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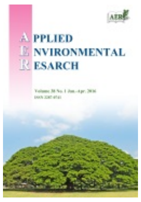

Natural Radioactivity in Groundwater in Phra Nakhon Si Ayutthaya Province

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Abstract

This research aims to study the specific activity of natural radioactivity in groundwater samples taken in the area of Phra Nakhon Si Ayutthaya Province, Thailand. Totally, sixty groundwater samples collected from wells in eight districts were monitored and determined for radionuclides ^{226}Ra, ^{232}Th and ^{40}K and gross alpha and beta were determined using high resolution gamma spectrometer and Canberra Tennelec Series 5 gas flow proportional counter, respectively. Most of the sixty wells serve for water consumption and some other wells for agricultural. The results showed that the average activity concentrations of the gross alpha and beta were 0.01±0.007 and 0.15±0.02 Bq L^{-1} and the specific activity of ^{226}Ra, ^{232}Th and ^{40}K were averagely 0.77±0.13, 1.03±0.19 and 15.56±1.28 Bq L^{-1}, and respectively. The activity concentrations of the gross alpha and beta and the specific activity of the radionuclides in these samples exhibited quite low as compared to the recommended reference level for human consumption reported by World Health Organization (WHO).

Keywords: Natural radioactivity; Groundwater; Phra Nakhon Si Ayutthaya Province

Introduction

Water is the most important source for life and makes up 70-75 % of total body weight. While 70 % of the world's surface is covered by water, only 0.3 % of the total water resources on earth is drinkable and suitable for daily use [1]. For instance, human uses of water are agriculture, industry, consumption, household uses and recreation. Natural disasters have been more severe than before due to environmental balance is gradually destroyed. Significant reduction of rainfall and longtime of dry spells directly affect agricultural and industrial sectors, as well as water consumption. Groundwater resource is an alternative providing leverage to tackle a shortage of water supply in many areas. Groundwater is acknowledged about its risk of natural contamination from mineral dissolution of their host rocks, as well as anthropogenic contamination caused by human activities. Moreover, natural radioactive materials are also concerned, especially using groundwater as water supply in communities. Consumption of contaminated groundwater may cause adverse health effects.

The radioactivity in groundwater occurs mainly from radionuclides of the natural decay chains of ^{238}U, ^{232}Th and ^{40}K. Some radionuclides can be easily dissolved in water depending on mineralogical and geochemical composition of soil and rock, redox condition and residence time of groundwater transporting through soil and bedrock, and reaction of groundwater with soil and bedrock [2]. When the decayed radionuclides are taken into human body through ingestion or inhalation, radionuclides will cause internal exposure [3]. Ingested radionuclides are absorbed into blood and accumulated in specific tissues causing damages [4]. Some radioactive materials can cause toxicity to kidney and increase risk of cancer.

Due to data of radioactive concentrations in groundwater in Thailand are quite rare, this study is to monitor natural radioactivity occurrence in groundwater located in Phra Nakhon Si Ayutthaya Province, Thailand. The objective of this study is to evaluate the gross alpha and beta activity concentrations and natural radionuclides activity concentration levels (^{226}Ra, ^{232}Th and ^{40}K) for consideration. The gross alpha and beta activity concentrations and radionuclides specific activity were determined using Canberra Tennelec Series 5 gas flow proportional counter and high resolution gamma spectrometer, respectively. In addition, the observed data in this study was useful for evaluation of radioactive contamination level in these groundwater samples.

Material and methods

1) Monitoring design

The study area in this work is located in Phra Nakhon Si Ayutthaya Province, 76 km northern distance from Bangkok, covering an area of 2,556 km^2 and dividing into sixteen districts. This province is located in the flat river plain of the Chao Phraya River valley. The presence of four rivers, i.e. Chao Phraya River, Pa Sak River, Lopburi River and Noi River, flowing through the city makes this province a major rice farming area of Thailand. Due to drought phenomena have recently occurred, groundwater is widely used throughout the province as an alternative water reserve. Groundwater in this province serves for both consumption and agricultural purposes. Sixty wells were designated for groundwater samples collection during May to October, 2016 as illustrated in Figure 1. Most of designated wells serve for consumption purpose through community water supply system. At least 2 L of groundwater were taken for each sampling. Then, they were preserved with 1N HCl to pH 2 in order to avoid loss of radionuclide fraction by adsorption with the container and to prevent some biological activities according to EPA methods of 900.0 and 901.1.

2) Determination of gross alpha and beta radioactivity

Approximately 1 L of each sample was filtered via filter paper (Whatman No.1) and put into 2-L beaker. The filtrate with certain volume was further evaporated on hot plate until almost dry and its residue appeared. Then, residue was transferred to clean planchet and further drying to evaporation under infrared lamp. After drying process, residue was weighted and stored in desiccator until analytical measurement. Weighted residues were analyzed for the radiation (in terms of gross alpha and beta) by Canberra Tennelec Series 5 gas flow proportional counter with counting time of 120 min per sample. In addition, the control sample (or blank sample) was required and had to be placed in the front sequence of sample series during the measurement. The gross alpha and beta counting system were calibrated using planchets containing of ^{241}Am and ^{90}Sr/ ^{90}Y standard sources, respectively. The counting efficiencies for the system were 23 % for alpha and 36 % for beta. Finally, the measurement result was used to calculate for radioactivity with the following equations (Eq. 1) as shown below.

3) Determination of radioactivity of radionuclides (^{226}Ra, ^{232}Th and ^{40}K)

One liter of each water sample was transferred into a Marinelli beaker. The beakers was subsequently firmly sealed for at least 4 weeks to ensure a state of secular equilibrium between radium isotopes and their respective daughters radionuclides measuring of gamma radiation [5]. The sample was analyzed for ^{226}Ra, ^{232}Th and ^{40}K using a high resolution gamma spectrometer. The efficiency calibration of the detector was performed using multinuclide distributed in 1.0 g cm^{-3} epoxy matrix calibration standards with approximate volume of 1 L in a Marinelli beaker. The efficiency calibration of the detector was performed by Cs-137 and Co-60, which the energy peak of Cs-137 was 662 keV and those of Co-60 were 1173 keV and 1,333 keV. Each water sample in a Marinelli beaker was determined for radionuclides using high resolution gamma spectrometer counting for 50,000 s. A result of which was further calculated for radioactivity with the equations (Eq. 2) as shown below.

4) Annual dose calculation

In this study, annual dose ingestion of ^{226}Ra contamination in groundwater was considered. Therefore, the radioactivity dose from annual consumption in each adult was estimated according to the following equation (Eq. 3) [6]; considering with consumption rate of 2 L d^{-1} and the conversion factors of 0.28 μSv Bq^{-1}.

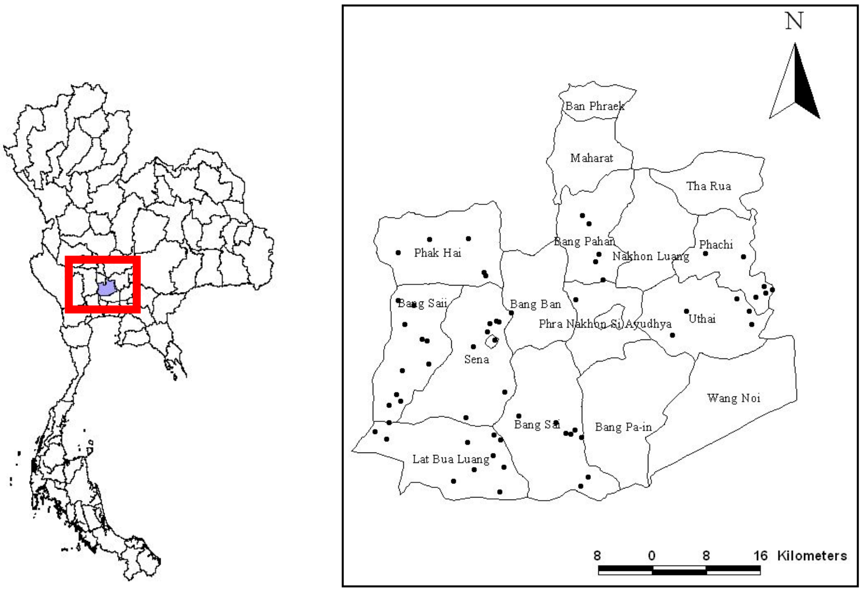

Figure 1 Sampling sites in Phra Nakhon Si Ayutthaya Province.

$$\text{Activity concentration (Bq L}^{-1}\text{)} = \frac{(A_s - A_b) \times 100}{\% \, \varepsilon \times V \times 60} \qquad \text{(Eq. 1)}$$

Where A_s is count rate of samples (cpm), A_b is count rate of background (cpm), % ε is efficiency percentage of the detector, V is volume of water (L) and 60 is conversion factor from dpm Bq^{-1}.

$$\text{Activity Concentration (Bq L}^{-1}) = \frac{N_{(cps)}}{\varepsilon \times V \times P_{\gamma}(E)} \quad \text{(Eq. 2)}$$

Where $N_{(cps)}$ is net counts of radionuclide in the sample (count per second), ε is the efficiency of energy detector, V is volume of water (L) and $P_{\gamma}(E)$ is opportunity to decay and emit gamma ray energy.

$$\text{Effective dose (mSv a}^{-1}) = A_w \times IR_w \times ID_F \quad \text{(Eq. 3)}$$

Where A_w is activity concentration (Bq L^{-1}), IR_w is intake of water for person in 1 year (730 L a^{-1}) and ID_F is the effective dose equivalent conversion factor (μSv Bq^{-1})

Results and discussion

The activity concentrations of gross alpha and beta and specific activity of the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in groundwater samples in Phra Nakhon Si Ayutthaya Province were concluded in Table 1. From the results, pH values of sixty samples were observed in the range of 6.7-8.4, most of which were in the neutral pH with the average of 7.6. Their conductivities were in the range of 232 - 3946 μc cm^{-1} where their average value was 902 μc cm^{-1}.

The activity concentrations of gross alpha were found in the range of 0.009-0.029 Bq L^{-1}, as well as those of gross beta were in the range of 0.063-0.345 Bq L^{-1}. The averages of activity concentration of gross alpha and beta observed in this area were 0.011±0.007 and 0.156±0.018 Bq L^{-1}, respectively, which were comparatively lower than their recommended reference levels by WHO of 0.5 and 1 Bq L^{-1}, respectively (Figure 2). Especially, the maximum concentration of gross alpha (0.029 Bq L^{-1}) was considered as only 6 % of the reference level (0.5 Bq L^{-1}). However, the maximum concentrations of gross beta (0.345 Bq L^{-1}) were in considerable percentage (about 35 %) of the reference level of 1 Bq L^{-1}.

1) Radium-226

The concentrations of radionuclides ^{226}Ra, ^{232}Th and ^{40}K were found in the ranges of 0.520-0.921, ND-1.033 and 12.9-18.8 Bq L^{-1}, respectively (Figure 3). Their averages of activity concentrations were 0.77±0.13, 1.03±0.19 and 15.6±1.29 Bq L^{-1}, respectively. In case of ^{226}Ra, their activity concentrations were observed in the range that its maximum concentration (0.921 Bq L^{-1}) was much closed to the recommended level of 1 Bq L^{-1} for water consumption announced by WHO.

Table 2 gathered several reports of radio-activity monitoring in various areas in order to compare with our study. Similarly, Zhuo et al. (2001) [7] reported the maximum value of 0.93 Bq L^{-1} was found in groundwater sample taken in Fujian province (Table 2). Nevertheless, some samples contained higher concentrations of ^{226}Ra were observed in groundwater samples taken in Egypt [8] and Yemen [9] and Serbia [10].

Table 1 Activity concentration (Bq L^{-1}) of natural radioactivity and associated characteristics in groundwater samples collected in Phra Nakhon Si Ayutthaya Province during May to October 2016

Parameter	Concentration			Standard deviation		
	Max	Min	Average	Max	Min	Average
Gross Alpha (Bq L^{-1})	0.029	0.009	0.011	0.010	0.004	0.007
Gross Beta (Bq L^{-1})	0.345	0.063	0.156	0.024	0.013	0.018
^{226}Ra (Bq L^{-1})	0.921	0.520	0.767	0.166	0.110	0.133
^{232}Th (Bq L^{-1})	1.033	ND	-	-	-	0.194
^{40}K (Bq L^{-1})	18.8	12.9	15.6	3.635	0.912	1.290
pH	8.4	6.7	7.6	-	-	-
Conductivity (μc cm^{-1})	3946	232	902	-	-	-

ND is Not Detectable

Table 2 The activity concentration of natural radioactivity in Bq L^{-1} of water samples reported in various studies

Country	Type of water	Activity concentration (Bq L^{-1})					Ref.
		Alpha	Beta	^{226}Ra	^{232}Th	^{40}K	
Turkey	Groundwater	0.08-0.38	0.12-3.47	-	-	-	[2]
Hungary	spring waters	0.03-1.75	0.03-2.01	-	-	-	[11]
Bangladesh	Surface water	0.45×10^{-3} - 1.36×10^{-3}	0.06-0.28	-	-	-	[12]
Nigeria	Underground water	0.31-14.49	0.02-27.5	-	-	-	[13]
China	Groundwater	-	-	0.001-0.94	-	-	[7]
Roi-Et Province	Groundwater	-	-	<0.006- 0.177	-	-	[14]
Egypt (Elba)	Groundwater	-	-	1.6-11.1	0.21-0.97	9.1-23	[8]
Yemen	Groundwater	-	-	0.86-3.09	0.46-2.01	7.84-18.02	[9]
Jordan	Hot spring water	-	-	3.8–6.8	1.42–2.37	23.2-34.8	[15]
Serbia	Groundwater	0.001-1.33	0.02-5.43	0.005-2.56	0.006-0.79	0.012-2.6	[10]
Phra Nakhon Si Ayutthaya	Groundwater	0.009-0.029	0.063-0.345	0.520-0.921	N.D.-1.033	12.9-18.8	Present work

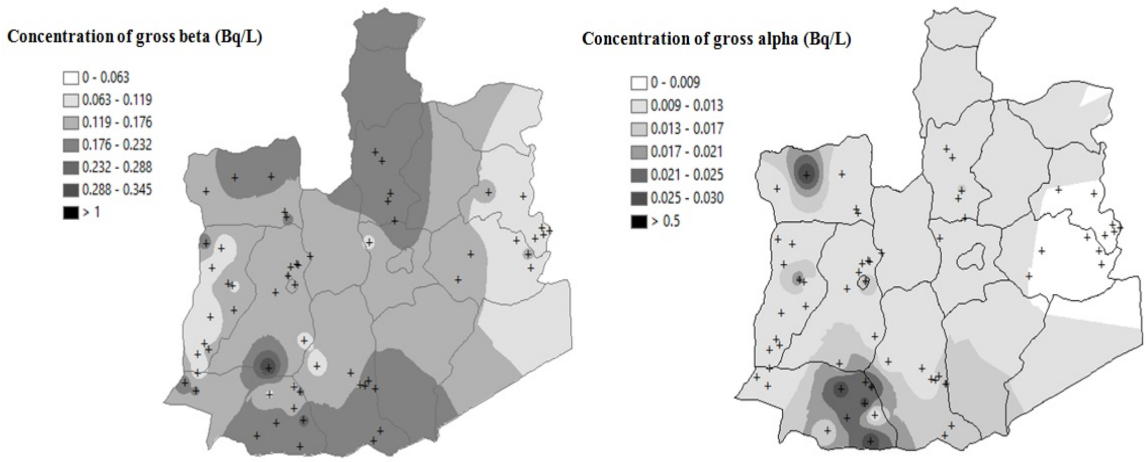

Figure 2 Interpolated radiological maps for gross alpha and beta in Phra Nakhon Si Ayutthaya Province.

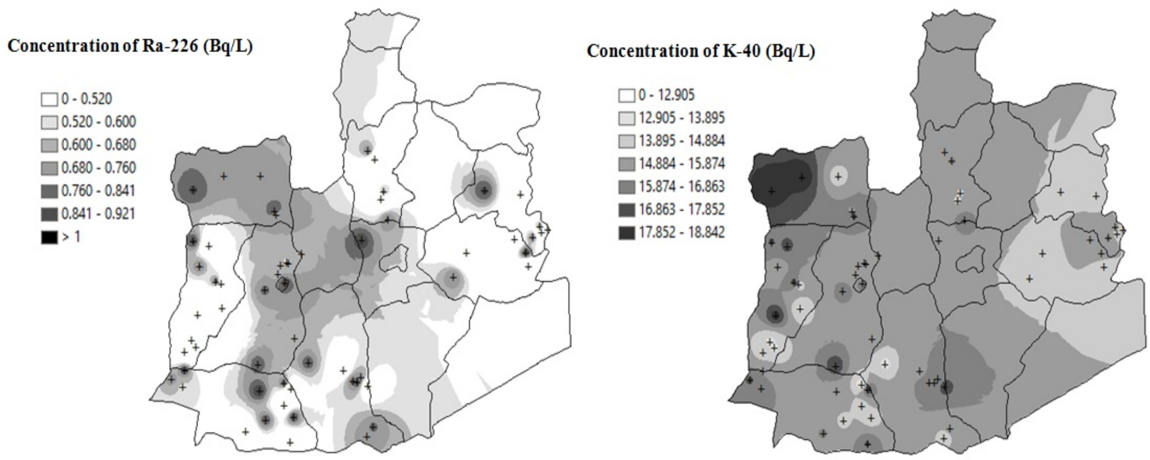

Figure 3 Interpolated radiological maps for ^{226}Ra and ^{40}K in Phra Nakhon Si Ayutthaya Province.

However, there is a report of ^{226}Ra monitoring in groundwater samples taken in Roi-Et Province, northeastern part of Thailand [14]. Those samples found in the range of <0.006-0.177 Bq L^{-1}, which were lower than in our study. This was probably due to different geology constituting water-bearing formations of groundwater system. Geological map of Thailand 1: 250,000 of Phra Nakhon Si Ayutthaya Province [16] illustrated some of igneous rocks in its geology, but there was none shown in that of Roi-Et Province [17]. Some literature reviews mentioned that ^{226}Ra and ^{228}Ra levels in ground-water related to certain types of rock (e.g. granite, sandstone) or rocks dissolution [18]. Anyway, some admitted that wide ranges of radioactivity were observed with little correlation to the type of rock or sediment constituting aquifer formation [19].

2) Thorium-232

In case of ^{232}Th (radiological maps were not shown here), there was no data of ^{232}Th concentrations in the study of groundwater in Roi-Et Province [14]. Usually, Th content in crustal rocks is observed at low level. In addition, Th typically forms complex compounds with ions in water either acidic or basic conditions. ^{232}Th concentrations observed in this study were averagely 1.0 ± 0.19 Bq L^{-1}, similar range of which was reported in groundwater samples collected in Yemen [9] and hot spring water samples in Jordan [15]. In contrast, the much lower concentrations of ^{232}Th in groundwater were mentioned in samples taken in Serbia [10].

3) Potassium-40

The ^{40}K concentrations monitored in this study were in moderate level in comparison to elsewhere [8-10, 15]. Theoretically, the abundance of ^{40}K activity observed in groundwater was usually explained by the relevance to potassium fertilizer application in agricultural activities and transportation through groundwater system. Anyway, higher concentrations of ^{40}K were found in hot spring water samples studied in Jordan [15]. Similar concentrations were observed in groundwater samples monitored in Yemen [9]. Much lower concentrations were reported in a study of groundwater in Serbia [10]. It is possible that moderate concentrations of ^{40}K activity found in this study area might be resulted from potassium fertilizer application. More statistical data of background level of ^{40}K activity and fertilizer application in Phra Nakhon Si Ayutthaya Province are required for in depth discussion.

4) Annual dose of ^{226}Ra ingestion

The annual dose calculation for adults, considering only ingestion from ^{226}Ra by the water consumption rate is 2 L d^{-1} in this study were found in the range of 0.106-0.188 mSv a^{-1}. Although ^{226}Ra level found in groundwater samples taken in Phra Nakhon Si Ayutthaya Province during May to October, 2016 (Table 2) complied with WHO guideline of 1 Bq L^{-1}, all results of annual dose calculation exceeded the individual dose criteria (IDC) of 0.1 mSv a^{-1} suggested by WHO (2011) [20]. However, this screening level for drinking water usually applied if either gross alpha activity or gross beta respectively exceeded 0.5 and 1 Bq L^{-1} guidance concentrations, which those in this study were not in the case (Table 2). Anyway, this study attempted the annual dose calculation for ^{226}Ra in order to compare with the study in Roi-Et province [14]. Thus, the annual doses of ^{226}Ra in our study were higher than those in Roi-Et province due to higher ^{226}Ra concentration as previously mentioned. The most frequency of 38 % (of total 60 data) were observed in range of 0.101-0.120 mSv a^{-1} as shown in Figure 4. Nevertheless, WHO [20] also suggested that this IDC (0.1 mSv a^{-1}) should be determined only in case of any radionuclide was to exceed the guideline concentration; consequently resulting in summary annual dose might exceed the IDC. Then, the continuation monitoring for a whole year should be applied before concluding that the water is unsuitable for consumption.

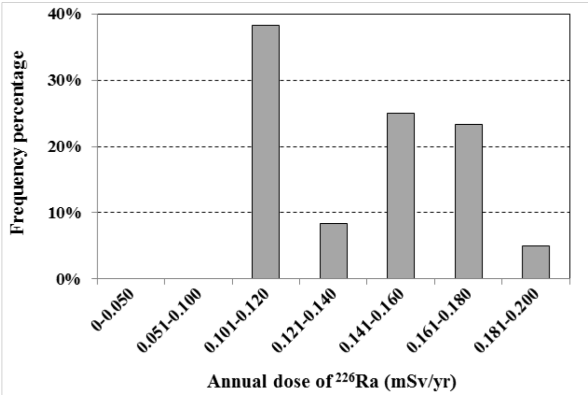

Figure 4 Frequency distribution of effective dose for ^{226}Ra in groundwater in water sample.

Conclusion

The natural radioactivity levels of gross alpha and beta and radionuclides: ^{226}Ra, ^{232}Th and ^{40}K observed in groundwater samples taken in Phra Nakhon Si Ayutthaya Province during May to October 2016 considerably complied with the guidance level for drinking water quality recommended by WHO. The ^{226}Ra levels in this area were in concern where their annual dose calculation resulted in exceeding the individual dose criteria adopted by WHO. However, this was too early to conclude these groundwater samples were unsuitable for drinking purpose. Different seasonal monitoring will be further studied and their results will be compared and discussed with data presented here. Therefore, the annual dose for ingestion of the radionuclides found in groundwater will be more evaluated and discussed.

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