

RADIOACTIVITY CONCENTRATIONS OF SOILS FROM THE VICINITY OF LYNAS PLANT AT GEBENG, KUANTAN AND ITS POTENTIAL RADIATION RISK

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ABSTRACT

Natural radioactivity in the surface soil from the vicinity of Lynas Rare-earth plant in Kuantan, Pahang had been studied with aim of evaluating environmental radioactivity of that area. The radioactivities of the samples have been measured using a low-background HPGe gamma detector. The concentration of ^{226}Ra ranged found to be between 8.7 – 76.9 Bq/kg (mean 33.7 Bq/kg); activity of ^{238}U from 8.7 to 80.5 Bq/kg (mean 36.9 Bq/kg); as for ^{232}Th , it ranged from 6.2 to 121.5 Bq/kg (mean 58.2 Bq/kg) and that of ^{40}K , the values was from 19.1 to 778.9 Bq/kg (mean of 221.3 Bq/kg). Activity standard deviation for ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K were 14.2 Bq/kg, 14.7 Bq/kg, 26.1 Bq/kg and 185.6 Bq/kg, respectively. To evaluate the risk of radiation hazard to human, several parameters, i.e. the radium equivalent activity, External hazard index, Representative level index and dose rate in air from the terrestrial natural gamma radiation were calculated with the mean values found to be 136.7 Bq/kg, 0.4, 1.0, 65 nGy/h, respectively. The data were discussed and compared with those obtained from the previous studies given in the literatures. This study found that the mean dose equivalent received by an individual at the vicinity area (outdoor) of Lynas plant was estimated to be 82.0 $\mu\text{Sv/yr}$, which is far below the annual dose limit of 1,000 $\mu\text{Sv/yr}$ for general public as stipulated in the national act.

Keywords: Dose equivalent, external hazard index, Lynas, natural radionuclides, radiation risk, surface soil

INTRODUCTION

Natural radioactivity contributes significantly to radiation doses received by mankind. Two prominent sources of external radiation are coming from cosmic rays and terrestrial gamma rays. Cosmic rays are mostly been shielded off by the ozone layer and contribute less radiation risk to human beings. Meanwhile, the primary source of terrestrial radiation received by humans is from the naturally occurring radioactive material (NORM) deposited in the earth crust, which derives essentially from ^{40}K , and the daughters of ^{238}U and ^{232}Th decay series such as radium, radon, actinium, protactinium, lead and polonium. These progenies first appear in the lithosphere level, deposited on the surface soil before it been washed and drained through several pathways such as weathering, erosion, fallout, rainwater and human activities into rivers transport and finally ended in the estuary (Ahmad-Taufek et al., 2004; Akram et al., 2004; Myrick et al., 1983; Nagaya and Saiki, 1967). The knowledge of the concentrations and distributions of radionuclides in the soils are of interest since it provides useful information for understanding changes in monitoring environmental contamination from natural radioactivity (Chiozzi et al., 2002; Sroor et al., 2001). Furthermore, data on natural radiation are important for the authorities to design rules and regulations for the purpose of radiation protection.

When the environment was contaminated by the enhancement of natural radioactivity from human activities (TENORM), human being present in such areas would be exposed to additional radiation from these natural radionuclides. Therefore, the radiation hazards risks to these people are of interests in order to ensure that no additional doses are imposed on to them. Among those radionuclides, radium-226 (^{226}Ra , uranium series progeny), uranium-238 (U-238), thorium-232 (Th-232) and potassium-40 (^{40}K) are at most concern due to their rich progenies that mostly emitting gamma energies. In most literatures, radiation risks were calculated from the specific activity of these few radionuclides (Ahmed and El-Arabi, 2005; Nabil et al., 2010; Narayana et al., 2001; Yang et al., 2005). The concentrations of ^{226}Ra , ^{238}U and ^{40}K on terrestrial especially in the areas of Peninsular Malaysia had been widely reported (Omar et al., 1990; UNSCEAR, 2000; Yahaya et al., 1997; Yasir et al., 2006) but none reported about the radiation risk.

Lynas Malaysia Sdn. Bhd. (Lynas), a rare earth refinery plant remains a thorny issue as the processing of lanthanide concentrate will also produce solid powdery waste which contains very low levels of NORM. The waste was reported to contain 6.3 Bq/g thorium and 0.38 Bq/g uranium and the lanthanide residue contains 6.6 Bq/g ^{228}Ra and 0.27 Bq/g ^{226}Ra radionuclides (The Academy of Sciences Malaysia, 2011). Operation of this plant has been protested by local residents and environmental pressure groups due to the potential chemical and radioactive waste hazards. The plant operations will produced pollutants such as chemicals, heavy metals and radioactive in forms of gases and liquids. These pollutants have potential to be released into the environment through the stacks, discharge drains and leakages. Therefore, it is crucial to establish a specific monitoring program to carry out the monitoring activities.

In this article, activity concentration of natural radionuclides for ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K in surface soils from the vicinity of Lynas plant at Gebeng, Kuantan, which were determined by HPGe gamma-ray spectrometry and the measured results were used to estimate the additional radiation doses originated from the presence of these radionuclides in the surrounding land. This enable a dosimetric evaluation for risk to people in that area of concern were estimated.

MATERIALS AND METHODS

Sampling and Sample Preparation

A total of 31 stations had been randomly selected and established within 3 km radius surrounding the plant, as illustrated in Fig. 1. A composite soil sample was collected where the top soil (< 5cm depth) were obtained randomly from a metre square area of each station. The surface soil samples were collected using a coring tool and covered a total surface area of at least 200 cm². Collected samples were placed into the HDPE plastic bag, properly labeled and sealed before transported back to the laboratory. Samplings process were repeated at the same monitoring station with an interval of approximately two-three months and the data used in this article is the mean value from each station, for the samples that were collected between May 2014 until November 2014.

The collected samples were weighted individually, about 1 kg each, and dried at 105°C for minimum duration of 24 hrs until a constant mass, then ground to pass through a 200-mesh sieve. Samples were transferred into a 600 ml marinelli beaker, sealed with thick PVC tape to inhibit radon gases from escaping. The sample weight between 400 – 1000 g with average sample weight about 800 g. All samples were stored for a period in excessive of 30 days (> 7 half-lives of ^{222}Rn and ^{220}Rn) to establish secular equilibrium (Dowdall and O'Dea, 2002; Yang et al., 2005) between parents and their respective radioactive progeny prior to gamma counting.

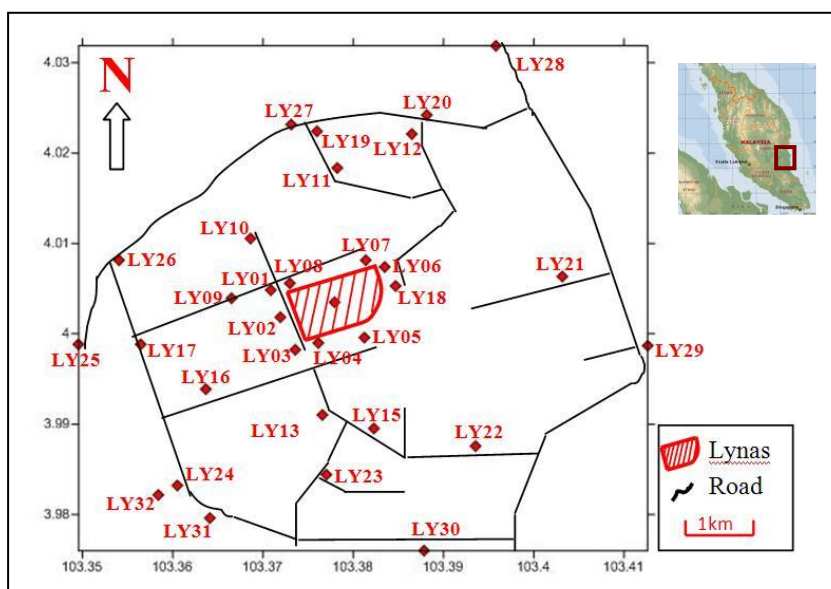


Figure 1: Sampling location

Experimental Setup

A sample spectrum was individually measured with the gamma ray spectrometry consisting of a high-purity germanium (HPGe) setup and multichannel analyzer of 16,384 channels. The detector used is a coaxial 3 inches diameter closed end, closed facing window geometry with vertical dipstick (500 – 800 μm) operated at 3,500 HV bias supply. The detector was shielded in a chamber made of lead, cadmium and copper (total thickness 11 cm) to reduce the background radioactivity. This p-type detector was designed to provide 25% relative efficiency with the FWHM resolution of 0.78 keV at 122 keV gamma-ray line of ^{57}Co and 1.80 keV at 1332 keV gamma-ray line of ^{60}Co . It was calibrated using procedures as reported earlier by Yii et al. (2009) using customized gamma multinuclides standard source comprising of ^{210}Pb , ^{241}Am , ^{109}Cd , ^{57}Co , $^{123\text{m}}\text{Te}$, ^{51}Cr , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{88}Y and ^{60}Co in the same counting geometry. Source used was manufactured by Isotope Products Laboratories, USA (source no. 1290-84 and 1755-30).

All samples were counted for 50,000 seconds using spectrometer and corrected for density and sampling date. A container of the same geometry filled with inert material counted was used to determine the background counts. Counting times were long enough to ensure a 2σ counting error of less than 10%. Previous studies reveal that minimum counting time of 10 hours (36,000 seconds) is sufficient to provide adequate counts under the various gamma-ray peaks (Ahmed and El-Arabi, 2005; Arogunjo et al., 2005; El-Reefy et al., 2006).

The ^{226}Ra , ^{238}U and ^{232}Th were measured through the gamma transitions of their progenies; ^{214}Pb (295.21 and 351.92 keV) and ^{214}Bi (609.31 keV, 1120.29 keV and 1764.49 keV) were used for determination of ^{226}Ra . U-238 activity was obtained through the emissions at the energy lines of 351.92 keV (^{214}Pb) and 609.31 keV, 768.36 keV, 1120.29 keV, 1238.11 keV and 1764.49 keV (all of ^{214}Bi). The energy lines of 238.63 keV (^{212}Pb), 338.4 keV, 911.07 keV, 969.11 keV (^{228}Ac) and 583.19 keV (^{208}Tl) were used to calculate the activity for ^{232}Th . Meanwhile, ^{40}K content was measured directly *via* its 1460.7 keV energy peak (El-Reefy et al., 2006; Mashra and Sadarivan, 1971; Yang et al., 2005).

Since the detection system provided a counting rate that was proportional to the amount of radioactivity in the samples, the radioactivity concentrations in the environmental samples were calculated using equation as reported by Yang et al. (2005) and Chen et al. (2005). The minimum detectable activity (MDA) for ^{226}Ra , ^{238}U and ^{232}Th was quantified to be 1 Bq/kg per dry weight (dry wt.), while ^{40}K was quantified at 5 Bq/kg after considering the size and the counting time of the sample.

RESULTS AND DISCUSSION

The activity concentrations for all radionuclides at all stations were found to be above the MDA value. The activity concentrations in some locations appear to be higher when compared to the other stations. The wide area of sampling could be the explanation for this observation. From this study, the concentration of ^{226}Ra was found to be ranged between 8.7 – 76.9 Bq/kg with a mean value of 33.7 Bq/kg; the activity of ^{238}U varied from 8.7 to 80.5 Bq/kg with a mean value of 36.9 Bq/kg; for ^{232}Th , it ranged from 6.2 to 121.5 Bq/kg with a mean value of 58.2 Bq/kg and that of ^{40}K was from 19.1 to 778.9 Bq/kg with a mean of 221.3 Bq/kg. The activity standard deviation of ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K was 14.2 Bq/kg, 14.7 Bq/kg, 26.1 Bq/kg and 185.6 Bq/kg, respectively.

Activity concentration for each radionuclide collected from the same location at different interval was averaged. The average activity concentration for ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K at each station is summarized in Fig. 2. The average activity for ^{226}Ra , ^{238}U and ^{232}Th have very similar pattern, i.e. they are quiet high at station LY 01, LY 02, LY 10, LY 13, LY 15, LY 23, LY 26 and LY 29 but low at station LY 05, LY 11, LY 12 and LY 32. Meanwhile, K-40 has a very different pattern as compared to the other three radionuclides with high average activity found at station LY 01, LY 02, LY 06, LY 07, LY 16, LY 19 and LY 26 but low at LY 04, LY 18, LY 27 and LY 30.

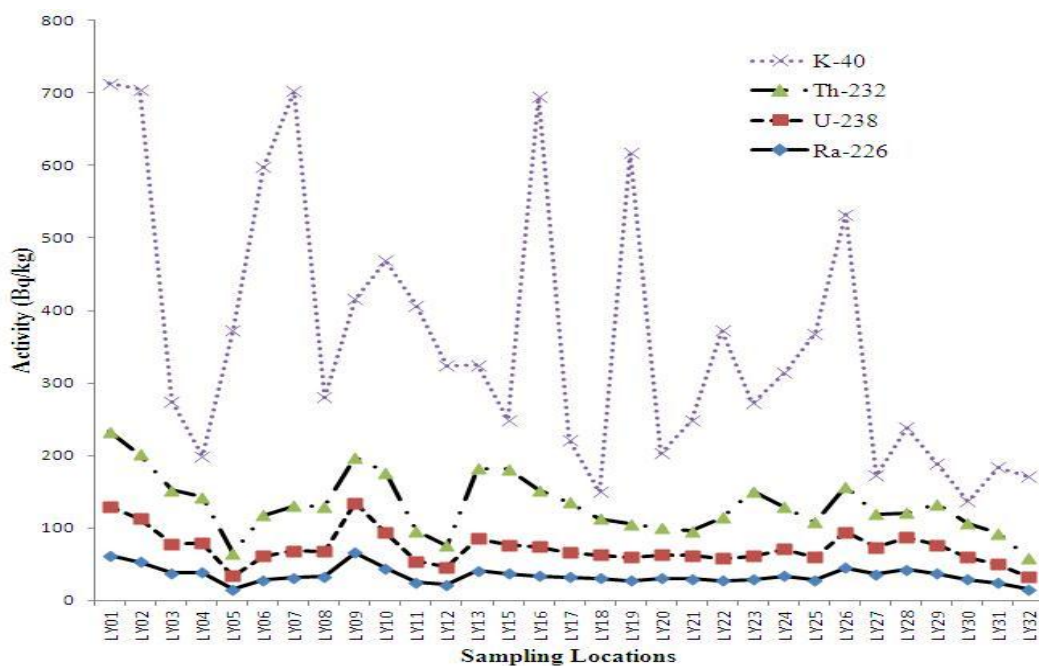


Figure 2: Average activity concentration ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K at sampling station

The high activity of ^{226}Ra and ^{238}U is found at station near point LY 01 and LY 09 (about 150 meter from Lynas) while low activity is observed near point LY 05, LY 12 and LY 32. Low activity found in these three locations mainly because these three stations are open and unsheltered area which very likely cannot prevent the runoff of mineral, studies are still underway to find the actual causes. Except those low activity stations, the activity concentration of ^{226}Ra that between 8.7 – 76.9 Bq/kg and of ^{238}U between 8.7 – 80.5 Bq/kg found in this study is normal for soil in Malaysia (^{226}Ra , 38 – 94 Bq/kg; ^{238}U , 49 – 86 Bq/kg) reported in UNSCEAR (2000).

Meanwhile, there are two obvious locations found with high activity of ^{232}Th , i.e near point LY 01 and LY 02; and at area near point LY 15 and LY 23. Again, low activity is observed at region near point LY 05 and LY 12. The activity concentrations for ^{232}Th ranged from 6.2 to 121.5 Bq/kg with a mean value of 58.2 Bq/kg. Besides of those low activity points, activity value found in this study also comparable to normal soil reading in Malaysia (range 63 – 110 Bq/kg, mean 82 Bq/kg) (UNSCEAR, 2000). The activity concentrations for radionuclides indicating that activity for ^{232}Th is generally higher when compared to that of ^{226}Ra in most stations. This mainly due to the nature of the decay properties for the parents as ^{228}Ra (daughter of ^{232}Th) which has shorter half life of 5.75 years will be producing progenies much faster as compared to ^{226}Ra which has half life of 1600 years (Krest et al., 1999; Moore et al., 1995).

On the other hand, the activity distribution of ^{40}K indicates several high activity locations near LY 01, LY 02, LY 06, LY 07, LY 16, LY 19 and LY 26. The activity concentrations for ^{40}K ranged from 19.1 to 778.9 Bq/kg with a mean of 221.3 Bq/kg, the mean is low as compared to the range reported in UNSCEAR (2000) (range 170 – 430 Bq/kg, mean 310 Bq/kg). Variation of ^{40}K from one location to another is possible as it has a highly mobilize (easy dissolve) radionuclides and widely used in industrial materials (fertilizer, chemicals, etc.) as Gebeng industrial area itself consists of various kind of heavy industries.

Calculation of Radiological Effects

The most widely used radiation hazard index is called the radium equivalent activity, Ra_{eq} . The radium equivalent activity is a weighted sum of activities of the ^{226}Ra (^{238}U), ^{232}Th and ^{40}K radionuclides based on the assumption that ^{226}Ra , 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma ray dose rate (Ahmed and El-Arabi, 2005; Stranden, 1979; Yang et al., 2005). Radium equivalent activity can be calculated from the following relation as suggested by Berekta and Mathew (1985).

$$\text{Ra}_{\text{eq}} = A_{\text{Ra}} + 1.43 A_{\text{Th}} + 0.077A_{\text{K}} \quad (1)$$

Where, A_{Ra} , A_{Th} , A_{K} are the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K , respectively. To be non-hazardous, the calculated Ra_{eq} should not exceed a maximum value of 370 Bq/kg (UNSCEAR, 1982).

Another radiation hazard index called the representative level index, I_{r} , is defined as the following formula (Alam *et al.*, 1999; NEA-OCED, 1979).

$$I_{\text{r}} = \frac{1}{150\text{Bq/kg}} A_{\text{Ra}} + \frac{1}{100\text{Bq/kg}} A_{\text{Th}} + \frac{1}{1500\text{Bq/kg}} A_{\text{K}} \quad (2)$$

Where, A_{Ra} , A_{Th} , A_{K} having the same meaning as in Eq. (1) above.

Besides these two indicators, External hazard index (H_{ex}) is another parameter that enables us to evaluate the additional radiological hazard of natural gamma-radiation to the people exposed to the radiation daily. The calculation was performed using equation as reported earlier by Yang *et al.* (2005) and Nabil *et al.* (2010).

$$H_{ex} = \frac{Q_U}{370} + \frac{Q_{Th}}{259} + \frac{Q_K}{4810} \leq 1 \quad (3)$$

where Q_U , Q_{Th} , and Q_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K , respectively.

Also, the total air absorbed dose rate (nGy/h) due to the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq/kg can be calculated using the formula given by Beck *et al.* (1972), Tzortzis *et al.* (2003) and UNSCEAR (1988).

$$D = 0.429A_U + 0.666A_{Th} + 0.042 A_K \quad (4)$$

Where, A_U , A_{Th} and A_K are the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K , respectively, in Bq/kg. Beck *et al.* (1972), derived this equation for calculating the absorbed dose rate in air at 1.0 m above ground from measured radionuclides concentration in environmental materials.

The Radium equivalent activity (Ra_{eq}), Representative level index (I_{yr}), External hazard index (H_{ex}) and total air absorbed dose rate at 1 m above ground of the present work, had been calculated from the mean NORM activity at each station, and illustrated as shown in Figs. 3 – 6 and summarized in Table 1 together with results from the other studies.

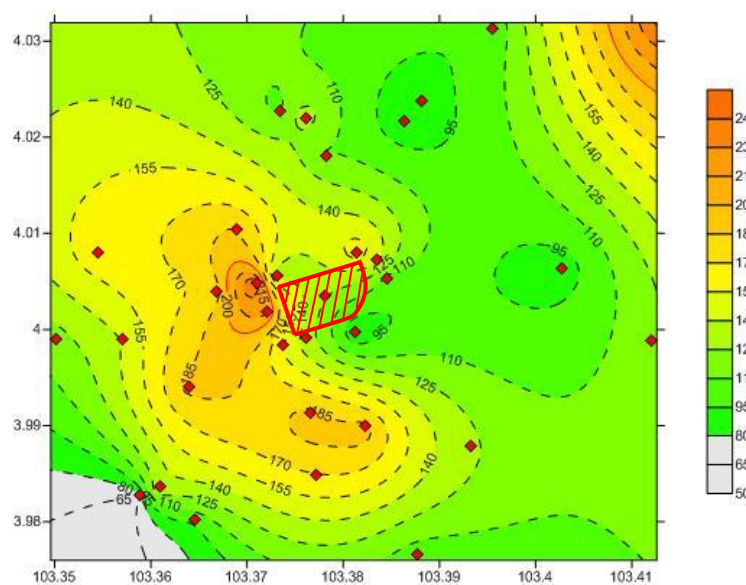


Figure 3: Ra_{eq} (Bq/kg) surrounding Lynas (Red shaded)

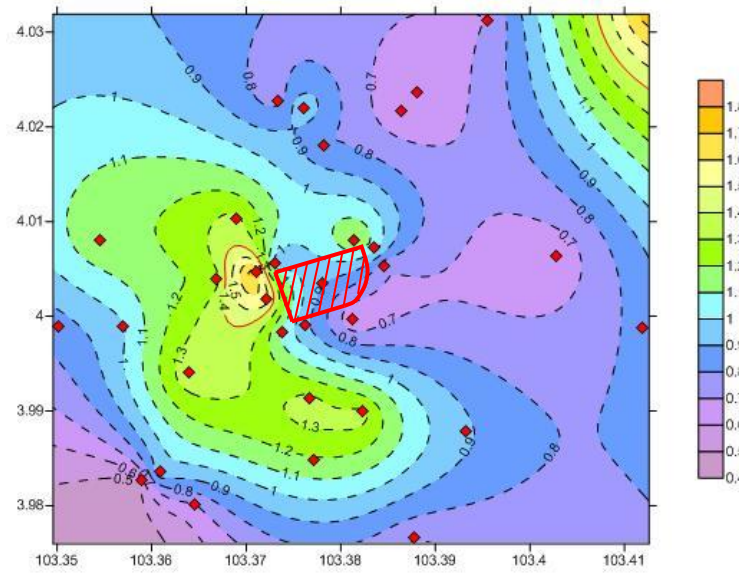


Figure 4: I_T value surrounding Lynas (Red shaded)

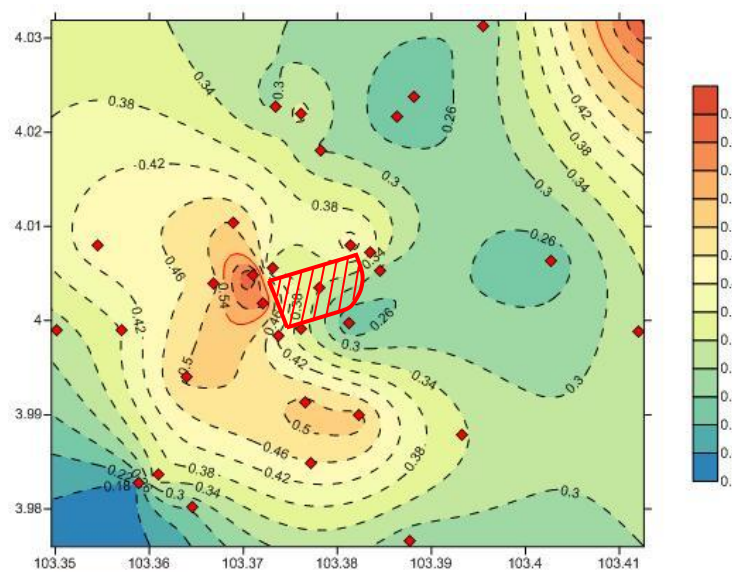


Figure 5: H_{ex} value surrounding Lynas (Red shaded)

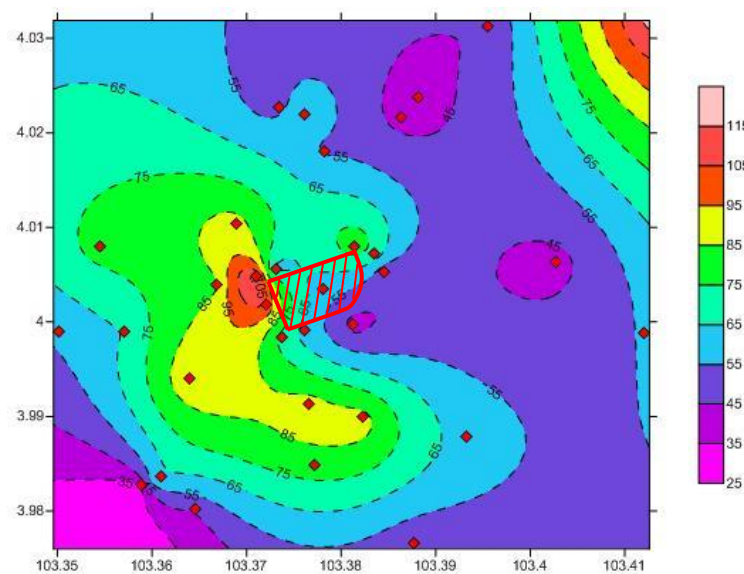


Figure 6: Total air absorbed dose rate (nGy/h) surrounding Lynas (Red shaded)

Figures 3 – 6 shows that, either approach of calculations, i.e. the Ra_{eq} , I_{yr} , H_{ex} and dose rate all indicating that the high radiation hazard locations are found near station LY 01 and LY 02. This location therefore had potential to cause risk of radiation exposure to people moving in that area. Radium equivalent activity and External hazard index is radiation hazard index, which is used to evaluate the radiation hazards of the gamma rays due to the present of NORM radionuclides. The calculated value for that of radium equivalent activity (Ra_{eq} , range 61 – 247 Bq/kg, mean 136.7 Bq/kg) and external hazard index (H_{ex} , range 0.2 – 0.7, mean 0.4) are well below the recommended value of 370 Bq/kg and 1, respectively, showing that the natural radiation at this area (base on radionuclide concentrations found in samples) does not necessary adding extra radiation risks to the people even though they are within the study area.

Meanwhile, calculated representative level index are between 0.4 – 1.7 (I_{yr} mean \sim 1.0). In general this value was also less than the background level ($I_{yr} \sim$ 1.5) in Peninsular Malaysia's soil. Only soil sample from LY 01, LY 09 and nearby areas having values slightly higher than that of the background level.

The calculated total air dose rate (Fig. 6) for all stations were ranged between 29 – 118 nGy/h with the mean absorbed dose rate of 65 nGy/h. This value is slightly higher than the estimated average global terrestrial radiation of 55 nGy/h (range 28 – 120 nGy/h) reported by UNSCEAR (1993), 57 nGy/h (range 18 – 93 nGy/h) reported by UNSCEAR in 2000 and those reported in Vietnam's surface soil of 62 nGy/h by Hien et al. (2002). However, this value is much lower as compared to the value reported for Peninsular Malaysia (mean 92, range 55 – 130 nGy/h), Thailand (mean 77, range 2 – 100 nGy/h) which also reported in UNSCEAR 2000 and some other place like in Nile Island of Egypt that reported a dose rate of 82.7 nGy/h by Ahmed and El-Arabi (2005) and 124 nGy/h at Xiazhuang granite area soil by Yang et al. (2005).

Table 1: Comparison of radium equivalent activity, representative level index, external hazard index and total air absorbed dose rate of the present work and other studies

Location/ Country	Sample	Radium Equivalent (Bq/kg)	Represent ative Level Index ($I_{\gamma r}$)	External Hazard Index (H_{ex})	Total air dose rate (nGy/h)	References
Nile Island, Egypt	Soil	152.9	1.3	0.4 ^a	82.7	Ahmed and El- Arabi (2005)
Brazil	Soil	147.8 ^a	1.1 ^a	0.4 ^a	72 ^a	Malanca <i>et al.</i> (1993)
India	Soil	86.7 ^a	0.6 ^a	0.2 ^a	74	Narayana <i>et al.</i> (2001)
Thailand	Soil	138.6 ^a	1.0 ^a	0.4 ^a	77	UNSCEAR (2000)
Japan	Soil	96.9 ^a	0.7 ^a	0.3 ^a	53	UNSCEAR (2000)
Xiazhuang, China	Soil	266.0	0.3 – 1.7	0.7	124	Yang <i>et al.</i> (2005)
Vojvodina, Yugoslavia	Agri. soil	150 ^a	1.1 ^a	0.4 ^a	71.2 ^a	Bikit <i>et al.</i> (2005)
Zheziang, China	Soil	184.9 ^a	1.4 ^a	0.5 ^a	90.6 ^a	Ziqiang <i>et al.</i> (1988)
China average	Soil	161.5 ^a	1.2 ^a	0.4 ^a	77 ^a	Wang (2002)
World average	Soil	89.2 ^a	0.7 ^a	0.2 ^a	43 ^a	UNSCEAR (1988)
World average	Soil	118.5 ^a	0.9 ^a	0.3 ^a	55	UNSCEAR (1993)
World average	Soil	128.7 ^a	0.9 ^a	0.3 ^a	59	UNSCEAR (2000)
Malaysia (Peninsular)	Soil	208.1 ^a	1.5 ^a	0.6 ^a	92	UNSCEAR (2000)
Lynas	Soil	61 – 247 (136.7)	0.4 – 1.7 (1.0)	0.2 – 0.7 (0.4)	29 – 118 (65)	<i>Present work</i> ^b

^aCalculated by the author using data given in the reference

^bValue in bracket is the mean value for all stations

The dose equivalent that an individual staying at the area might be expected to receive from such terrestrial natural gamma radiation was estimated to be from 36.6 to 148.9 μSv per year with mean dose equivalent at 82.0 μSv per year. This value is far below than the annual dose limit of 1,000 μSv per year for general public. In obtaining these values, it was assumed that people spent on the average 20% of their time outdoors and a conversion factor of 0.72 Sv/Gy was used (UNSCEAR, 1988; Yang et al., 2005).

The results for the radium equivalent activity, representative level index, external hazard index and total air dose rate of the present work and other studies are compared in Table 1. It is found that, in general, the value of Ra_{eq} , $I_{\gamma r}$, H_{ex} calculated for the soils in this study is comparable to those reported elsewhere and less than recommended figures, indicating that the risk of radiation

exposure for the people in this vicinity are smaller as compared to the general Peninsular Malaysia's background radiation received by them. Therefore, base on the outcome of the study in 2014, there is no evidence of radioactive enhancement found at the Lynas's surrounding vicinity.

CONCLUSIONS

From this study, it can be concluded that:

1. The concentration of ^{226}Ra were ranged between 8.7 – 76.9 Bq/kg with mean value of 33.7 Bq/kg; the activity of ^{238}U from 8.7 to 80.5 Bq/kg with mean value of 36.9 Bq/kg; ^{232}Th ranged from 6.2 to 121.5 Bq/kg with mean value of 58.2 Bq/kg and ^{40}K from 19.1 to 778.9 Bq/kg with mean 221.3 Bq/kg. The activity standard deviation of ^{226}Ra , ^{238}U , ^{232}Th and ^{40}K was 14.2 Bq/kg, 14.7 Bq/kg, 26.1 Bq/kg and 185.6 Bq/kg, respectively.
2. The calculated hazardous value for radium equivalent activity (Ra_{eq} , range 61 – 247 Bq/kg, mean 136.7 Bq/kg); external hazard index (H_{ex} , range 0.2 – 0.7, mean 0.4); representative level index between 0.4 – 1.7 (I_{yr} mean ~ 1.0); total air dose rate between 29 – 118 nGy/h with the mean dose rate of 65 nGy/h were performed.
3. Location near station LY 01 and LY 02 have highest potential causing radiation hazard to the people whilst the other locations pose low radiation risk.
4. Estimated mean dose equivalent of 82.0 μSv per year will be received by an individual from terrestrial natural gamma radiation at the study area, but this value is still far below the annual dose limit for general public.
5. No evidence of radioactive enhancement found at the study area.

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