine, 1995; Rajesh et al., 2013; Ramasamy et al., 2015; Idriss et al., 2016).

The airborne radiometric survey of Yemen is the only indicator of the occurrence of radioactive anomalies. Juban District is a populated area among areas that have elevated radiation. It is exposed to high radiation because of gamma radiation, and consumption of groundwater containing high concentrations of radionuclides. The distribution of natural radionuclides among different media of the environment such as groundwater, soil and rock were investigated. In addition, the corresponding health risk due to natural radionuclides in Juban's environment was assessed. This study will be applied in other areas in Yemen that the airborne gamma-ray surveys show are good candidates as radioactive anomalies resources. Finally, this study would add to the knowledge about behaviour and distribution of radionuclides in groundwater from highland areas in semi- arid environment.

1.6 Outline of thesis

This thesis was divided into five chapters. Chapter One explains the introduction to the research. It includes the background of study, problem statement, objectives, scope of research and thesis outline.

Chapter two consists of the literature review of the research. It contains an introduction to radioactivity, basic concepts and terminology of radiation, related health hazards, radioactivity in environment and in water, behavior of natural radionuclides in water, disequilibrium in uranium series, health standards and criteria for radionuclides in water, natural radioactivity studies in different countries, and radiation studies in Yemen.

Chapter Three presents the description of geological and hydrological settings of Juban District and the methodology used in this study to meet the objectives of the thesis. This includes measurement of gamma dose rate, sample collection, rock and soil samples collection, water samples collection, sample preparation, experimental set-up and equipment, rock samples measurement, gamma spectrometer measurement, X-Ray Diffraction measurement, Energy Dispersive X-Ray Fluorescence (EDXRF) measurement, measurement of ^{232}Th , ^{226}Ra , and ^{40}K activity concentrations in soil samples, measurement of natural radionuclides in water samples, measurement of the physicochemical parameters of water, measurements of the total concentration of U, Th, K, and Fe in water samples, ICP-MS measurement for U and Th concentrations, Atomic Absorption Spectrometer (AAS), Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES), measurement of ^{222}Rn concentrations, measurement of U isotopes by alpha spectrometer, statistical analysis of calibrated data, assessment of radiological health effects and spatial distribution of natural radiation..

Chapter Four presents the experimental results, analysis of data and discussion of research results and estimation of radiological health effects in Juban District. Chapter Five presents conclusions and recommendations.

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