RADIOLOGICAL HAZARDS ESTIMATION BY NATURAL RADIOACTIVITY IN WATER SAMPLES ALONG BATANG HARI RIVER, JAMBI

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Received: 25-01-2023 Revision Received: 31-01-2023 Accepted: 30-05-2023

DOI : 10.17146/jstni.2023.24.1.6797

Keywords: activity concentration, Batang Hari River, gamma spectrometry, radiological hazard, water. Abstract Illegal mining activity is still operated along the watershed of Batang Hari, which could potentially be contaminated by natural radionuclides, which are Uranium-238 (²³⁸U), Thorium-232 (²³²Th), Radium-226 (²²⁶Ra), Polonium-210 (²¹⁰Po), Potassium (⁴⁰K), and Lead-210 (²¹⁰Pb). This study aimed to measure activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in river water, groundwater, and drinking water samples along Batang Hari River and then calculate radiological hazard parameters in the sample. The sample collects from 3 different locations from 3 sample types Muaro Jambi District, Jambi City, District Batanghari, Tebo District, and Bungo District. The samples were measured using gamma spectrometry High Purity Germanium (HPGe) for 17 hours. The results showed that the average value of the radiology hazard parameters sample consists of Ra_{ea} 0.307653, AEDE that is 1.771 x10⁻⁶, and each external and internal hazard index are 0.00083 and 0.00155, respectively. The radiological hazard parameters in water samples along Batang Hari River, Jambi, are below the recommended limit: Raeq is 370 Bq/L, AEDE is one mSv/y, and hazard index H_{ex} and H_{in} is 1. This study showed that the water is safe and fulfills the radiological aspect of water quality requirements.

INTRODUCTION

Batang Hari River is the longest river on Sumatera Island that flows through two provinces, West Sumatera and Jambi. Batang Hari River becomes a life source for civilians, such as an irrigation water source for rice fields, fisherman's livelihood, baths, wash and outhouses, and a transportation track. It is also used as the primary source of some Regional Water Drinking Companies distributed to civilians and even more for illegal gold mining (1).

Along the Batang Hari River area, illegal gold mining activity still operates. In Batang Hari Regency, illegal gold mining activity still exists at Danau Embat Village, Sungai Ruan Ilir Village, Maro Sebo Ilir District, Muara Tembesi District Rambutan Masam Village, Pasar Muara Tembesi Village, Rantau Kapas Mudo Village. Besides, illegal gold mining activity at Batang Hari operates in Muara Bulian District (2). Tebo Regency also has illegal gold mining activity in Tanjung Pucuk Village, VII Koto District, specifically in the PT Tebo Multi Agro area. Moreover, illegal gold mining activity in Bungo Regency has operated in most subdistricts in Tanjung Gedang Village, Muaro Bungo District (3).

Gold mining produces matters such as Gold (79Au), Uranium (92U), and Lead (26Fe).

Water sources from rivers, even underground located in gold mining areas, could potentially be contaminated by natural radionuclides, including Uranium-238 (²³⁸U), Torium-232 (²³²Th), Radium-226 (²²⁶Ra), Polonium-210 (²¹⁰Po), Potassium (⁴⁰K), and Plumbum-210 (²¹⁰Pb) (4). Generally, radiation exposure from these radionuclides does not cause any health effects instantly, but it can increase cancer risk slowly. Radiation with a low level in the environment can be a minor contributor to cancer risk (5), so we need to be aware of natural radiation received by the body. Uranium isotopes ($^{238}\text{U},\ ^{234}\text{U},$ and $^{235}\text{U})$ are radiotoxic, which may not obey. Some radionuclides with decay chains from ²³⁸U and ²³⁵U had high radiotoxic characteristics (6). Natural radionuclides ²³⁸U, ²³²Th, and ⁴⁰K can enter the body by inhalation or ingestion. Suppose the radionuclide concentration inside the body is in large amounts. In that case, it can cause health effects such as developing cancer, so radionuclide measurement in the study area needs to be done (7).

Measurement of radionuclides ²³⁸U, ²³²Th, and ⁴⁰K for the river, underground, and drinking water and estimation of radiological hazard to evaluate the water quality consumed along the Batang Hari River must be carried out. The priority of this study is to know ²³⁸U, ²³²Th, and ⁴⁰K radionuclide activity concentration in the water and estimate radiology hazard in the water sample, including Ra_{eq}, AEDE, H_{in}, and H_{ex}.

EXPERIMENTAL SECTION

Materials

Water Sample

The water sample measured is from the region that flows by the Batang Hari River, which is river water, underground water, and drinking water.

Source of Radionuclide Standard

The source radionuclide standard is the typical source for gamma spectrometry calibration. It contains ⁶⁰Co, ¹³⁷Cs, ¹³³Ba, ²¹⁰Pb, and ²¹⁴Am radionuclides.

Instrumentation

Gamma Spectrometry

The activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in water samples were then measured utilizing Gamma Spectrometry High Purity Germanium GEM-F5930-1 series, made by ORTEC-USA.

Inspector

Inspector was used as a voltage and pulse amplifier to differentiate the pulse based on the gamma energy.

Computer Set

The computer was used to monitor gamma-ray spectrum countings.

Maestro Software

Maestro software used was Maestro Software for Windows 7.01 version. It is used to identify radioactive materials that radiate gamma rays.

Digital Scales

Digital scales were used to measure mass from a water sample.

Marinelli Tube 1 L

Marinelli tube 1 L was used to place for sample.

Methods

Sample Collection and Preparation

Samples were taken from five districts flowed by the Batang Hari River: Muaro Jambi Regency, Jambi City, Batanghari Regency, Tebo Regency, and Bungo Regency. The water samples, including underground and drinking water, were taken from each river of the regency. A total of 15 water samples, roughly twenty liters each, were collected from the tube from each selected location. All the collected samples were adequately marked for identification. All samples were labeled with code names based on the area sample taken and given a number based on the water source. The sample was given 01 code if the sample was river water, 02 for underground water, and 03 for drinking water. The exact sample code names can be seen in Table 1.



Figure 1. Water Sampling Locations along the Batang Hari River, Jambi.

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INO.	Sample Code	Source	Location	Latituae	Longituae
1.	KJ01	River	Kota Jambi	1°35′14,32′′S	103°35'4,23''E
2.	KJ02	Underground	Kota Jambi	1°35′43,65″S	103°35'2,11''E
3.	KJ03	Drinking	Kota Jambi	1°35′51,22″S	103 ° 35'29,15''E
4.	MJ01	River	Muaro Jambi	1°34′17,12″S	103 ° 30'35,61''E
5.	MJ02	Underground	Muaro Jambi	1°34'3,17''S	103°31'3,88''E
6.	MJ03	Drinking	Muaro Jambi	1°34'0,08''S	103°31'15,22''E
7.	BH01	River	Batang Hari	1°42′11,50′′S	103°6'18,60''E
8.	BH02	Underground	Batang Hari	1°42′8,30′′S	103°6'23,80''E
9.	BH03	Drinking	Batang Hari	1°41′55,57″S	103°6'13,28''E
10.	TB01	River	Tebo	1°34'27,12''S	102°43'21,23''E
11.	TB02	Underground	Tebo	1°34'36,54''S	102 ° 43'15,45''E
12.	TB03	Drinking	Tebo	1°34'32,13''S	102°43'41,11''E
13.	BG01	River	Bungo	1°28′15,27″S	102°8'23,82''E
14.	BG02	Underground	Bungo	1°28′48,33″S	102°8'27,36''E
15.	BG03	Drinking	Bungo	1°28′54,61′′S	102°8'38,42''E

Table 1. Water Sample's Code along Batang Hari River, Jambi

The collected samples were heated until they became 1 L to make the water reach the radionuclide measurement standard of the Environment Safety Laboratory at BRIN appropriately. The water sample was then pondered to ensure the sample mass was 1 L. Afterward, the piece was placed into a Marinelli beaker and sealed for 40 days to establish radioactive equilibrium between radionuclides and their daughter decay so that exposure to gamma radiation could happen. The sampling procedure was done according to the methodologies recommended by BRIN (8).

Background Radiation Measurement

Background radiation was measured by measuring an empty Marinelli beaker using gamma spectrometry for 17 hours. It aimed to know early radiation before calibration and ²³⁸U, ²³²Th, and ⁴⁰K radionuclide measurements of the sample.

Gamma Spectrometry Calibration

Gamma energy calibration measurement was a qualitative analysis that aimed to know the background condition of the standard source counting room. It consists of energy calibration and efficiency calibration. Calibration energy was done to see the relation between the channel and gamma energy (keV) in the authoritative source. Meanwhile, efficiency calibration aimed to know the radionuclide efficiency and energy in the standard reference. They need to be done before radionuclide measurement is done. The quality control procedure was done in this calibration to ensure the tool performs itself (9). The efficiency of each energy has a specific energy and to calculate using Equation [1] (10):

$$\varepsilon_{\gamma} = \frac{N_s - N_{bg}}{A_{t P_{\gamma}}}$$
[1]

Where N_s is the standard count of radiation (count per second), N_{bg} is the background count of radiation (count per second), A_t is the activity in measurement (Bq), and P_V is a yield of gamma energy (%) from the sample.

Measurement of Radionuclide Activity Concentration

Activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K measurements were done using a gamma spectrometry *High Purity Germanium* (HPGe) detector. The measurement was done for 17 hours for each sample. The measurement process and spectrum-controlled analysis using a computer with *Software Maestro for Windows* 7.01 version.

Radioactive concentration measurement in the sample was determined using Equation [2] (10).

$$C_{sp} = C_{avg} \pm U_T$$
^[2]

$$C_{avg} = \frac{N_s - N_{bg}}{\varepsilon_{\gamma} p_{\gamma} m}$$
^[3]

$$U_T = C_{avg} \times \sqrt{\left(\frac{U_N}{N_S}\right)^2 + \left(\frac{U_\varepsilon}{\varepsilon_\gamma}\right)^2 + \left(\frac{U_P}{P_\gamma}\right)^2 + \left(\frac{U_m}{m}\right)^2}$$
[4]

Whereas C_{sp} shows the radioactive matter in the corrected sample (Bq/L), C_{avg} shows the radioactive matter in the average corrected sample (Bq/L). U_T indicates the uncertainty in measurement (L), and $\varepsilon \gamma$ is the efficiency of observed gamma energy (%). M is the sample mass (L), U_N is the uncertainty of the count sample (%); U_{ε} is the uncertainty of efficiency gamma (%); U_p is the yield uncertainty (%), and U_m is the uncertainty of sample mass (%).

Calculation of Radiological Hazard Parameter

The radiological hazards parameter was estimated from the sample's Uranium, Thorium, and Potassium activity concentration. Then, radiological hazards in the sample were previewed through some estimation models; such is the annual effective dose equivalent (*AEDE*), Ra_{eq} , Hex, and H_{in} (11).

The absorbed dose can be calculated from ²²⁶Ra, ²³²Th, and ⁴⁰K concentrations in the water by assuming the other radionuclide, like ¹³⁷Cs, ⁹⁰Sr, and ²³⁵U decay series, can be ignored if the total dose is so tiny toward the environment background. Generally, ²²⁶Ra ²³²Th and concentration estimations are based on radioactive equilibrium condition assumptions by measuring their daughter isotopes directly. The absorbed dose in the air at 1 m height on the surface is not contributed to radiology risk toward the exposed individual. It can be considered AEDE from terrestrial gamma radiation. Using the conversation coefficient, 0.7 Sv/Gy absorbed dose in the air towards the effective dose received by an adult and 0.2 Sv/Gy as outdoor filler factor based on UNSCEAR. AEDE can be determined by using Equation [4] (11):

$$AEDE = D \ x \ 8760 \ x \ 0.2 \ x \ 0.7 \ x \ 10^{-6}$$
 [5]

$$D = 0.462A_U + 0.604A_{Th} + 0.0417A_K \quad [6]$$

Whereas A_U is the value of uranium activity concentration (Bq/L), A_{Th} is the value of thorium activity concentration (Bq/L), and A_K is the value of Potassium activity concentration (Bq/L).

Radium Equivalent Activity (Ra_{eq}) allowed the index universally to analyze radiation exposure of primordial radionuclides (Uranium, Thorium, and ⁴⁰K). Ra_{eq} was calculated from the Bq/L unit activity concentration shown in Equation [7] (11).

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.77A_K$$
[7]

Limit radiation exposure cause natural radioactivity in the sample towards the allowed equivalent dose of 1 mSv/y is the External Hazard Index (H_{ex}). The maximum H_{ex} must be appropriate with the highest Radium Equivalent Radium, 370 Bq/L (12). It can be determined by Equation [8] (11):

$$H_{ex} = \left(\frac{A_U}{370} + \frac{A_{Th}}{370} + \frac{A_K}{4810}\right)$$
[8]

Meanwhile, Internal Hazard Index (*H*_{in}) can be calculated using Equation [9] (11):

$$H_{in} = \left(\frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right)$$
[9]

RESULTS AND DISCUSSION

 238 U activity concentration in the water sample ranges from 0.00±0.00 Bq/L to 1.29±0.72 Bq/L with average 0.266±0.428 Bq/L, 232 Th activity concentration from 0.01±0.12 Bq/L to 0.04±0.10 Bq/L with average 0.025±0.119 Bq/L, and 40 K activity concentration from 0.31±0.03 Bq/L to 0.68±0.04 Bq/L with average 0.467±0.034 Bq/L. Minimum Detected Concentration (MDC) shows that activity concentration is below the detected limit of the equipment. The highest activity concentration is in the MJ03 sample code 238 U 1.29±0.72 Bq/L whereas the value surpassed the UNSCEAR recommended limit. The limit corresponding to an activity concentration of 238 U is 10 Bq.L⁻¹ (8).

²³⁸U is the primary contaminant in Gold mining activity. The bigger the Gold mining activity in the area, the higher the ²³⁸U activity concentration (9). ²²⁸U activity concentration can be increased because of a natural process, such as absorption from the soil, or technological processes involving radioactive material establishment naturally, such as phosphate fertilizer production or mineral and sand processing (13). Otherwise, uranium activity concentration can be high because leaching effect. It can cause factors that influence uranium activity concentration in the sample (14). Uranium comes from rock, sediment, or soil source and elements related to uranium decay, water distance towards uranium source, water hydraulic isolation degree towards the surface or underground water, and oxidated water condition. Mediator concentration can increase uranium solubility (15).

The highest ²³²Th activity concentration is in the underground water sample with code TB02, which is 0.04±0.03 Bq/L, while the highest ⁴⁰K activity concentration is 0.68±0.04 Bq/L in drinking water sample code MJ03. The area sampling of these samples is Tebo Regency and Muaro Jambi Regency. Those sampling areas are Tebo Regency and Muaro Regency, which have high plantation activity.

Plantations usually use a large amount of fertilizer that, most of which consists of Nitrogen (N), Phosphor (P), Potassium (K), and ⁴⁰K that have natural potassium overflow. The fertilizer also contains some natural radioactive (NORM) (16). Some of the fertilizer material can be

cleaned (separately) to the river so it will cause sedimentation towards sediment alongside the river. Radioactive material accumulation can occur in the riverbank sediment, and soil can be erased and sedimented from one place to another alongside the river. It can increase natural radioactivity in a specific area (17). The lowest activity concentration in the sample is ²³²Th. Generally, Thorium characteristic is complex to solute in water (18), so the value is low.

Based on the type of water, the highest radionuclide activity concentration in the river water sample is 40 K, 0.61±0.04 Bq/L in the TB01 sample code (Figure 2). It occurred because the sampling was done at Tebo Regency, with high plantation activity. The highest activity concentration in the underground water is shown in Figure 3 in TB02 sample code, which radionuclide is ²³⁸U that 0.69±0.01 Bq/L. Not only plantation activity but illegal gold mining activity is still operated there. The higher the Gold mining activity in an area, the higher the ²³⁸U activity concentration will be (19). Figure 4 shows that the highest drinking water activity concentration is in the MJ03 sample. However, the average ²³²Th and ⁴⁰K activity concentration in the water sample is still under the recommended limit allowed by PERKA BAPETEN, WHO, and UNSCEAR.





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Figure 4. Histogram of ²³⁸U, ²³²Th, and ⁴⁰K activity concentration in drinking water samples along the Batang Hari River, Jambi.

No.	Sample Code	R _{ea} (Bq/ L)	AEDE (mSv/y)	H _{ex}	H _{in}
			Jambi City		
1.	KJ01	0.05324	3.091x10 ⁻⁷	0.00012	0.000144
2.	KJ02	-0.08437	-3.96 x10 ⁻⁷	-0.00014	-0.00023
3.	KJ03	0.05940	3.493 x10 ⁻⁷	0.00014	0.00016
			Muaro Jambi		
4.	MJ01	0.51864	2.959 x10 ⁻⁶	0.00138	0.002618
5.	MJ02	0.51589	2.961 x10 ⁻⁶	0.00138	0.002610
6.	MJ03	1.37096	7.799 x10 ⁻⁶	0.00368	0.007192
			Batanghari		
7.	BH01	0.07832	4.535 x10 ⁻⁷	0.00018	0.000211
8.	BH02	0.02772	1.81 x10 ⁻⁷	7.5x10 ⁻⁵	7.48 x10 ⁻⁵
9.	BH03	-0.03333	-1.4 x10 ⁻⁷	-4.4 x10 ⁻⁵	-9 x10 ⁻⁵
			Tebo		
10.	TB01	0.08127	4.941 x10 ⁻⁷	0.00021	0.000274
11.	TB02	0.78339	4.442 x10 ⁻⁶	0.00207	0.003982
12.	TB03	1.16711	6.641 x10 ⁻⁶	0.00313	0.0061
			Bungo		
13.	BG01	0.0594	3.493 x10 ⁻⁷	0.00014	0.00016
14.	BG02	-0.02871	-1.1E x10 ⁻⁷	-3.1 x10 ⁻⁵	-7.8 x10 ⁻⁵
15.	BG03	0.04587	2.802 x10 ⁻⁷	0.00011	0.000124
	Average	0.307653	1.771 x10 ⁻⁶	0.00083	0.00155
	Minimum	-0.08437	-3.96 x10 ⁻⁷	-0.00014	-0.00023
	Maximum	1.37096	7.799 x10 ⁻⁶	0.00368	0.007192
	Threshold	370	1	1	1
		(20)	(20)	(11)	(11)

Table 2. Radiological Ha	azard Parameter
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The radiological hazards parameter in the water sample is shown in Table 2. The average value of Ra_{eq} in Bq/L unit is 0.307653, that still below the recommended limit of 370 Bq.L⁻¹ (20). The value of *AEDE* in the water sample shows the external radiation exposure reached up to 7.799 x10⁻⁶ mSv/y. The average value of *AEDE* is 1.771 x10⁻⁶ mSv/y is still under the recommended limit of 1 mSv/y.

The average value of the external hazard index (H_{ex}) in the water sample is 0.00083, while the internal hazard index (H_{in}) is 0.00155. These hazard indexes are still below the recommended limit, so the water is safe if reviewed based on radiation safety. The limit of H_{ex} and H_{in} allowed is 1 (21).

Therefore, the river, underground, and drinking water from along the Batang Hari River are still safely limited because the activity concentration of radionuclides is still under the threshold. Hence, the water sample's radiological hazard parameters, such as Ra_{eq}, AEDE, and external and internal hazard radiation, are low.

CONCLUSION

From this study, we can conclude that all ²³⁸U, ²³²Th, and ⁴⁰K activity concentrations in the water sample along Batang Hari River were detected using gamma spectrometry. All activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K are still under the recommended limit by PERKA BAPETEN No. 9 2009, WHO *Guidelines for Water Quality* 2011, and UNSCEAR *Report* 2000. Radiological hazard parameters include *Raeq, AEDE,* and external and internal hazard index. The average value of the radiological hazard parameter is still under the recommended limit. Therefore, the water is safe and fulfils the radiology standard water quality requirement.

ACKNOWLEDGEMENTS

The author would like to thank Wahyudi from the Central of Safety and Radiation Metrology Safety of Research and Innovation National Agency (PTKMR-BRIN), for helping with the research. Acknowledgment also goes to Maulida from the Directory of Laboratory Management, Facility of Research and Technology Area of Research and Innovation National Agency (DPLFRKST–BRIN).

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