MEASUREMENT OF RADON-222 AND RADIUM-226 IN WELL WATER

OF KELANTAN

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MEASUREMENT OF RADON-222 AND RADIUM-226 IN WELL WATER OF KELANTAN

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Dedicated to God, my beloved parents, my dearest husband (Mohd Faizi bin Abu) and my lovely twin boys (Ahamd Fathi Zulkarnain and Ahmad Fitri Zunnurain) iii

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ABSTRACT

The Environmental Protection Agency (EPA) of USA sets the Maximum Contamination Level (MCL) for ²²²Rn in drinking water to be 11.1000 Bq/l and for ²²⁶Ra to be 0.1100 Bq/l. This study was conducted to determine the levels of ²²²Rn and ²²⁶Ra in private well water in the state of Kelantan. A total of 25 samples of well water were collected from various districts in the state. The levels of ²²²Rn and ²²⁶Ra dissolved in well water were measured using the liquid scintillation counting technique. ²²²Rn activity concentrations were found within the range of 0.0491 to 4.3127 Bq/l for both unfiltered and filtered samples. Meanwhile, activity concentration of ²²⁶Ra measured is between 0.0278 to 1.9670 Bq/l for both unfiltered and filtered samples, exceeding the MCL value. The annual effective doses due to the intake of natural radionuclide from well water for three different age groups were estimated based on the amount of water consumed. The average annual effective dose of 226 Ra calculated for babies in the range of 0.12 to 1.79 μ Sv/y. The average annual effective dose of 226 Ra for children was in the range of 0.11 to 0.34 μ Sv/y while, for the adults, it was in the range of 0.11 to 0.33 μ Sv/y for ²²⁶Ra. The annual effective dose intake for the babies was found to be higher than other groups but within the limit recommended by World Health Organization (WHO). The doses for the other two groups are below the recommended value.

ABSTRAK

Agensi Perlindungan Persekitaran atau Environmental Protection Agency (EPA) dari Amerika Syarikat menetapkan aras pencemaran maksimum atau Maximum Contamination Level (MCL) untuk ²²²Rn di dalam air minuman adalah 11.1000 Bq/l dan untuk ²²⁶Ra adalah 0.1100 Bq/l. Kajian ini dijalankan untuk mengenalpasti aras ²²²Rn dan ²²⁶Ra dalam air perigi persendirian di negeri Kelantan. Sebanyak 25 sampel air perigi telah dikumpul dari pelbagai daerah di negeri tersebut. Aras ²²²Rn dan ²²⁶Ra terlarut dalam air perigi diukur dengan menggunakan teknik pembilang sintilator. Kepekatan aktiviti bagi ²²²Rn adalah dalam julat 0.0491 sehingga 4.3127 Bq/l untuk sampel tidak dituras dan dituras. Sementara itu, aktiviti bagi ²²⁶Ra, kepekatan yang diukur adalah antara 0.0278 sehingga 1.9670 Bq/l untuk kedua-dua sampel vang tidak dituras dan dituras, iaitu melebihi nilai MCL. Dari sampel, boleh anggar dos tahunan yang berkesan untuk pengambilan radionuklid semulajadi dalam air perigi bagi tiga kumpulan umur yang berbeza telah dianggarkan berdasarkan jumlah air yang digunakan. Purata pengambilan dos tahunan berkesan bagi ²²⁶Ra yang dikira untuk bayi adalah dalam lingkungan 0.12 sehingga 1.79 µSv/y. Bagi kanak-kanak pula, purata dos tahunan berkesan adalah dalam lingkungan 0.11 sehingga 0.34 μ Sv/y. Manakala purata pengambilan dos tahunan berkesan untuk orang dewasa dalam lingkungan 0.11 sehingga 0.33 µSv/y. Pengambilan dos tahunan berkesan untuk bayi didapati lebih tinggi daripada kumpulan lain tetapi masih berada dalam had seperti yang disyorkan oleh World Health Organization (WHO). Dos untuk kedua-dua kumpulan yang lain adalah di bawah nilai yang disyorkan.

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LIST OF SYMBOLS

А	Alfa
eta	Beta
Λ	Half life
Ao	Original activity
A _t	Current activity
Bq	Becquerel
Bq/l	Becquerel per litre
С	Concentration of radionuclide
CA	Concentration in Air
Co	Original Radon concentration in water
CT	Concentration in Toluene
Cw	Concentration in Water
D	Annual effective dose
D _T	Diffusion coefficient for Radon between Air and Toluene
D_W	Diffusion coefficient for Radon between Air and Water
E	Conversion factor
HNO ₃	Nitric Acid
Ι	Annual intake of drinking water
pCi/l	Pico Curie per little
POPOP	[1, 4-Bis (5-phenyl-2-oxazoly)]
PPO	(2, 5-diphenylozaxole).
Ra	Radium
Rn	Radon
R _N	Total activity of Radon
S	Second

Sv/y	Sievert per year
Т	Elapsed time
Th	Thorium
U	Uranium
VA	Volume of Air
V t	Volume of Toluene
V_{W}	Volume of Water
Z	Atomic number
²¹⁴ Bi	Bismuth-214
²²² Rn	Radon-222
²²⁶ Ra	Radium-226
²²⁸ Ac	Actinium-228
²¹² Pb	Lead-212
²³⁴ U	Uranium-234
²³⁵ U	Uranium-235
²³⁸ U	Uranium-238
⁴⁰ K	Potassium-40

LIST OF ABBREVIATIONS

ADL	Annual Dose Limit
AMCL	Alternate Maximum Contaminant Level
ANM	Agency Nuclear Malaysia
BSRP	Basic Safety Radiation Protection
СРМ	Cycles per Minute
DLR	Detection Limit for the purpose of Reporting
DPM	Disintegrations per Minute
F	Filtered
GPS	Global Positioning System
KLTN	Kelantan
LSC	Liquid Scintillation Counter
MCL	Maximum Contaminant Level
MMMP	Multi-Media Mitigation Program
ND	Non Detectable
NIRS	National Inorganics and Radionuclide Survey
NORM	Naturally Occurring Radioactive Material
NURE	National Uranium Resource Evaluation
SSI	Swedish Radiation Protection Institute
UF	Unfiltered
UNSCEAR	United Nations Scientific Committee on the Effect of Atomics
	Radiation
USEPA	United States Environmental Protection Agency
WHO	World Health Organisation
yr	Year

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CHAPTER 1

INTRODUCTION

1.0 Introduction

Most sources of drinking water for rural people, especially those living far inland and are still lagging behind in development, most of them get the source of drinking water from groundwater and surface water. Some of the drinking waters are obtained from groundwater sources such as springs, wells and boreholes. Meanwhile, surface water source is obtained from rivers, lakes and streams. Normally, drinking water from groundwater sources has higher concentrations of radon and radium than surface water.

Radon is a radioactive nuclide which is quite short-lived, half of a given amount decay is only 3.82 days. It is also a chemically inert gas and highly soluble in water. The presence of radon in the environment it as the most dangerous radioactive elements than the others. It also do not combine with any particular element and thus move freely in the ground. It is produced continuously in rocks and minerals. Radon is produced through α -decay of Radium-226 and it is a daughter product of the Uranium-238 natural decay series. Radon-222 is easily soluble in water under pressure, especially well water since it is one of the sources of groundwater. High concentrations of ²²²Rn are more worrisome to the water users, especially its effects on health to the residents. By drinking water from groundwater, it will increase the risk of human exposure to radiation effects. Almost 90% of stomach cancer deaths caused by inhaling radon released to the indoor air from water. Only about 10% of the deaths were from cancers of internal organs, mostly the stomach, caused by ingestion of radon in water (USEPA, 1999). Cothern *et al.* (1986) proved that there was a risk of death of about 1-7% from lung cancer for the people of the United States. Most cancer associated with indoor radon levels in buildings due to groundwater resources.

In most countries, the level of radon concentrations measured in the water supply used by the people can usually reached 20 Bq/l. But there are some cases in some countries where it is above 100 Bq/l. However, the results of epidemiological studies found no association between radon content in drinking water and cancer of the digestive system and other systems until now. The World Health Organisation (WHO) has recommended guidelines for the quality of drinking water with the amount of radon in public drinking water sources to be above 100 Bq/l (WHO, 2009).

²²⁶Ra is parent of ²²²Rn and it is an alpha emitter with a half-life of 1600 years. It can be found in groundwater and it is supplies a continuous source of ²²²Rn. The concentration of ²²⁶Ra can reach up to a few Bq/l and its present in the system depends on the availability of water or rock. Most people are concerned about the presence of ²²⁶Ra in drinking water as it is the main source of water in their daily activities. United State Environmental Protection Agency (USEPA) has reported that the Maximum Contaminant Level (MCL) for the combination of the two radionuclide, ²²⁶Ra+²²⁸Ra reported as 0.185 Bq/l. However, ²²⁶Ra in drinking water have been reported to be as much as 0.110 Bq/l (USEPA, 2000). Known, ²²⁸Ra is owned by Thorium-232 series and it decays by beta emission. ²²⁸Ra is controlled by the low solubility of thorium and it is transported on solids. Meanwhile ²²⁶Ra is owned by ²³⁸U series and it decays by alpha emission. ²²⁶Ra can also be transported by water flow by long-term transport of some fertilizers into the waterways. If ²²⁶Ra

is present within large amounts in drinking source, it is dangerous as it is the toxic chemical (Zhuo *et al.*, 2001). Among some natural radionuclide present in water resources, only ²²²Rn and ²²⁶Ra can be dangerous to the people who use the water sources in daily activities especially when it is used as drinking water.

When the concentration of ²²⁶Ra is at a high level, it will increase health risks, especially its potential as bones seeker. This is a worrying situation especially when it involves the ²²⁶Ra content in private well water, where the people who use the water are less concerned about the presence of dangerous radionuclide such as ²²⁶Ra in the water. In addition, the rural people are less concerned with the treatment to remove radionuclide in view of the costs payable by them. A survey was conducted by the Swedish Radiation Protection Institute (SSI) indicated that the maximum concentration of radium in groundwater in Sweden was up to 2.5 Bq/l (SSI, 1996).

There are various methods practiced to analyze the ²²²Rn and ²²⁶Ra content in water sources. However, the most widely used method is to use the Liquid Scintillation Counter (LSC). This is because, conventional LSC has an advantage in terms of performance, especially if it involves counting large numbers of samples and analysis of samples can be run in a short period of time. In addition, it can also be used to measure low concentrations of ²²²Rn.

One of the aims of this study is to measure the concentrations of 222 Rn and 226 Ra in groundwater from well water by using liquid scintillation techniques. There are many studies that have been conducted using this method, especially for the analysis of aqueous samples of water taken by the many authors (for example Schönhofer, 1989; Beata Kozlowska *et al.*, 1999; Chalupnik *et al.*, 1992; Escobar *et al.*, 1996; Suomela, 1993).

This study focuses on natural radioactivity content in well water samples, collected from several districts in Kelantan. The determinations of natural radionuclide in well water are useful to study the environmental sanitation and human health for those who use the well water for their daily activities.

1.2 Statement of Problem

In rural areas, groundwater is more likely to be the source of drinking water, cooking, cleaning, bathing, and also agricultural activities. Nowadays, most people who use sources of groundwater are not aware of the hazards of radioactive sources from groundwater resources. They use the water source to carry out daily activities, without knowing the presence of radioactive sources in the water. Groundwater is being contaminated due to agricultural runoff or disposal of liquid waste. ²²⁶Ra maybe mobile in surficial environment, specifically in the reducing environment but at high level of ²²⁶Ra it can cause health risks. Meanwhile ²²²Rn has the ability to migrate through the pores of the media through by diffusion process, such as through the rock materials. In this study, the method used for the evaluation of ²²²Rn and ²²⁶Ra levels in well water is using LSC.

1.3 Objectives of the Research

The objectives of the study are:

- i) To measure the radioactivity concentration of ²²²Rn and ²²⁶Ra in well water samples collected from Kelantan.
- ii) To estimate the annual effective dose due to the intake of natural radionuclide from well water for three different age groups.

1.4 Statement of Hypothesis

Following are the hypothesis of this study:

- i) There exist of radioisotope ²²²Rn and ²²⁶Ra in groundwater from Kelantan.
- ii) Due to the nature of soil and rock in Kelantan, the activity concentration of ²²⁶Ra is probably high.
- Babies are among those who have a higher annual effective dose from
 ²²⁶Ra than children and adults.

1.5 Scope of Study

A sampling of groundwater for two isotopes of the ²³⁸U series, ²²²Rn and ²²⁶Ra was conducted in 5 districts in Kelantan to determine the concentration of ²²²Rn and ²²⁶Ra in groundwater from rural area for domestic usage. The method used for the evaluation of natural radionuclide in well water is LSC. The estimation of annual effective dose due to the intakes of groundwater was calculated for three different age groups. The dose estimation study is very useful by determining the amounts of radioactivity intakes and their effects to the human health and populations.

1.6 Thesis Outlines

This thesis consists of five chapters. The first chapter consists of introduction, research objectives, importance of this research, and scope of study and research methodology. Chapter two is the literature review. It covers the work of relevant studies carried out. Chapter three explains the methods and equipment used in the experimental work. Chapter four shows the data obtained from the field measurement, statistical presentation of data using histograms, analysis of groundwater using LSC, and estimate the annual effective dose due to the intake of natural radionuclide for three different age groups. Finally chapter five presents the conclusions of the project and the suggestions.

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