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Radionuclides (^{40}K , ^{232}Th and ^{238}U) and Heavy Metals (Cr, Ni, Cu, Zn, As and Pb) Distribution Assessment at Renggam Landfill, Simpang Renggam, Johor, Malaysia

E Zaidi^{1*2}, MJ FahrulRazi², ATS Azhar³, ZAM Hazreek³, A Shakila², MS Norshuhaila⁴, M Omeje⁵

¹Research Centre for Soft Soil, Universiti Tun Hussein Onn Malaysia, Johor, MALAYSIA

²Faculty of Science, Technology and Human Development, Universiti Tun Hussein Onn Malaysia, Johor, Malaysia

³Faculty of Civil and Environmental Engineering, Universiti Tun Hussein Onn Malaysia, Johor, Malaysia

⁴Faculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia, Johor, Malaysia

⁵College of Science and Technology, Department of Physics, Covenant University, Ota, Ogun State, Nigeria

Corresponding author: zaidi@uthm.edu.my*, saifulaz@uthm.edu.my

Abstract. The assessment of radioactivity levels and the distribution of heavy metals in soil samples at CEP Farm landfill, Renggam in Johor State was to determine the activity concentrations of naturally occurring radionuclides and heavy metal concentrations of this landfill. The background radiation was monitored to estimate the exposure level. The activity concentrations of radionuclides in soil samples were determined using HPGe gamma ray spectroscopy whereas the heavy metal concentration was measured using X-RF analysis. The mean exposure rate at the landfill site was $36.2 \pm 2.4 \mu\text{R hr}^{-1}$ and the annual effective dose rate at the landfill site was $3.19 \pm 0.22 \text{ mSv yr}^{-1}$. However, residential area has lower mean exposure dose rate of about $16.33 \pm 0.72 \mu\text{R hr}^{-1}$ and has an annual effective dose rate of $1.43 \pm 0.06 \text{ mSv yr}^{-1}$ compared to landfill sites. The mean activity concentration of ^{40}K , ^{238}U and ^{232}Th at landfill site were $239.95 \pm 15.89 \text{ Bq kg}^{-1}$, $20.90 \pm 2.49 \text{ Bq kg}^{-1}$ and $40.61 \pm 4.59 \text{ Bq kg}^{-1}$, respectively. For heavy metal compositions, Cr, Ni and Cu have mean concentration of $232 \pm 10 \text{ ppm}$, $23 \pm 2 \text{ ppm}$, and $46 \pm 19 \text{ ppm}$, respectively. Whereas, Zn has concentration of $64 \pm 9 \text{ ppm}$ and concentration of $12 \pm 1 \text{ ppm}$ and $71 \pm 2 \text{ ppm}$ was estimated for As and Pb respectively. The higher activity concentration of ^{40}K down the slope through leaching process whereas the higher activity level of ^{238}U content at the landfill site may be attributed to the soil disruption to local equilibrium.

1. Introduction

Human kind are exposed to background radiations unknowingly and this exposure to natural occurring radiations are not preventable [1]. About 80 % exposures to the radiation of the world collectively originated from natural sources and 99% of world population exposed to radiation dose occurs as a result of natural sources contribution. In developing countries such as Malaysia, the increase in waste



generation cannot be overemphasized which can be attributed to some factors as increasing population level, rapid urbanization, rise in civilian living standards and glooming economy. The waste emanating from such areas, residential, industrial residues and other domestic places dumped into the landfill. In areas such as landfill site usually has a high contaminants of heavy metals due to gathering of different waste materials. Moreover, heavy metals elements usually exist as anion in soils which may lead to the condition of the soil to become acidic due to the positive charge (ion exchange) that develop in soil [2].

In this case, the source of the contamination can be categorized into elements in heavy metal and radionuclides. The heavy metal elements such as Cr, Ni, Cu, Zn, As and Pb tend to become toxic when found in higher concentration in a soluble form. On the other hand, radioactive elements such as ^{238}U , ^{232}Th , and ^{40}K may be found in waste disposal with various activities and the radionuclide concentration. Radionuclide were carried some distance away by wind or water, or from the waste itself before coming to rest in soil [3]. Radionuclide in environment is normally emitted gamma ray and also known as terrestrial radiation and its concentration depending on the geographical condition and geological formation [1, 4, 5].

Other contributions mainly occurs as a result of anthropogenic activities such as dumping of waste to inappropriate area and quarry site activities. The exposure of human kind to this natural radioactivity is mostly depends on the impact of these activities on types of the soil and geological formation of the area. Previously, study related to contaminated soil with its parameter relationship and treatment has widely being studied [6-11].

CEP Farm landfill Renggam (N 01°55.406' E 103°22.684') is a municipal landfill site which receive waste disposal from the district of Batu Pahat, Simpang Renggam and Kluang. The accumulation of huge amount of waste could contribute to high level of radionuclide concentrations and heavy metals in the study area. This study aims at assessing the background radiation level of the radionuclides and the concentrations of heavy metals which significantly will play a vital role in monitoring the contamination level in future since no report of this kind yet in the landfill site.

2. Materials and Methods

2.1 Sample collection and preparation

Soil samples were collected at four places of interest which surrounded by Rengam-Jerangau soil species with FAO classification of Haplic Nitisols and Geric Ferrasols respectively. This particular soil has sandy and clay loam structure and very common soil series in the oil palm plantations area. In this study the sampling point at this landfill are assigned at the leachate area, the peak of waste disposal, entrance and the roadside area. Point 1, 2, 3 were located at the leachate area. Point 4, 5, and 6 were located at the peak of waste disposal. Point 7 and 8 were located at the entrance, while Point 9 and 10 were located at the roadside. Soil samples were taken at a depth of 10 cm and 2 kg for each sample and kept in a plastic bag. The soil samples were then brought to the Research Centre of Soft Soil (RECESS) laboratory, University Tun Hussien Onn Malaysia (UTHM) for sample preparation purposes. The soil samples were put inside a tray and dried in an oven at 106°C for 24 hours. The dried soils were then grinded by using a grinding machine in order to get the fine granule powder. After finished the grinding process, the soil powder was then sieved at 75µm aperture sieve. The sample powder were loaded in Marinelli beakers and sealed for 4 weeks in order to achieve secular equilibrium. The remaining sieved soils were undergone pelletizing process. A mass ratio in gram (8:2) of soil and wax were weighed by using an analytical balance. A mixture of soil and wax were loaded into a mould and compressed at 15 tonnes by using hand-hydraulic compressor into a pallet form. The heavy metal and radionuclide analysis was performed using X-ray Fluorescence (XRF) and Hyper Pure Germanium detector (Gamma Spectroscopy) respectively.

2.2 Radiation dose measurement

The measurement of gamma radiation exposure was conducted by using a gamma ray survey meter. The background dose at this landfill site was measured five times for each sampling point in order to achieve a good statistical analysis. The radiation dose was measured in $\mu\text{R hr}^{-1}$ and the annual dose was measured in mSv yr^{-1} . The average of exposure dose, the standard deviation and the standard error for every measurement of each sampling point were calculated using the following equations [12];

$$\text{Average, } \bar{x} = \frac{\sum_{i=1}^n x_i}{n} \quad 2.1$$

$$\text{Standard deviation, } SD = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n}} \quad 2.2$$

$$\text{Standard error, } SE = \frac{SD}{\sqrt{n}} \quad 2.3$$

Where \bar{x} an average and n is a number of samples. The annual dose rates were simply converted from exposure dose by using this relationship [2];

$$\text{Dose Rate (mSv yr}^{-1}\text{)} = 0.0833 \times \text{Exposure Rate (}\mu\text{R h}^{-1}\text{)} \quad 2.4$$

2.3 Activity concentration of the radionuclide

Measurements of the activity concentration of ^{40}K , ^{238}U and ^{232}Th in Bq kg^{-1} dry weight of the collected samples were carried out by using HPGe gamma ray spectrometry with 20% relative efficiency. Each sample in the marinelli beaker was place in a lead-shielded in order to reduce gamma ray background from surrounding. The gamma transitions used for ^{238}U were 352.46 keV and 609.6 keV. Those for ^{232}Th and ^{40}K gamma transition were 583.53 keV and 1460.28 keV, respectively. All gamma transition energies are tabulated in Table 1 and the correlation between efficiency and the gamma energies is illustrated in Figure 1. The following equation was used to determine the activity concentration of radionuclides in soil samples [13];

$$A_s = A_{\text{net}} / (\epsilon \times I_{\gamma} \times m \times t) \quad 2.5$$

Where A_{net} is the number of counts of a full photopeak from a sample corrected from background, m is the mass of sample in kg, I_{γ} is the emission probability per decay of the specific peak, t is the sample measurement time in second, and ϵ is the absolute photopeak efficiency of the detector at a particular photopeak.

2.4 Elemental composition of the heavy metal

The elemental composition of the heavy metal in the soil sample was determined using an XRF spectroscopy system. The standard sample of SOIL-7 from IAEA, Vienna, Austria has been used as a reference sample. Here, S2 PICOFOX software was used to extrapolate the composition and distribution of heavy metals. The concentration of heavy metals was measured in parts per million (ppm). The heavy metal elements was identified by using X-ray florescence spectrometry (XRF). It

determines the 13 elemental compositions that are non-destructive and precise. The samples were analysed simultaneously which employed to determine the elemental compositions especially for tracing the heavy metal elements.

3. Results and Discussions

3.1 Exposure rate at the sampling site

From Figure 2, it can be observed that the exposure level is inhomogeneously distributed for each sampling points. This variation in distribution of exposure level is due to different sampling points from different locations between the landfill and roadside areas.

The exposure rate for Point 1, which at the leachate area has average rate of $42.00 \pm 0.57 \mu\text{R hr}^{-1}$. The exposure rate at this point was very higher compared to the rest of the points due to the accumulation of radionuclide in leachate zone and may be attributed to the combination of waste materials and other geological features. The integration of accumulation of radionuclide and geological features at this leachate zone may result to the higher background level of radiation exposure measured in the area which could serve as the trend of redeposition of radionuclides on interaction with the geological formation. Points 5 has an average exposure rate of $35.2 \pm 0.72 \mu\text{R hr}^{-1}$ and lower than Point 1. At point 4, 5 and 6 which are at the peak of waste contribute, an average exposure rate of $30.8 \pm 1.98 \mu\text{R hr}^{-1}$. The surface of the waste disposal peak has less absorption of radioactive substance, thus this indicates that less dilution process occur at this point. Point 7 and 8 which were located at the entrance point have an exposure rate values of $26.8 \pm 0.44 \mu\text{R hr}^{-1}$ and $23.3 \pm 0.72 \mu\text{R hr}^{-1}$, respectively, with the mean value of $25.0 \pm 1.27 \mu\text{R hr}^{-1}$. The lower average reading of exposure rates between the leachate area and peak of waste disposal was due to the different soil species which mainly covered by the sedentary soil which may not support the oxidative nature of uranium. Point 9 and 10 were located at the roadside which has an average reading of exposure rate of $36.6 \pm 2.68 \mu\text{R hr}^{-1}$. A higher reading of exposure rates occurred at the roadside as it is mainly covered by the bitumen or asphalt [14]. The average exposure rate at landfill site between Point 1 to Point 6 was $36.2 \pm 2.4 \mu\text{R hr}^{-1}$.

3.2 Annual dose rate at sampling site

The annual effective dose rates at the sampling sites were converted from the exposure rate by using Equation 2.4. The annual effective dose received from this survey at each sampling point is shown in Figure 3. At points 1, 2 and 3 which is the leachate area give an average annual effective dose of $3.67 \pm 0.02 \text{ mSv yr}^{-1}$ whereas point 4, 5 and 6 which are at the peak area of the site contribute an average annual dose rate of $2.70 \pm 0.17 \text{ mSv yr}^{-1}$. Point 7 and 8 which located at the entrance give an average annual effective dose rate of $2.18 \pm 0.11 \text{ mSv yr}^{-1}$ whereas point 9 and 10 which are at the roadside contributed average annual effective dose rate of $3.21 \pm 0.23 \text{ mSv yr}^{-1}$. Here, the landfill sampling point from point 1 to point 6 gives an average annual effective dose of $3.19 \pm 0.22 \text{ mSv yr}^{-1}$. From the data obtained, the average annual effective dose rate at the landfill site was slightly higher than the dose limit for the civilian which was 1 mSv yr^{-1} [1]. The increment of its annual dose could be contributed by several factors such as the accumulation of radionuclide, geological formation and also various polluted and toxic compound residue in this area.

3.3 Activity concentration of radionuclides

The activity and the activity concentrations naturally occurring radionuclides (^{40}K , ^{238}U and ^{232}Th) in each soil samples were determined by using HPGe gamma spectroscopy. The activity concentration was measured in Bq kg^{-1} .

3.3.1 Potassium (^{40}K)

The activity concentration of Potassium (^{40}K) at sampling points 1, 2 and 3 was measured with maximum average value of $254.34 \pm 26.80 \text{ Bq kg}^{-1}$ as tabulated in Table 2. The minimum average

value of the activity concentration was measured at the entrance to the site (Point 7 and 8) with the value of 170.51 ± 5.25 Bq kg⁻¹. Higher activity concentration of 40K at leachate area could be due to the accumulation of element that are mobile through the slope of the waste disposal peak edge. The concentration of 40K found to be higher as compared to 238U and 232Th because 40K was the most abundant elements for about 2.4% of the Earth's mass [15].

3.3.2 Uranium (238U)

The average activity concentration of 238U at the leachate area (Point 1, 2 and 3) was 24.82 ± 2.99 Bq kg⁻¹. Based on the results presented in Table 3, leachate area has the maximum average activity concentration of 238U as compared to other sampling point with activity level of 24.82 ± 2.99 Bq kg⁻¹. Such higher level of 238U at sampling point 1, 2 and 3 may be due to the occurrence of uranyl ion, [UO₂]²⁺ which derived from the interaction with the acid rain as such may enhance the redeposition uranium. This could lead to uranyl ion, [UO₂]²⁺ migration into the ground water system and the local equilibrium could be disrupted [15]. Activity concentration seems to be declined from Point 4 to Point 8 which covers from the peak of waste disposal to the entrance area. Entrance contribute the lowest average of activity concentration of 6.29 ± 0.94 Bq kg⁻¹. As overall, it is clearly observed that 238U activity reduces as the sampling point is far away from the dump site which obeys the inverse square law.

3.3.3 Thorium (232Th)

According to Table 4, the maximum average activity concentration of 232Th was located at leachate area (Point 1, 2 and 3) which has 48.16 ± 5.13 Bq kg⁻¹ as the activity level. The lowest average activity concentration was 11.01 ± 0.30 Bq kg⁻¹ which located at the entrance area reported at point 7. An anomaly measurement occurs at the roadside with the average activity concentration of 232Th 16.49 ± 0.63 Bq kg⁻¹ was noted. In natural environment, the activity concentration of 232Th is higher than that of the activity concentration of 238U because 232Th was much more abundant in nature [15 – 16].

3.4 Elemental compositions of heavy metals

In Table 5, it can be observed that chromium (Cr) has the highest concentration at each sampling point. The average concentration of Cr at the landfill site (leachate area and peak of waste disposal) was 232 ± 10 ppm. The contribution of Cr concentration was due to the association of natural soil with ultrabasic rocks such as sandy soils or maybe from the type of soils [17]. The average concentration of nickel (Ni) at the landfill site was 23 ± 2 ppm. Copper (Cu) has the average concentration of 46 ± 19 ppm due to the accumulation of Cu in waste disposal, anthropogenic activities and also the large number of corroded metals on dumpsite [18 – 19]. An anthropogenic activities led to the high level of zinc (Zn) concentration at the landfill site for about 64 ± 9 ppm which could be attributed to the acidification and decreasing soil adsorption ability [20,21]. Meanwhile, the concentration of arsenic (As) at the landfill site is 12 ± 1 ppm. A less amount of As concentration could be due to the surrounding environment that cover by an agriculture activities. Pesticides containing As could disperse to the air and being absorbed by the soil at the landfill site [3]. Meanwhile the concentration of lead (Pb) at the landfill site was 71 ± 2 ppm. The Pb accumulates at the landfill site due to the major source of Pb pollution by continuous disposal of lead containing waste into the dumpsite [3, 18]. It also notices that there is a significant trend of Pb distribution between leachate, entrance, waste peak and roadside area which may have enhanced the level at the peak location.

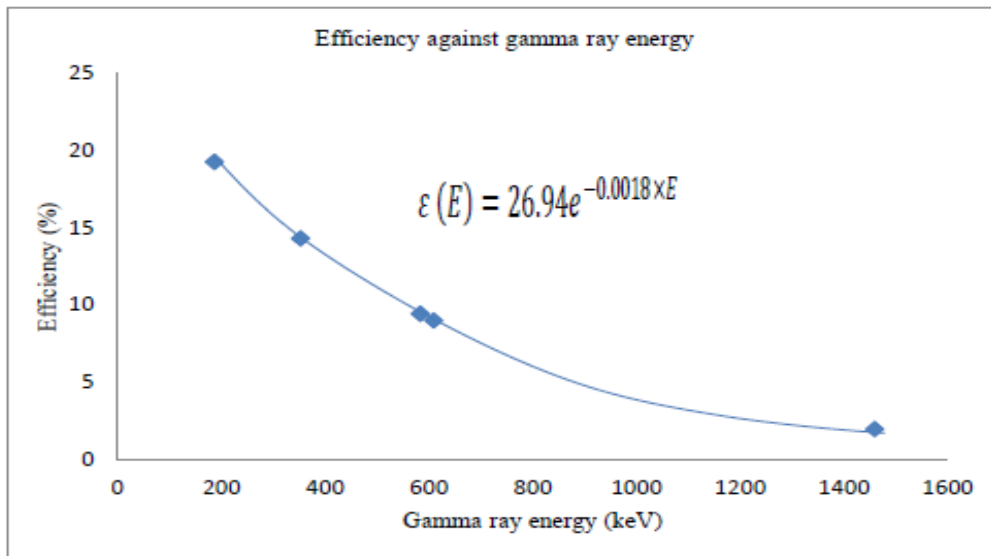


Figure 1: Efficiency as a function of gamma ray energies for HPGe detector.

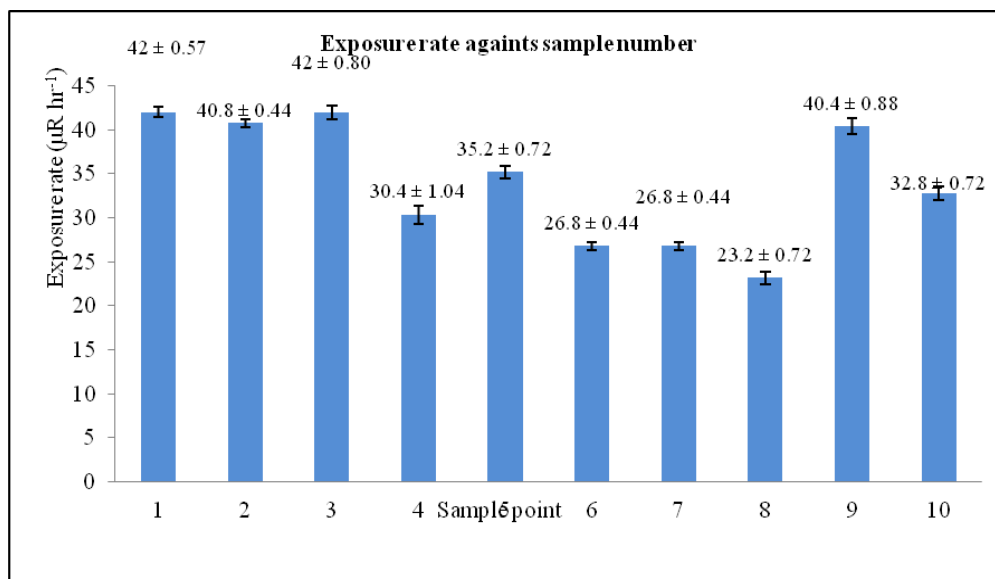


Figure 2: The distribution of exposure rate at sampling point location.

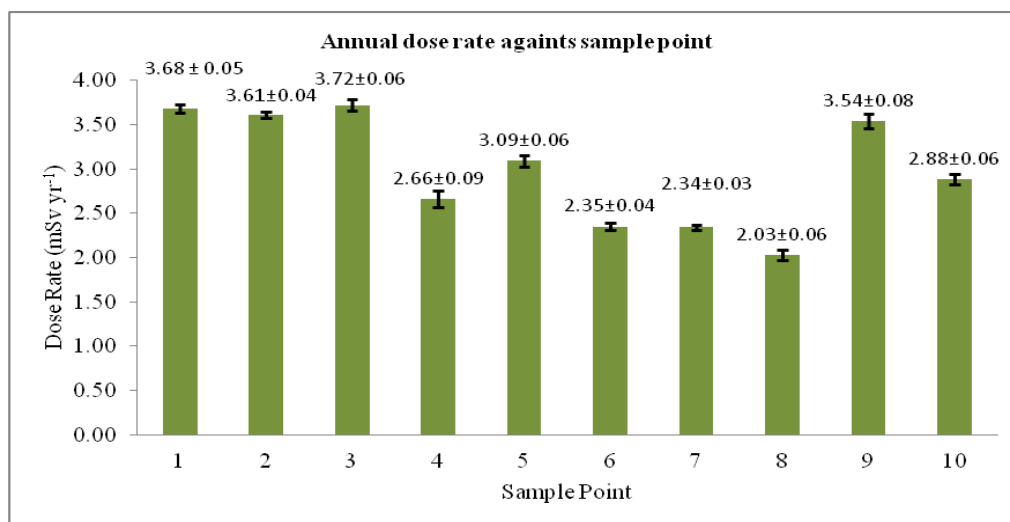


Figure 3: The distribution of annual dose rate at each sampling location.

Table 1: The measured energy peak efficiency as a function of gamma ray energies.

Radionuclide	Lr (%)	Energy (keV)	Efficiency (%)
Ra-226	3.52	186.7	19.25
U-238	35.34	352.4	14.29
Th-232	30.55	583.5	9.43
U-238	45.16	609.6	8.99
K-40	10.66	1460.2	1.95

Table 2: The activity concentration and radionuclide concentration of ⁴⁰K.

Location	Sample no.	Activity Concentration (Bq kg ⁻¹)			
		Activity	±	Mean	±
Leachate	1	303.57	8.32	254.34	26.80
	2	192.12	5.05		
	3	267.33	7.00		
Peak	4	249.73	7.21	225.57	12.41
	5	229.46	6.15		
	6	197.51	5.61		
Entrance	7	163.09	4.30	170.51	5.25
	8	177.92	4.92		
Roadside	9	213.68	5.90	190.46	16.42
	10	167.24	4.93		

Table 3: The activity concentration and radionuclide concentration of ^{238}U .

Location	Sample number	Activity Concentration (Bq kg ⁻¹)			
		Activity	±	Mean	±
Leachate	1	31.14	0.54	24.82	2.99
	2	18.47	0.32		
	3	24.84	0.43		
Peak	4	20.16	0.46	16.98	2.37
	5	19.59	0.36		
	6	11.18	0.27		
Entrance	7	7.63	0.19	6.29	0.94
	8	4.96	0.17		
Roadside	9	10.58	0.30	11.55	0.69
	10	12.52	0.34		

Table 4: The activity concentration and radionuclide concentration of ^{233}Th .

Location	Sample no.	Activity Concentration (Bq kg ⁻¹)			
		Activity	±	Mean	±
Leachate	1	56.54	0.96	48.16	5.13
	2	35.87	0.58		
	3	52.08	0.83		
Peak	4	29.07	0.66	33.06	4.46
	5	43.86	0.72		
	6	26.26	0.55		
Entrance	7	11.01	0.30	14.55	2.50
	8	18.09	0.42		
Roadside	9	17.38	0.45	16.49	0.63
	10	15.60	0.40		

Table 5: The heavy metal concentration by the place of interest.

Elements	Leachate		Entrance		Peak		Roadside	
	ppm	±	ppm	±	ppm	±	ppm	±
Cr	213	14	232	33	251	4	266	39
Ni	27	2	35	1	20	1	16	3
Cu	18	5	37	17	74	29	14	2
Zn	71	14	78	33	58	8	32	2
As	11	1	13	1	14	1	19	1
Pb	75	2	30	12	67	2	19	5

4. Conclusion

Mean exposure rate at the landfill site was $36.2 \pm 2.4 \mu\text{R hr}^{-1}$ and the annual effective dose rate at the landfill site was $3.19 \pm 0.22 \text{ mSv yr}^{-1}$. The higher exposure rate and annual effective dose rate may be due to the accumulation of radioactive substances inside the waste disposal area. However, the mean exposure rate at residential area was $16.33 \pm 0.72 \mu\text{R hr}^{-1}$ and has annual effective dose rate of $1.43 \pm 0.06 \text{ mSv yr}^{-1}$. Landfill site has larger mean exposure rate and annual dose rate compared to residential area which is attributed to the effect of the waste deposits. The mean activity concentration of ^{40}K , ^{238}U and ^{232}Th at landfill site were $239.95 \pm 15.89 \text{ Bq kg}^{-1}$, $20.90 \pm 2.49 \text{ Bq kg}^{-1}$ and $40.61 \pm 4.59 \text{ Bq kg}^{-1}$, respectively which was inferred to be the reaction of the waste materials, acid rain and other geological sources of the soil. The higher activity concentration of ^{40}K may due to the mobilisation of ^{40}K down the slope through leaching process. ^{238}U content at landfill site may be attributed to the waste disposal or soil disruption to local equilibrium. The mean concentration of heavy metal elements of Cr, Ni, Cu, Zn, As and Pb at landfill were $232 \pm 10 \text{ ppm}$, $23 \pm 2 \text{ ppm}$, $46 \pm 19 \text{ ppm}$, $64 \pm 9 \text{ ppm}$, $12 \pm 1 \text{ ppm}$ and $71 \pm 2 \text{ ppm}$, respectively. Different species of heavy element has different factors that affect the concentration in waste disposal. Some of the factors that influence could be consist of soil sample species, accumulation of elements, mobility and solubility of metals in soil, anthropogenic activities and the metal content residues.

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