Gamma radiation measurements and dose rate in waste soil samples of OK-Tedi Gold – Copper mine in Western Province of Papua New Guinea

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Abstract

The specific activity of natural radionuclides in the twelve different mining waste soil samples from OK Tedi gold -copper mine in Western Province of Papua New Guinea were analyzed to study the radiation effects in that region. The mean specific activities of ^{238}U, ^{232}Th, $^{40}K$and ^{235}U in the samples were found to range from 53.1 to 85.0 Bq/kg with an average value of 68.7± 5.0 Bq/kg, 41.6 to 130.0 Bq/kg with an average value of 90.5±31.4 Bq/kg, 0.6 to 1.3 Bq/kg with an average value of 0.8 ± 0.2 Bq/kg and 55.4 to 253.7 Bq/kg with an average value of 174.7± 55.9 Bq/kg respectively. The concentrations of ^{238}U and ^{232}Th were found to be 5.6 and 22.2 ppm respectively which are greater than the world average values of 2.64 and 11.1 ppm. The Radium equivalent activity was calculated and ranges from 144.5 to 248.6 Bq/kg with average activity of 197.2 ± 39.2 Bq/kg. The absorbed dose rate was calculated for the samples and ranges from 64.40 Bq/kg to 109.90 Bq/kg with an average value of 86.4 ± 16.9 Bq/kg. The average annual indoor dose of 605.3 ± 118.3 μGy/yr and outdoor annual effective dose of 151.3 ± 29.6 μGy/yr was estimated. The external hazard indices were calculated and observed to vary from 0.4 to 0.7 with average value equal to 0.5 which is lower than the permissible value of 1.0. The internal hazard indices were found ranging from 0.6 to 0.7 with average value equal to 0.7 which is lower than permissible value of 1.0. The excess lifetime cancer risk (ELCR) was calculated using risk factors of International Commission on Radiological Protection and Biological Effects of Ionizing Radiation and was found to range from $1.6x10^{-4}$ to $2.7x10^{-4}$ with an average value of $2.1x10^{-4}$ which is greater than the world average value of $1.45x10^{-4}$. Though the ELCR value is slightly greater, the radiation level in that region is less than the safe permissible level.

Keywords: *Natural radionuclides, Dose rate, Radium equivalent, Excess lifetime cancer risk.*

1. Introduction

Natural radiation contributes some percentage of total radiation exposure to humans in varying amounts geographically, and is a great concern to humanity because they contaminate the environment. Papua New Guinea is an Oceania country north of the Australian continent and is situated in the ring of fire thus bring about its challenges of induced environmental radiation hazard. The country is rich in renewable and non - renewable energy sources. In the non - renewable sector it has a large deposit of carbons and

minerals. The excavation of minerals such as gold, copper, nickel and gas and other minerals begin in 1852 during a gold rush period when the discovery of an abundance of gold deposit in some areas of the country (www.sl.nsw.gov.au). The large-scale mining came into operation in the late 1900 and so the OK-Tedi gold -copper mine begin its operation in 1981 and started producing in 1984(www.mrdc.com.pg/ok-tedi-no2-ltd.html, www.panda.org/discover/knowledge_hub/). In the process of mining, the large volume of earth crust was excavated to the surface including the low level of radionuclides mainly Uranium, Thorium, Actinium and Potassium and their progenies are the naturally occurring radioactive elements in the world. Specifically, Potassium- 40, Uranium-238 and Thorium-232 are major source of natural radioactivity. (Md. Samiul Ehsan et al., 2019). The naturally occurring crustal elements when undergoing decay process may release radiation energy which contributed to elevate the level of natural background radiation dose above accepted global average threshold. The elevation of the background radiation dose when exposed to by the population either inhaling, ingestion or whole body exposure could result in different forms of carcinomas (UNSCEAR, 2000). Since the mine is located in the upper stream of mighty Fly River whose water way is used for transport of equipment, cargo, people and materials to operate and sustain the mine operation can become a major source of ionization radiation transport, hence the basic knowledge of radiological parameters and radioactive contents redistribution of the earth crust and process mine waste is important. The necessity for the safety and upkeep of natural environment is paramount hence measurements and analysis of waste ore samples is mandatory. There were many studies associated with radiation exposure of natural radioactive elements in the world (Karalhan and Bayulken, 2000: Matiullah, et al., 2004; Tsortsiz and Tsetos, 2004; Veiga et al., 2006; Kessaratikoon and Aweakechi, 2008; Dimovska et al, 2011).

Papua New Guinea is situated in the 'Ring of Fire' region where earthquakes and volcanic eruptions are common. During mining process, earth materials from the interior of the earth are brought to the surface and the waste material are dumped outside. There is high chance of finding radioactive elements in the soil. In this study, the natural radioactivity concentrations of ^{238}U, ^{232}Th, ^{235}U and ^{40}K are analyzed in processed soil samples collected from waste ore from the mining site. The results are used to calculate the isotope contents in the soil samples. The gamma dose rate in air, the annual effective dose rate, radium equivalent and the internal and external hazard index were calculated to assess the radiation hazard associated with absorbed gamma dose rate.

2. Materials and Methods

Fig. 1 shows the location of OK Tedi Gold mine in Papua New Guinea. The mining site is 05^{o} 12' 28" South Latitude and 141^{o} 8' 20" East Longitude. People stay around the mining site where the waste is dumped. Twelve samples were collected from different locations around the mining waste and analyzed. The estimated population in this study area is estimated to be 20,000 (2011 Census). The area is under the influence of tropical climate with two seasons, rainy and sunny, interchangeably. There are no such studies carried out in this region on natural radioactivity till date.

Twelve samples were collected from different locations of the region where mining waste is dumped near Ok Tedi and were labelled A, B, C, D, E, F, G, H, I, J, K and L. The samples were filtered with a mesh of 1 mm X 0.5 mm mesh. The processed samples are then filled into 20 ml containers similar to Marinelli beaker and kept for a period of 30 days for the radionuclides progeny to reach equilibrium. The soil samples were then analyzed in a LB 2045 NaI (Tl) Gamma ray Spectrometer.

Fig. 1 Location of OK Tedi Gold mine in Papua New Guinea

2.1 *Experimental set up*

Preliminary Adjustments: A Cs^{137} test source is used for calibration. The calibration function automatically modifies the high voltage until Cs 137 photopeak lies on 662 keV energy line. The high voltage found is saved. The background function is used to measure the background spectrum in the range 0 – 2048 keV. All the sources in the vicinity of the detector are removed and the background spectrum is measured for half an hour. To get the correct activity of the sample, the background spectrum is subtracted from the total measurement result. This is done by the machine automatically. The period of counting of the sample was set at 12 hours. Neither ^{238}U nor ^{232}Th emits gamma rays. In our study, the activity of ^{238}U is estimated from the activity of ^{210}Pb , the activity of ^{232}Th is estimated from ^{228}Th and activity of Actinium series is estimated from ^{235}U. The specific activity of each sample is calculated using the formula (Ayse, 2017).

$$A = \frac{P}{\varepsilon f_\gamma t m} \qquad \text{(eq. 1)}$$

where P is the total net counts under the above-mentioned photo peak, ε is the measured photo peak efficiency, f_γ is the gamma ray intensity, t is the sample measurement time and m is the sample weight.

3. Results and Discussion

3.1 Specific activities of the natural radioactive elements

The specific activity of ^{238}U varies from 53.1 to 85.0 Bq/kg with an average value of 68.7$\pm$ 5.0 Bq/kg, the specific activity of ^{232}Th ranges from 41.6 to 130.0 Bq/kg with an average value of 90.5$\pm$31.4 Bq/kg, the specific activity of ^{235}U ranges from 55.4 to 253.7 Bq/kg with

an average value of 174.7± 55.9 Bq/kg and the specific activity of ^{40}K varies from 0.6 to 1.3 Bq/kg with an average value of 0.8 ± 0.2 Bq/kg. The specific activities of the natural radioactive elements are given in Table 1. The graphical representations of these results are shown in Figure 2.

Table 1. Specific activities of ^{238}U, ^{232}Th, ^{235}U and ^{40}K

Samples	Specific activity (Bq/Kg)			
	^{238}U	^{232}Th	^{235}U	^{40}K
1	58.4	73.3	236.7	1.2
2	71.7	57.5	244.4	0.9
3	85.0	41.6	252.0	0.6
4	80.8	54.1	153.7	0.6
5	76.6	66.6	55.4	0.6
6	64.9	98.3	120.3	0.6
7	53.1	130.0	185.1	0.7
8	65.7	125.0	153.9	1.0
9	78.3	119.0	122.6	1.3
10	69.0	85.8	218.6	0.6
11	55.7	101.7	210.9	0.9
12	65.6	125.0	153.9	1.0
Average	68.7±5.0	90.5±31.4	174.7±55.9	0.8±0.2

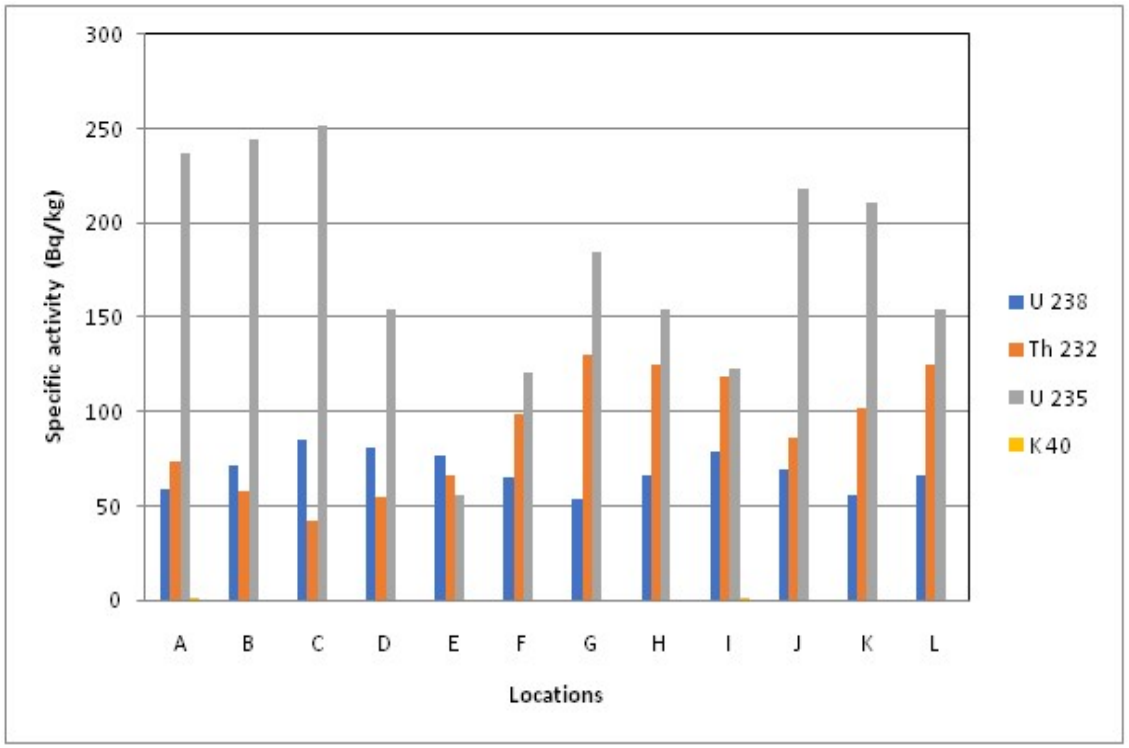

Fig. 2 Specific activities of ^{238}U, ^{232}Th, ^{235}U and ^{40}K for the waste ore samples collected from OK-Tedi Gold-Copper mine.

3.2 Estimation of radioactive elements in soil samples using Gamma ray spectrometry

The amount of radioactive isotopes in a given sample can be estimated as follows (Rasha et al., 2018). Let m (kg) be the mass of the isotope in the given sample of M (kg) with activity B (Bq/kg), mass number A, Avogadro number N_0, decay constant λ, half-life period $T_{1/2}$ and branching ratio Γ.

The number of radioactive isotopes in the sample, $n = \frac{mN_0}{A}$

Number of decays/sec, $n_d = \lambda n = \frac{\lambda m N_0}{A}$ where $\lambda = \frac{0.693}{T_{1/2}}$

Number of actual γ rays emitted/sec, $N_\gamma = n_d \Gamma = \frac{mN_0\lambda\Gamma}{A} = M$

From this, the mass of the radioactive isotope in the given sample can be estimated using the relation $m = \frac{AMB}{N_0\lambda\Gamma}$

Table 2. Average Specific activity and Isotope concentration of ^{238}U, ^{232}Th, ^{235}U and ^{40}K in the soil samples of OK-Tedi Gold-Copper mine.

Natural radioactive elements	Average Specific activity	Isotope Concentration (ppm)	World average Concentration (ppm)
^{238}U	68.7	5.6	2.64
^{232}Th	90.5	22.2	11.1
^{235}U	174.7	0.004	0.019
^{40}K	0.8	0.0026%	0.012%

Table 2 represents the specific activity and concentration of ^{238}U, ^{232}Th, ^{235}U and ^{40}K in ppm. The percentage of ^{235}U to ^{238}U is found to be 0.04% which is less than the naturally available ^{235}U to ^{238}U ratio of 0.012%. The main contribution of radiation is from ^{238}U and ^{232}Th. The activity of ^{238}U and ^{232}Th decay series radionuclides from all the waste ore samples collected from the mining sites were found to be 68.7 Bq/kg and 90.5 Bq/kg respectively which are within the range for ^{238}U and higher for ^{232}Th in comparison to world average range of 20 – 70 Bq/kg (IAEA 2004). The specific activity of 1 ppm of Uranium in rock sample is 12.35 Bq/kg and 1 ppm of Thorium is 4.06 Bq/kg (IAEA, 1989). In our study, concentrations of ^{238}U and ^{232}Th were found to be 5.6 and 22.2 ppm which are greater than the world average concentrations of 2.64 and 11.1 ppm. For K-40, 1% is equal to 313 Bq/kg in rocks. In our study, it is found to be 0.8 Bq/kg, equivalent to 0.0026%, which is lower than 0.012%, the world average value of ^{40}K in Natural Potassium (IAEA 1989).

3.3 Absorbed Dose rates in OK Tedi gold-copper mine waste ore samples

The approximate values of dose rates of OK-Tedi samples are calculated using the specific activity of the waste ore samples. The absorbed dose rate due to gamma radiations in air at 1m above the ground for uniform distribution of ^{238}U, ^{232}Th and ^{40}K were calculated using the guidelines of UNSCEAR 2000.

$$D(nGyh^{-1}) = 0.462A_U + 0.604A_{Th} + 0.04A_K \quad \text{(eq. 2)}$$

Where, A_U, A_{Th} and A_K are the specific activity of Uranium, Thorium and Potassium

$$\text{Outdoor Annual Estimated Dose (AED)} = D\ (nGyh^{-1})\ X\ 8760\ h\ X\ 0.2 \quad \text{(eq. 3)}$$

$$\text{Indoor Annual Estimated Dose (AED)} = \text{D (nGyh}^{-1}\text{) X 8760 h X 0.8} \quad \text{(eq. 4)}$$

Here, 0.2 and 0.8 are the outdoor and indoor occupancy index. Using equations (1), (2) and (3), the Dose rate, the outdoor and indoor estimated dose were calculated. The absorbed dose, the outdoor and indoor Average Estimated Dose of the waste ore samples are given in Table 3.

3.4 *Radium equivalent (Ra_{equ})*

$$\text{Ra}_{\text{eq}} = \text{A}_{\text{U}} + 1.43\text{A}_{\text{Th}} + 0.07\text{A}_{\text{K}} \quad \text{(eq. 5)}$$

Where, A_U, A_{Th} and A_K are specific activities of ^{238}U, ^{232}Th and ^{40}K (Bq/kg)

The calculated values of Ra_{eq} are tabulated in Table 3. The average value is observed to be 197.2±39.2 Bq/kg. The recommended maximum value for Ra_{eq} is 370 Bq/kg. So the radiation level is observed to be much lower than the average global vale.

3.5 Radiation Hazard Indices

Radiation Hazard Indices were calculated using the following equations.

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{(eq. 6)}$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{(eq. 7)}$$

The values of H_{ex} and H_{in} for the waste ore samples were given in Table 3. H_{ex} varies from 0.4 to 0.7 with an average value of 0.5±0.1 and H_{in} varies from 0.6 to 0.9 with an average value of 0.7±0.1. The values of internal and external radiation hazard indices must be less than unity for radiation hazard to be negligible.

Table 3. Absorbed dose, outdoor and indoor AED, Radium Equivalent, Radium Hazard Index and Excess Lifetime Cancer Risks for OK-Tedi Gold -Copper mine.

Samples	Specific Activity (Bq/kg)				Absorbed Dose(nGyh^{-1})	Outdoor AED (μGyh^{-1})	Indoor AED (μGyh^{-1})	$Ra_{eq.}$ (Bq/kg)	H_{in}	H_{ex}	ELCR
	^{238}U	^{232}Th	^{235}U	^{40}K							
1	58.4	73.3	236.7	1.2	71.3	124.9	499.7	163.3	0.6	0.4	0.0017
2	71.7	57.5	244.4	0.9	67.9	119.0	475.8	154.0	0.6	0.4	0.0017
3	85.0	41.6	252.0	0.6	64.4	112.8	451.3	144.5	0.6	0.4	0.0016
4	80.8	54.1	153.7	0.6	70.0	122.6	490.6	158.2	0.6	0.4	0.0017
5	76.6	66.6	55.4	0.6	75.6	132.5	529.8	171.9	0.7	0.5	0.0019
6	64.9	98.3	120.3	0.6	89.4	156.6	626.5	205.5	0.7	0.6	0.0022
7	53.1	130.0	185.1	0.7	103.1	180.6	722.5	239.0	0.8	0.6	0.0025
8	65.7	125.0	153.9	1.0	109.9	192.5	770.2	244.5	0.8	0.7	0.0027
9	78.3	119.0	122.6	1.3	108.1	189.4	757.6	248.6	0.9	0.7	0.0027
10	69.0	85.8	218.6	0.6	83.7	146.6	586.6	191.7	0.7	0.5	0.0021
11	55.7	101.7	210.9	0.9	87.2	152.8	611.1	201.2	0.7	0.5	0.0021
12	65.6	125.0	153.9	1.0	105.8	185.4	741.4	244.4	0.8	0.7	0.0026
Average	68.7 ±5.0	90.5± 31.4	174.7±5 5.9	0.8± 0.2	86.4 ± 16.9	151.3 ± 29.6	605.3 ± 118.3	197.2 ± 39.2	0.7 ± 0.1	0.5 ± 0.1	0.0021 ± 0.0004

3.6 *Excess Lifetime Cancer Risk (ELCR)*

If the average life expectancy of a human being is 70 years, then Excess Lifetime Cancer level is the probability of developing cancer over a lifetime at a given radiation exposure level. It is calculated as

ELCR = AED X DL X RF (eq. 8)

Where, RF is the risk factor whose value is taken as 0.05, DL is the duration of life and AED is the Annual Effective Dose.

The calculated values of ELCR are given in Table 3. The average value of ELCR is observed to be 0.0021 which is higher than the global average value of 0.00145.

4. Conclusion and recommendations

An LB 2045 Gamma Ray Spectrometer was used to determine the specific activity of ^{238}U, ^{232}Th, ^{235}U and ^{40}K from 12 soil samples collected from the waste ore of the OK Tedi Gold-copper mine in Western Province of Papua New Guinea. The respective Specific activities were then used to estimate the absorbed dose, annual indoor and outdoor effective dose, Radium Equivalent Dose, Radiation Hazard Indices and Excess Lifetime Cancer Risk. The mean concentrations of 5.6 ppm and 22.2 ppm for U238 and U232 respectively are higher than the global average values of 2.46 and 11.1 ppm respectively. The mean concentrations of 0.004 ppm and 0.007% for U235 and K40 respectively are lower than the world average values of 0.019 ppm and 1.3% respectively in the UNSCEAR 2000 Report. The average annual indoor dose of 605.3 ± 118.3 $\mu Gyh^{-1}$is lower than the maximum threshold of 1000 $\mu Gyh^{-1}$and outdoor annual effective dose of 151.3 ± 29.6 μGyh^{-1} is less than the global threshold. The average radium equivalent of 197.2 ± 39.2 Bq/kg is lower than the global average of 370 Bq/kg. The average Radiation Hazard Indices for indoor is 0.7 which is lower than the threshold value of 1.0 mSv and for outdoor it is 0.5 which is lower than the threshold of 1.0 mSv. The estimated average Excess Lifetime Cancer Risk value is 0.0021± 0.0004 which is higher than the global value of 0.00145. The ranges for the estimated parameters were also calculated. The radiation level around the Ok Tedi mining region is found to be less than the accepted global level of radiation.

5. References

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