Determinaton of internal and external hazard index of natural radioactivity in well water samples

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Abstract. Well water is an environmental component that is always used by people but is vulnerable to contaminated. The impact of the contamination of water bodies could cause danger to human health. Water pollution could be caused by radioactivity that exceeds recommended limits. Source of radionuclide pollutants in water bodies could be caused by the operation of nuclear facilities. Analysis of radioactivity has been carried out on several samples of well water near the TRIGA 2000 reactor facility, followed by calculating the internal and external hazard index of radiation dose. The results of the analysis using a gamma spectrometer show that the radionuclides contained in well water samples are not fission products, but they are Ra-226, Bi-214, Pb-214 and K-40 in the form of natural radionuclides. The highest radioactivity in the sample was 5.808 Bq/L from K-40 and the lowest was 0.103 Bq/L from Bi-214. The internal and external hazard index has been determined where the internal hazard index was 0.013-0.023 while the external hazard index was 0.006-0.012 which is below the maximum hazard index value based on UNSCEAR which is ≤ 1. So, based on the results of this study on the radioactivity parameters show that well water is still safe to be used.

1. Introduction

Water could not be separated from human life and it has been becoming the most important part of humans. Through water usage in daily life such as cooking, bathing, drinking, and so on sustainably, thus people often use groundwater as clear water sources to accomplish their needs. Groundwater could be whether deep wells water or shallow wells water. Dug wells define as one of the good constructions used by small people and individual houses generally and widely. The depth of ground wells water ranged among 7-10 meter from ground level, whereas groundwater from the artesian well (pump) clarify as groundwater layer that obtained from deeper drilling or subsoil that is far from the surface. It is undeniable if the water is vulnerable to contamination. It could be caused by several reasons such as industrial waste disposal, environmental pollution, acid rain, and any other pollutants, including radionuclide activity. Radioactivity derived from nuclides with an unstable condition could emit radiation in the form of alpha, beta, and gamma. Radionuclides themselves could come from artificial and naturals.

Naturally occurring radioactive material (NORM) was the source of natural radiation found in the circle of occurring along with the universe. Radionuclide which categorized as NORM are usually derived from U-238, Th-232, K-40, Ra-226, Rn-222, and Rn-220 [23]. Whilst, technologically enhanced occurring radioactive materials (TENORM) are radionuclide that enhanced its concentration or exposed
to the environment since human activities employ raw material from the earth’s crust. TENORM can be produced from industrial activities that utilize isotope also through other activities at the nuclear facility. Refers to Rohanda (2013), radionuclide which generated on nuclear fission reaction could be form as Sr-90, Cs-137, Pu-238, Pu-239, Pu-240, Am-241, and Am-243 [10].

Natural radionuclides might be classified as hazardous and toxic materials, due to its character and concentration, both direct and indirect endanger the health of human life and other living creatures [21]. Radionuclide brings external radiation hazard and internal radiation hazard toward the human. In terms of external radiation hazard is gamma radiation which radiated from each radionuclide, whilst internal radiation danger could be caused by the process of radioactive ingestion and inhalation which are radionuclides as a result of uranium and thorium decay. The radiation hazard can be seen based on the radiation hazard index [5].

As naturally occurring radionuclides belonging to the uranium and thorium series present in drinking water usually give radiation doses higher than those provided by artificial radionuclides, and it could be a greater concern. Consequently, a study of natural radioactivity analysis in a well water sample was necessary. The aim of this study was to determine radioactivity in a well water sample followed by determining the internal and external hazard index of its radioactivity accepted by people.

2. Materials and method

2.1. Material and apparatus
This study has been conducted with the following materials and apparatus as follows: Well water samples, pH universal, filter paper, seal tape, jerrycans 5 L, beaker glass 1000 mL, and graduated cylinder 500 mL, spatula, funnel, Marinelli (sample container) 500 mL, Gamma-ray reference standard in point geometry (Am-241, Co-60, and Cs-137) from LMRI, multi nuclides standard 500 mL in epoxy matrix within Marinelli geometry from Eckert & Ziegler, GPS Garmin model and Montana 650 type, oven Memmert model, an unit of gamma spectrometer from Canberra consist of a high purity germanium (HPGe) coaxial detector type (model GX2518) with gamma energy resolution (FWHM) of 1.87 (1333 keV of Co-60) associated with preamplifier model 2002CSL, amplifier model 2022, 16 k digital multichannel analyzer multiport II, and Genie 2000 gamma spectra analyzer software. To ensure a low background environment, the HPGe detector was enclosed within a 10 cm lead shield layered internally with 2 mm copper.

2.2. Method

2.2.1. Survey and sampling. A Survey has been carried out to determine the availability of well water used by people near the TRIGA 2000 reactor facility. Sampling points have been determined to represent the four cardinal directions. Well water samples were taken from various regions that represent every direction wherein the TRIGA 2000 reactor served as the center point. As much as 5000 mL from each well water sampling location was gathered then it put into jerrycans then brought it to the laboratory.

2.2.2. Preparation of the sample. The pH of the sample has been checked using pH universal, then preserved by adding nitric acid until pH 2 for preventing absorption toward wall containers. Secondly, the well water sample was filtered to separate it from rough materials. Water samples gained as much as 5000 mL concentrated into 500 mL by using the oven at temperatures of 150-175 °C in order to accelerate evaporation. Afterward it was cooled down to room temperature, then put it in 500 mL Marinelli. In the last step, it was sealed by using seal tape then stored for one month to reach secular equilibrium.
2.2.3. **Calibration of gamma spectrometer**

2.2.3.1 **Energy channel and efficiency calibration.** At the beginning of the radionuclide analysis process by using a gamma spectrometer, two calibrations were performed, they are energy calibration and efficiency calibration. Each calibration process was held to determine that the gamma spectrometer instrument could be employed to measure the sample adequately.

In this study, energy calibration was carried out using gamma reference sources Am-241, Cs-137, and Co-60 that have been properly certified energy values. The standard has been counted for 300 seconds. Thus, it can convert channel numbers into energy values in terms of keV. The function was to represent the differences in each of the energy (low, medium, and high energy). Am-241, Cs-137, and Co-60 standard source that has been employed. The next step was processing and setting the spectrometer system by applying Genie 2000 software. It was gained four spectra that appear, in which two spectra have belonged to Co-60. Those two spectra represent high energy which was 1173.4 energy and 1332.4 KeV. While both medium energy and low energy were represented by one spectrum of each energy that was 661.6 keV belongs to Cs-137 and 59.6 keV belongs to Am-214. The data of energy calibration can be seen in table 1.

### Table 1. Data of gamma energy calibration.

| Energy (keV) | Channel |
|-------------|---------|
| 59.6        | 281     |
| 661.6       | 3111    |
| 1173.4      | 5515    |
| 1332.4      | 6267    |

By referring to table 1, a curve has been created. Energy calibration was conducted to determine the correlation between channel numbers also called channel as y and gamma energy (keV) as x. The curve can be seen in figure 1.

![Energy calibration curve](image-url)

**Figure 1. Energy calibration curve.**

The curve obtained was a linear model of equation of \( y = ax + b \) with the value of \( R^2 = 1 \). The regression value of 1 indicates that the correlation between channel and energy was a straight linear line. It can be concluded that the detector is still valid for analyzing samples at low, medium, and high energy.

The calibration efficiency by utilizing the standard 500 mL multi-nuclides in the epoxy matrix in a Marinelli container that has been determined based on the certified reference with known activity values and uncertainty of 3.0 – 3.1% from each nuclide (supplied by Eckert and Ziegler, USA). Nuclides spectra
were found in this standard source include Cd-109, Co-57, Te-123m, Cr-51, Sn-113, Sr-85, Cs-137, Y-88, and Co-60 which represent low energy range (88 keV from Cd-109) to high energy (1333 keV from Co-60). The standard was counted for 61200 seconds. Furthermore, the count value was obtained from the nuclide spectra processed by Genie 2000. Equation (1) was used to get the value of measurement efficiency of the detector:

$$\varepsilon = \frac{\text{cps}}{A \cdot \gamma \cdot V} \cdot 100\%$$  \hspace{1cm} (1)

In the manner of:

- A = standard activity (Bq)
- Iγ = emitted gamma intensity (%)
- cps = count rate
- V = volume (L)
- ε = efficiency (%)

Absolute efficiency of detector values for all nuclide energy which arises was needed. It could be performed by interpolating from values obtained. Hence, the equation from its interpolation will be employed in sample quantitative analysis.

Efficiency calibration was carried out by employing a multi-nuclide standard in the epoxy matrix. Exclude having a density that closes to water that was 1 which suitable with the samples of this present study, epoxy also accomplished the standard materials requirements. It required for stability and sustainability thus it would be used in a long period of time relatively. Unlike water that was easily affected by environmental conditions. If gamma rays hit a material, it will experience a reduction in intensity caused by its absorption (matrix). Gamma attenuation correction factors are of major concern for precise gamma spectrometry. It caused by systematic uncertainty in the full energy peak efficiency due to the differences between the matrix density and chemical composition of the reference material and the other bulk samples [7], [8]. The amount of materials absorption named the self-absorption coefficient. Moreover, Therefore, it was necessary to analyze the efficiency value of the detector by using a standard source identical to the samples. Furthermore, spectra arising from the sample have a different energy, although it was not significantly. So, the fitting function of its efficiency was required [9].

The efficiency calibration curve can be seen in figure 2.

**Figure 2.** Calibration curve of efficiency.

Based on the result of standard source assay, we obtain two types of efficiency curves, first was the power curve for a range of gamma energy 88 - 159 keV represented mathematically by the equation of:

$$y = -2E-06x^2 + 0.0004x + 0.0371$$  \hspace{1cm} [Eγ 88 - 159 keV]

$$y = 3.6811x - 0.849$$  \hspace{1cm} [Eγ 122-1333 keV]

$$R^2 = 1$$

$$R^2 = 0.9858$$
and second was the polynomial curve for a range of gamma energy 122 - 1333 keV represented mathematically by the equation of:

\[ y = 3.6811x^{-0.849} \]  

In term of y value was efficiency and x represent gamma energy. Nevertheless, both of them have a regression value close to 1. It pointed out the strong correlation between efficiency value toward energy. Hence the equation could be utilized to find nuclide efficiency on the samples.

In order to determine the background interference in the environment around the detector, an empty Marinelli was assayed in the same manner as the samples. The background spectra were used to correct the net peak area of gamma rays measured from samples.

2.2.4. Analysis of samples. The previous steps such as sample preparation, energy and efficiency calibration were passed. Thus well water was ready to be counted up by utilizing gamma spectrometer within 61200 seconds. The activity of each radionuclide was counted using equation (4):

\[ A = \frac{cps}{\varepsilon \cdot I_y \cdot V} \]  

In the manner of:
- \( A \) = standard activity (Bq)
- \( I_y \) = emitted gamma intensity (%)
- \( cps \) = count rate
- \( V \) = volume (L)
- \( \varepsilon \) = efficiency (%)

2.2.5. Determination of internal hazard index (\( H_i \)). The internal hazard is represented by \( H_i \) [3]. Internal radiation exposure which is defined in relation to an internal hazard could be calculated through the internal hazard index value by employing the equation (5) [1], [4].

\[ H_i = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \]  

In terms of \( A_U \) means uranium activity, \( A_{Th} \) means thorium activity, and \( A_K \) means potassium activity.

2.2.6. Determination of external hazard index (\( H_e \)). To calculate the external hazard index of gamma radiation, it could refer to the equation (6) [6]:

\[ H_e = \frac{A_U}{370} + \frac{A_{Th}}{258} + \frac{A_K}{4810} \]  

In terms of \( A_U \) means uranium activity, \( A_{Th} \) means thorium activity, and \( A_K \) means potassium activity.

3. Results

3.1. Survey and determination of sampling location
The use of well water by the community began to decrease, especially in urban areas. This is due to the availability of service installations from providers that supply clean water to urban homes. The water source can come from mountain springs or the results of processing (recycling). However, there are still people who use well water as a source of water for their daily activities. This is evident from the results of a survey that has been conducted.
In this study, especially the well water samples were taken from four locations which represent each cardinal direction towards the TRIGA 2000 reactor facility as a central point. Well water samples were taken from four different residential areas near the TRIGA 2000 reactor facility. The map of the sampling point can be seen in Figure 3 and sampling points are in where the coordinate data can be seen in table 2.

![Map of well water sampling location](image)

**Table 2.** Coordinate data of a well water sampling location.

| Sample ID | Coordinate | Resident area       |
|-----------|------------|---------------------|
|           | Latitude   | Longitude           |                      |
| SWB       | 06°53’11,8”| 107°36’17,9”        | Kelurahan Cipaganti |
| SWS       | 06°53’39,5”| 107°36’29,8”        | Kelurahan Lebak Siliwangi |
| SWT       | 06°53’09,3”| 107°37’11,8”        | Kelurahan Sadang Serang |
| SWU       | 06°52’47,1”| 107°36’34,8”        | Kelurahan Dago        |

The sampling points were determined based on the possibility of a critical pathway for radionuclide dispersion from the TRIGA 2000 reactor facility. It could be from the direct release into the airborne then deposited either wet or dry manner. In addition, direct leakage from the facility then carried away into deep groundwater. Well water availability and the possibility of its use by the people for several activities such as bathing, washing, cooking, and even being used as a reserve source of drinking water was chosen. The depth from the ground surface of well determined as the sampling location varies, which is 2 - 30 meters and also the distance to the center point, which is 298 - 1390 meters. But the pH
values of all samples are the same, which is 7. Its data were obtained from the survey result that was conducted before the sampling is carried out shown in table 3.

**Table 3. Utilization of well water samples.**

| Sample | Utilization                          | Depth (meter) | Distance (meter) | pH |
|--------|--------------------------------------|---------------|------------------|----|
| SWB    | Bathing, Washing, Cooking            | 5             | 298              | 7  |
| SWS    | Bathing, Washing                     | 2             | 745              | 7  |
| SWT    | Bathing, Washing, Cooking, Drinking  | 10            | 1390             | 7  |
| SWU    | Bathing, Washing, Cooking, Drinking  | 30            | 903              | 7  |

From table 3, we see that all well water taken (SWB, SWS, SWT, and SWU) are has been used by the people for bathing, washing, cooking and drinking, except SWB and SWS were not used for drinking because they are visually quite turbid.

3.2. *Result of sample radioactivity analysis*

Each sample was tested using a gamma spectrometer in 61200 seconds sequentially. A relatively long time was chosen in order to improve detection capabilities and minimize high error values. Considering radioactivity value in the environment samples had tiny value generally that able to improve net peak area, thus it was possible to be detected massively by the instruments.

The result of the sample spectra analyzed by calculating the correction factor of background count to ensure the net count derived from a radionuclide contained in each sample. Qualitative analysis conducted by observing the energy spectrum that appears. Furthermore, quantitative analysis carried out by using the equation of activity calculation and substitution of the equation of efficiency curve from the previous standard.

Table 4 presents the radioactivity concentrations of natural nuclides detected for the well water samples respectively. The observed activities concentration of the radionuclides content in the well water ranged from 1.697 to 3.792 Bq/L for Ra-226, 0.236 to 0.584 Bq/L for Pb-214, 0.103 to 0.117 Bq/L, and 1.081 to 5.808 Bq/L for K-40, respectively.

The major contribution to the terrestrial gamma radiation field comes from K-40 and from radionuclides generated through the sequence of decay transformations of three alpha-emitting primeval radionuclides, i.e. U-238, Th-232, and U-235 [2]. Based on Table 4, it could be discovered that natural radionuclide which dominates well water samples had derivative U-238. It was a primordial radionuclide element and K-40. U-238 had various types of progeny with different half-lives, such as Pb-214 had half-live within 26.8 seconds, Bi-214 within 19.9 seconds and Ra-226 within 1.6 x 10 years. Each sample contained natural radionuclide Ra-226 and K-40.
Table 4. The result data of analyzing qualitative and quantitative samples.

| Sample ID | Nuclides | Progenies | Activity (Bq/L) |
|-----------|----------|-----------|----------------|
| SWB       | U-238    | Ra-226    | 3.342 ± 0.021  |
|           |          | Pb-214    | 0.522 ± 0.003  |
|           |          | Bi-214    | 0.117 ± 0.001  |
|           |          | K-40      | 5.808 ± 0.036  |
| SWS       | U-238    | Ra-226    | 1.697 ± 0.009  |
|           |          | Pb-214    | 0.584 ± 0.003  |
|           |          | K-40      | 1.439 ± 0.008  |
| SWT       | U-238    | Ra-226    | 2.943 ± 0.016  |
|           |          | Pb-214    | 0.459 ± 0.003  |
|           |          | Bi-214    | 0.103 ± 0.001  |
|           |          | K-40      | 5.113 ± 0.028  |
| SWU       | U-238    | Ra-226    | 3.792 ± 0.022  |
|           |          | Pb-214    | 0.236 ± 0.001  |
|           |          | K-40      | 1.081 ± 0.006  |

From table 4 we can see either quantitatively and qualitatively, in all the samples contained a variety of natural radionuclides such as Ra-226, Pb-214, Bi-214, and K-40 with varying values of each. This shows that in the analyzed well water samples there were no artificial radionuclides or critical radionuclides for example Cs-137 and I-131 were released from activities at the TRIGA 2000 nuclear facility.

3.3. Hazard index

Based on activity value was gained, conducted, and calculated in determining the radiation hazard index of U-238, Th-232, and K-40. In this present study U-238 could be represented by the activity of uranium progenies there were Ra-226, Pb-214, and Bi-214 [1]. From each sample, it was found neither the activity of thorium progenies. Therefore, using equation (5) and (6) could be calculated the hazard index of both internal and external.

![Figure 4. Internal hazard index value.](image-url)
Internal radiation explained as the radiation which occurs if human consuming something that emits radiation then the radiation entered and radiated to the human’s body directly and resulted in radiological hazards. Figure 4 yielded that the index hazard of internal radiation of all the samples was on the range 0.013-0.023 value. The highest value belonged to SWB and SWU that was 0.023 while the smallest was 0.013 which showed by SWS samples, also SWB samples which having value 0.022. This indicates that all sample was safe since it had the smallest value below the threshold which recommended by UNSCEAR that was ≤ 1 [6].

In terms of external hazard in this present study was the use of well water as needs accomplishment thus people would interact and exposed with the water directly. If the water had an external hazard index value more than 1 value and it happened to sustain, it would make human body exposure to radiation source directly. It would affect skin health and eye health in a long time.

![Figure 5. External hazard index value.](image)

External radiation hazard index value that exhibited in figure 5 can be concluded that samples were on the range of 0.006-0.012. The highest value was on the SWB sample of 0.012 while the lowest was the SWS sample of 0.006. There are two samples where indicate almost the same value, i.e. SWT and SWU. Those values could be categorized as micro due to threshold that recommended by UNSCEAR was ≤ 1 [6].

4. Conclusion
This present study concludes that radionuclides found in the sample were K-40 and uranium progenies were Ra-226, Pb-214, and Bi-214. The average radioactivity from R-226, Pb-214, and K-40 in the four samples were 2.944 ± 0.017, 0.450 ± 0.003, 3.360 ± 0.020, respectively. Whereas Bi-214 which was only found in two samples was 0.169 ± 0.001. The highest radioactivity value was shown by the SWB sample that was K-40 of 5,808 ± 0.036 Bq/L while the lowest value was showed by the SWT sample that was Bi-214 of 0.103 ± 0.001 Bq/L.

All samples analyzed have a hazard index value both internal and external still below the maximum recommendation (≤ 1), i.e. the highest value for the external hazard index is 0.023 and the internal hazard index is 0.012. This can be interpreted that the estimated internal and external dose received from the use of well water by the public around the TRIGA 2000 reactor facility is still below the 1 mSv/year limit.

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