NATURAL RADIOLOGICAL STUDIES OF KELANTAN AND TERENGGANU STATES, MALAYSIA

NURADDEEN NASIRU GARBA

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My father, Alhaji Nasiru Garba Getso;

My mother, Hajiya Yahanasu Nasiru Getso;

My late sister, Fatima Nasiru Getso; and

My brothers and sisters

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ABSTRACT

Natural environmental radioactivity arises mainly from primordial radionuclides such as ^{40}K and also from ^{238}U and ^{232}Th decay series and have always been present in a variety of concentrations in every part of the earth's mantle and in the tissue of every living being. Natural radioactivity can be found almost everywhere; in soil, public water supplies, oil and atmosphere. It poses a measurable exposure to human beings. The present study was aimed of providing the base line data of Terrestrial Gamma Radiation Dose rates (TGRD), natural radioactivity concentrations and the corresponding radiological health hazards in the environments of Kelantan and Terengganu states. TGRD were measured using a micro roentgen survey meter model 19 manufactured by Ludlum, from 150 and 145 locations at Kelantan and Terengganu states, respectively. A total of sixty (60) soil samples and ten (10) water samples from major rivers were collected with thirty six (36) soil and five (5) water samples collected from Kelantan and twenty four (24) soil and five (5) water samples collected from Terengganu, respectively. The soil samples were analyzed using a high purity germanium detector (HPGe) and Genie 2000 software, while the water samples were analyzed at Malaysian Nuclear Agency using atomic absorption spectrometry (AAS) for ^{40}K and inductively coupled plasma mass spectrometer (ICP-MS) for U and Th concentrations. The measured TGRD mean values of 209 nGy h^{-1} and 150 nGy h^{-1} which are about three times the world and two times Malaysian averages of 59 nGy h^{-1} and 92 nGy h^{-1} respectively. The mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the soil samples were found to be 82 Bq kg^{-1}, 123 Bq kg^{-1} and 643 Bq kg^{-1} for Kelantan, and 79 Bq kg^{-1}, 84 Bq kg^{-1}, and 545 Bq kg^{-1} for Terengganu. ^{226}Ra and ^{232}Th in Kelantan are three times and in Terengganu twice the world average values of 32 Bq kg^{-1} and 45 Bq kg^{-1}, while ^{40}K is slightly higher than the world average value of 420 Bq kg^{-1}. For water samples, the mean concentrations of U and Th and activity concentration of ^{40}K was found to be 13 mBq L^{-1}, 4 mBq L^{-1} and 1119 mBq L^{-1} for Kelantan and 0.71 mBq L^{-1}, 0.23 mBq L^{-1} and 56 mBq L^{-1} for Terengganu, respectively. The health hazard impact of radium equivalent (Ra_{eq}), annual effective dose (*AED*), and external radiation hazard index (H_{ex}) which are indicators of radiological health hazards were computed as 307 Bq kg^{-1}, 1.28 mSv y^{-1} and 0.83 for Kelantan and 242 Bq kg^{-1}, 0.92 mSv y^{-1} and 0.65 for Terengganu, respectively. Statistical relationships between TGRD with underlying geological formations and soil types were obtained. Isodose contour maps which shows the distribution pattern of TGRD for both states were produced. Radiological health due to TGRD and natural radioactivity are on the average higher than both the world average and Malaysian average but were still within the recommended values of 370 Bq kg^{-1}, 0.48 mSv y^{-1} and unity, thus should not pose any significant danger to the populations.

ABSTRAK

Keradioaktifan alam sekitar semulajadi terbebas ke alam sekitar terutamanya daripada radionuklid purba seperti ^{40}K dan daripada siri reputan ^{238}U dan ^{232}Th dan yang sentiasa wujud dengan pelbagai kepekatan di setiap lapisan mantel bumi dan tisu setiap hidupan. Keradioaktifan semulajadi boleh didapati hampir di mana-mana; di dalam tanah, bekalan air awam, minyak dan atmosfera. Ia menyebabkan pendedahan yang perlu dipertimbangkan kepada manusia. Kajian ini bertujuan untuk menyediakan data dasar kadar dos sinaran gama daratan (TGRD), kepekatan keradioaktifan semulajadi dan hazad kesihatan radiologi di negeri Kelantan dan Terengganu. TGRD diukur menggunakan meter survey mikro Roentgen model 19 yang dikeluarkan oleh Ludlum di 150 dan 145 lokasi masing-masing di negeri Kelantan dan Terengganu. Enam puluh (60) sampel sejumlah tanah dan sepuluh (10) sampel air dari sungai utama telah dikumpulkan, dengan tiga puluh enam (36) sampel tanah dan lima (5) sampel air diambil dari Kelantan, manakala dua puluh empat (24) tanah dan lima (5) sampel air diambil daripada Terengganu. Sampel tanah telah dianalisis menggunakan pengesan germanium ketulenan tinggi (HPGe) dan perisian Genie 2000, manakala sampel air dianalisis di Agensi Nuklear Malaysia menggunakan spektrometri serapan atom (AAS) untuk ^{40}K dan spektrometer jisim plasma teraruh (ICP-MS) untuk kepekatan U dan Th. Purata TGRD yang diukur adalah 209 nGy j^{-1} dan 150 nGy j^{-1}, kira-kira tiga kali ganda nilai purata dunia dan dua kali purata Malaysia iaitu masing-masing 59 nGy j^{-1} dan 92 nGy j^{-1}. Purata kepekatan keaktifan ^{226}Ra, ^{232}Th dan ^{40}K dalam sampel tanah didapati bernilai 82 Bq kg^{-1}, 123 Bq kg^{-1} dan 643 Bq kg^{-1} untuk negeri Kelantan, dan 79 Bq kg^{-1}, 84 Bq kg^{-1}, dan 545 Bq kg^{-1} bagi negeri Terengganu. ^{226}Ra dan ^{232}Th di Kelantan adalah tiga kali dan di Terengganu adalah dua kali ganda nilai purata dunia ia iatu 32 Bq kg^{-1} dan 45 Bq kg^{-1}, manakala ^{40}K adalah sedikit lebih tinggi daripada nilai purata dunia iaitu 420 Bq kg^{-1}. Bagi sampel air, purata kepekatan keaktifan U, Th dan ^{40}K didapati bernilai masing-masing 13 mBq L^{-1}, 4 mBq L^{-1} dan 1119 mBq L^{-1} di negeri Kelantan, dan 0.71 MBq L^{-1}, 0.23 MBq L^{-1} dan 56 mBq L^{-1} di negeri Terengganu. Hazad kesihatan setara radium *(Ra_{eq})*, dos berkesan tahunan (*AED*), dan indeks hazad sinaran luaran (H_{ex}) yang merupakan petunjuk hazad kesihatan radiologi telah dikira, masing-masing bernilai 307 Bq kg^{-1}, 1.28 mSv thn^{-1} dan 0.83 di negeri Kelantan dan 242 Bq kg^{-1}, 0.92 mSv thn^{-1} dan 0.65 di Terengganu. Hubungan statistik antara TGRD dengan pembentukan geologi dan jenis tanah telah diperolehi. Peta kontur isodos yang menunjukkan corak taburan TGRD di kedua-dua negeri telah dihasilkan. Secara purata, kesan kesihatan radiologi daripada TGRD dan keradioaktifan semula jadi didapati lebih tinggi daripada kedua-dua purata dunia dan Malaysia, tetapi masih dalam nilai yang dicadangkan iaitu 370 Bq kg^{-1}, 0.48 mSv thn^{-1} dan uniti, oleh itu ia mungkin tidak menimbulkan bahaya yang besar kepada penduduk.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATION

AAS	-	Atomic absorption spectrometry
AEDE	-	Annual effective dose equivalent
AELB	-	Atomic energy licensing board
AGDE	-	Annual gonadal dose equivalent
ANOVA	-	Analysis of variance
BEIR	-	Biological effects of ionizing radiation
DAAPM	-	Department of agriculture Peninsular Malaysia
DGGS	-	Department of geological survey
DNA	-	Deoxyribonucleic acid
DOS	-	Department of statistics
EIA	-	Environmental impact assessment
FAO	-	Food and agriculture organization
FEPE	-	Full energy peak efficiency
FWHM	-	Full width at half maximum
GIS	-	Geographic information system
GPS	-	Global positioning system
HPGe	-	High purity germanium
HPIC	-	High pressurized ionization chamber
IAE	-	Internal annual effective dose
IAEA	-	International Atomic Energy Agency
ICLARM	-	International centre for living aquatic resources mgt.
ICP-MS	-	Inductively coupled plasma mass spectrometry
ICRP	-	International commission on radiological protection
ICRU	-	International commission on radiation units and measurements

INWQS	-	Interim national water quality standards for Malaysia
MCA	-	Multichannel analyzer
MDA	-	Minimum detectable activity
NM	-	Nuclear Malaysia
NPP	-	Nuclear power plant
OECD	-	Organization for Economic Co-Operation and Development
OAE	-	Outdoor annual effective dose
ppb	-	parts per billion
ppm	-	parts per million
QC	-	Quality control
RF	-	Risk factor
RIA	-	Radiological impact assessment
ROI	-	Region of interest
SC	-	Collective effective dose
SPSS	-	Statistical package for social sciences
SSDL	-	Secondary standards dosimetry laboratory
TAE	-	Total annual effective dose
TGRD	-	Terrestrial gamma radiation dose rate
UTM	-	Universiti Teknologi Malaysia
UNESCO	-	United Nation Educational Scientific and Cultural Organization
UNSCEAR	-	United Nations Scientific Committee on the Effects of Atomic Radiation
US NRC	-	United States Nuclear Regulatory Commission

LIST OF SYMBOLS

A	-	Activity
E	-	Energy
w_T	-	Tissue weighting factor
w	-	Channels
L_c	-	Critical limit
D	-	Absorbed dose
Q	-	Quantity
N_p	-	Net peak area
X_t	-	Life time
σ	-	Uncertainty
ε	-	Efficiency
γ	-	Gamma radiation
L_D	-	Detection limit
L_t	-	Life expectancy
λ	-	Decay constant
B_R	-	Branching ratio
Ra_{eq}	-	Radium equivalent
R	-	Cancer risk
R_L	-	Life time cancer risk
T_1	-	Half-life of parent nuclides
T_2	-	Half-life of progeny nuclides
A_1	-	Activity of the parent nuclides
A_2	-	Activity of the progeny nuclides
H_{ex}	-	External hazard index
H_{in}	-	Internal hazard index
E_D	-	Effective dose

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Human beings are continuously exposed to natural radiation from different source which includes soil, building materials, air, water, the universe and even within their own bodies (Kurnaz, 2013). Natural sources of radiation are known to be the most significant source of public exposure to ionizing radiation (Faanu *et al.*, 2011). Gamma radiation emitted from primordial radioisotopes is one of the main external sources of radiation on earth (UNSCEAR, 1993, 2000). Ninety nine percent (99%) of the total radiation exposure to the population (excluding medical exposure) was known to be due to the natural radiation sources, with only a very little contribution from nuclear power production and nuclear weapons testing (UNSCEAR, 2000).

Assessment of human exposure to gamma radiation is very significant, since natural radiation sources are the largest contributor of external dose to the world population (Lee *et al.*, 2009; Martin and Harbison, 1972). Terrestrial radionuclide originates from the earth crusts and came into existence with the creation of the planet (Wicks, 2011). Some of these radionuclides have half-lives of the order of billions of years, such that it takes a long time for them to decay and become non-radioactive, and are part of the human and non-human biota till today.

Natural radioactivity arises mainly from ^{238}U and ^{232}Th decay series and ^{40}K, which exist at trace amounts in all ground formation (Tzortzis *et al.*, 2004). Level of exposure to natural radioactivity majorly depends on the geological formations and soil types. They occurs at different levels in different part of the world (UNSCEAR, 1993), as such terrestrial background radiation monitoring due these radionuclides in soil is very essential. Several studies had been carried out performed worldwide, that measured the terrestrial gamma radiation dose rate (TGRD) and activity concentration of natural radionuclides in soil (Ahmad and Khatibeh, 1997; Al-Jundi, 2002; Fatima *et al.*, 2008; Karahan and Bayulken, 2000; Matiullah *et al.*, 2004; McAulay and Morgan, 1988; Quindos *et al.*, 1994; Ramli, 1997; Ramli *et al.*, 2009b; Ramli *et al.*, 2003; Ramli *et al.*, 2005b; Saleh *et al.*, 2007; Tahir *et al.*, 2005).

Drinking water may also contain significant levels of radioactive nuclides that could pose a risk to human health. These radionuclides usually enters the water supply system at any point, or at several points prior to consumption. For this reason, naturally occurring radionuclides in water are often less controllable (Garba *et al.*, 2012).

1.2 Problem statement

Studies and surveys of natural radioactivity is very significant in health physics for various practical and fundamental scientific reasons (Abd El-mageed *et al.*, 2011). The ever increasing mining activities, nuclear industries and other contamination sources (Agricultural activities) which are widespread necessitate the need to evaluate the terrestrial natural radioactivity. The data will be kept as a baseline information to determine the change in the environmental due to other human activities in future. The data will be needed in formulating safety and national standard guidelines for Malaysia in line with international recommendations.

In view of the possibility of Malaysia having operational nuclear power plant (NPP) by the year 2030 (Saleh *et al.*, 2014a), measurement and monitoring of terrestrial gamma radiation dose as well as the determination of the primordial radionuclides levels in soil and water are great important in NPP site assessment. This will enable areas of needs be identified and steps be taken to fulfill the International Atomic Energy Agency (IAEA) and Atomic Energy Licensing Board (AELB) of Malaysia requirements for environmental radiological data for nuclear power plant. Despite the great interest shown in measuring environmental radioactivity in Malaysia, literature have shown that the states of Kelantan and Terengganu were not fully covered (Alias *et al.*, 2008; Hamzah *et al.*, 2008; Hamzah *et al.*, 2012; Hamzah *et al.*, 2011a; Hamzah *et al.*, 2011b; Hamzah *et al.*, 2011c).

Therefore, the current study being the first of its kind in Kelantan and Terengganu, aims to study the TGRD and natural radioactivity in soil and major rivers in Kelantan and Terengganu and also investigate the influence of geological formations and soil types on the TGRD and activity of ^{226}Ra, ^{232}Th and ^{40}K in soil in their environments. The information obtained could be used to predict the natural radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K without resorting to full-scale laborious measurements, and sampling in areas difficult to access. This study will also assess the true picture of radiological effects to the population if any, due to external and internal exposure to natural radioactivity in Kelantan and Terengganu states.

1.3 Objectives of the study

The principal aim of this research is to provide baseline data / radiological information of the Kelantan and Terengganu states, Malaysia. The specific objectives are;

(a) To determine the terrestrial gamma radiation dose rates (TGRD) and produce a digital isodose mapping of the study areas using a geographic information system (ArcGIS) software as baseline data.

(b) To determine levels of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides in surface soil for each district and in water samples from major rivers in the two states and their radiological implication.

(c) To evaluate the relation between the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations and TGRD based on geological formations and soil types information in the two states and to make analytical comparison with previous studies.

(d) To enhance/improve the model for predicting environmental radioactivity based on geological formations and soil types information using SPSS.

1.4 Significances of this study

Radioactivity exists everywhere in nature, a such assessment TGRD and ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in the environment has become necessary in order to determine the amount of change in the environmental radioactivity levels with time, which is significant for environmental protection (Sroor *et al.*, 2001). Investigations of natural radioactivity in the environment is very important in order to identify hot spot or areas with high natural radiation and also to establish a baseline data which can serve as a useful information in assessing any future changes in the background natural radiation level due to human activities or any other artificial activities (Ibrahiem *et al.*, 1993; Lee, *et al.*, 2009; Quindos, *et al.*, 1994).

1.5 Scope of the study

In the present study attention is focused mainly on the unmodified natural radiation (terrestrial radiation) and its associated health effects. The work covers in situ measurement of terrestrial gamma radiation dose rate using a portable detector model 19, micro Roentgen (μR) meter, manufactured by Ludlum USA, and terrestrial (primordial) radionuclides in the soil and water samples of major rivers using hyper pure germanium, HPGe, detector, inductively coupled plasma mass spectrometer ICP-MS instrument model ELAN 6000 and atomic absorption spectroscopy, AAS, respectively. The study covers the entire Kelantan and Terengganu states involving all geological formations and soil types.

A comparative analysis and statistical tests were conducted to test the difference and normality of the data collected. The mean activity concentrations for each district in the two states were measured and compared.

Radiological health parameters such as annual effective dose, the collective effective dose, mean lifetime dose, life time cancer risk, radium equivalent, external and internal hazard indices, annual gonadal dose equivalent and mean annual effective dose equivalent were computed from results obtained from in situ TGRD measurements and ^{232}Th, ^{226}Ra and ^{40}K activity concentrations in soil. These will provide basic information for environmental radiological assessments and be used as a base reference level for future changes in the radiological environment due to human activity.

Isodose map of the study area were drawn using Geographic Information System, GIS, Arc View. The digital map shows the TGRD levels in the area.

1.6 Thesis outlines

This thesis will be 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 outlines.

Chapter Two presents the literature review of the study. It includes introduction, radioactivity, environmental radioactivity, terrestrial sources, cosmic sources, artificial sources, radioactive decay law, activity, specific activity, radioactive equilibrium, secular equilibrium, transient equilibrium, no equilibrium, quantity and units of radiation, exposure, absorbed dose, equivalent dose, effective dose, radiological hazards implication, natural radioactivity in soils, natural radioactivity in rocks, natural radioactivity in igneous rocks, natural radioactivity in sedimentary rock, natural radioactivity in metamorphic rocks, natural radioactivity in water, natural radioactivity studies in different countries, TGRD in different countries, TGRD in Malaysia, natural radionuclides activity concentration in the soils of different countries, studies of ^{226}Ra, ^{232}Th and ^{40}K activity in soil across Malaysia, activity of ^{226}Ra, ^{232}Th and ^{40}K in the rivers of some countries, activity of ^{226}Ra, ^{232}Th and ^{40}K in the rivers across Malaysia, radiation health effects, study area, Kelantan, Terengganu, geological formations of Kelantan, geological formations of Terengganu, soil types, alluvial soils, sedentary soils, miscellaneous soils, and soil types of Kelantan and Terengganu, respectively.

Chapter Three presents the description of the study areas and methodology used for of the research to meet the objectives of this study, this includes; experimental, measurement of TGRD, sample collection, soil sample collection, water sample collection, sample preparation, experimental set-up and equipment, gamma-ray spectroscopy, energy calibration, efficiency calibration, data analysis of gamma ray spectrum, measurement of U, Th and ^{40}K in water ICP-MS for U and Th measurement, AAS for ^{40}K, assessment of radiological effects, annual effective dose, radium equivalent (Ra_{eq}), external and internal hazard indices, average life time effective dose

$ALtE_D$ and cancer risk (*R*), statistical analysis, normality test, descriptive statistics, the anova test, t-test, regression, assessment of radiation hazard, ordinary kriging method.

Chapter Four presents the experimental results, analysis of data and discussion of research results.

Chapter Five presents conclusions and recommendations.

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