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Assessment of Natural Radioactivity Level and Radiological Index in the Vicinity of Lynas Rare-earth Processing Plants

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The findings of the study on assessment of natural radioactivity level and radiation hazard parameters in the vicinity of Lynas rare-earth processing plants are reported. This study aims to quantify the environmental levels of natural radionuclides in soil collected in the vicinity of Lynas rare-earth processing plants and thereby to assess potential radiological hazards to the environment. About 31 terrestrial sampling locations were chosen for collection of the soil samples. The activity concentrations of the naturally occurring radioactive material (NORM) members, i.e. ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K were measured using HpGe gamma spectrometer after reaching 30 days of secular equilibrium with their daughters. The mean activity concentration of ^{226}Ra ^{228}Ra ^{238}U, ^{232}Th and ^{40}K in soil samples were 35 Bq/kg, 62 Bq/kg, 38 Bq/kg, 60 Bq/kg and 245 Bq/kg, respectively. The estimated Ra_{eq} and H_{ex} readings due to natural environmental radiation in respectively lower than the recommended value of 370 Bq/kg and unity. Meanwhile, the total air absorbed dosage rate was slightly higher than the estimated average global terrestrial radiation but much lower compared to other regions in Malaysia. The results indicated that the radiation hazard in the vicinity of the Lynas rare-earth processing plants was negligible. Thus, it could be concluded that there were no additional radiation level and no radiological hazard effects to the people living in the surrounding areas.

Key words: NORM; radioactivity; radiological; rare-earth processing

Assessment of natural ambient radioactivity and its radiological effects in the environment play an important role to protect the health hazard on the environment and general public due to the radiation (Kasoga *et al.* 2015). The contribution of ambient radioactivity to the background radiation in the environment is mainly from two prominent natural sources, i.e. high-energy cosmic ray particles from the atmosphere and radioactive nuclides that originated from the earth crust which is present everywhere in the environment, including the human body (UNSCEAR 2000). The primary source of terrestrial radiation received by humans is of the naturally occurring radioactive material (NORM) associated and deposited with the formation of the earth's crust such as rocks, soils, ores, minerals, sediment, etc. The NORMs which are derived principally 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 has been washed and drained through

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several pathways such as weathering, erosion, fallout, rainwater and human activities into rivers transport and finally ended in the marine environment through estuary (Ahmad-Taufek 2004; Akram *et al.* 2004). In the terrestrial environment, these radioactive series are present in the soil and soil acts as a medium for transferring radionuclides in air and vegetables (Balakrishnan 2015).

These radioactive nuclides release radiation everywhere in our surroundings to which humans are exposed (Kasoga *et al.* 2015). In other words, although natural radioactivity is present at very low levels almost anywhere in the natural environment, everyone is exposed to it in air, food, and water. These amounts in the air are usually so small and do not constitute a health hazard (Yii *et al.* 2016). Also, the radiation exposure to the general public is due to unplanned and uncontrolled of human activities (e.g. industrial activities). Thus, uncontrolled activities involving NORM can contribute to ambient radioactivity in the environment, thus unwanted exposure and dispersal pose a risk to human health and the environment. Industrial activities such as oil and gas extraction, coal fired power generation, phosphate industries, zircon/zirconium industry, mining and processing of metals and rare-earth, etc. have been reported as potential sources of elevated naturally occurring radionuclides. The presence of NORM with elevated radionuclides concentrations could be an issue at any stage of an operation from the mineral feed stock, intermediate products, final products and the wastes generated during the process (IAEA 2005). In the past, the issue of NORM and their potential hazards associated with human health and contaminating the environment often raised concerns by the public but now many efforts on such environmental monitoring are implemented, and the problem can be solved and the potential radiological hazards can be reduced.

Study of the background or ambient radioactivity present in natural environmental samples such as vegetation, water, air, and soil provide vital information as they help to monitor the radioactivity levels found in the surroundings and also give an indication of changes in the radiation levels due to human activities (Hu *et al.* 2010). These studies also help to identify and evaluate the distribution of radionuclides in the area (IAEA 2010). Therefore, the present study is carried out with the primary objective to quantify the environmental levels of natural radionuclides in the soil collected at the vicinity of Lynas rare-earth processing plants and thereby to assess potential radiological hazards to the environment.

EXPERIMENTAL DETAILS

Sampling Area

Lynas rare-earth processing plants are situated in Gebeng Industrial Estate, adjacent to the Balok River. The estate lies within the capital city Kuantan in the Pahang state. The study areas were established within 3 km radius surrounding the plant and located within at a Latitude 3.97^{o} to 4.03^{o} North and Longitude 103.34^{o} to 103.42^{o} East (*Figure 1*).

Soil Sample Collection

About 31 soil samples were collected at randomly selected sampling points surrounding the Lynas Plants (*Figure 1*) in May 2014 to September 2015 which involved seven trips of sampling activities. Approximately 1 kg of composite surface soil samples was taken using a coring tool at less than 5 cm depth with

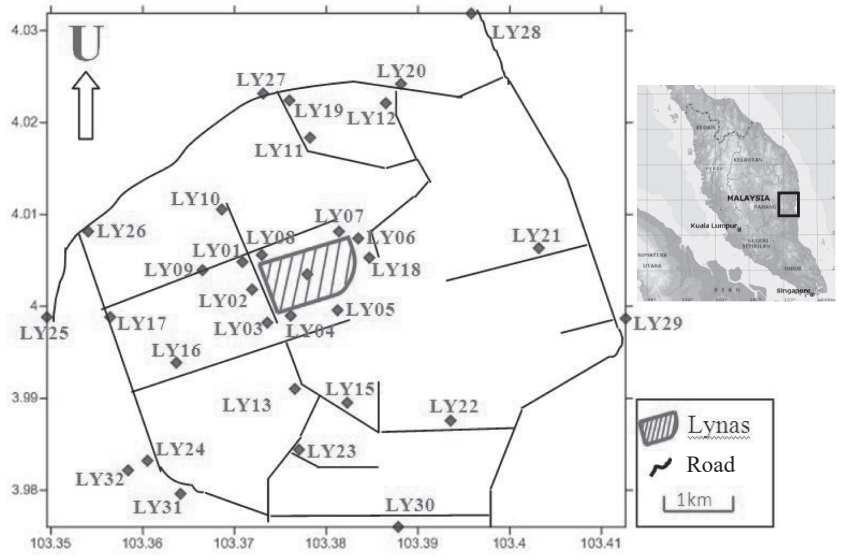

Figure 1. Location of the sampling points at the study area.

one-metre square area for each point. Stones, pebbles, vegetation, dried leaves, roots, etc. were cleared and removed from soil samples; then the collected samples were place into HDPE plastic bag, properly labelled and sealed before brought back to the laboratory for further analysis.

Sample Preparation

The collected soil samples were individually weighed, transferred into a steel tray and dried at 105°C in an electric scientific oven for a minimum of 24 hours until a constant weight was achieved. Then, the dried samples were ground to powder form and passed through a 200 mesh sieving machine. Samples were transferred into a 600 ml Marinelli beaker, weighed and sealed with thick PVC tape to inhibit radon gases from escaping (Yii *et al.* 2016). All samples were then stored for a period in excessive of 30 days (>7 half-live of ^{222}Rn and ^{220}Rn) to establish secular equilibrium between parents and with their respective radioactive progeny before to gamma counting (Dowdall 2002; Yang *et al.* 2005).

Gamma Counting

Procedure for gamma counting was summarized from the technical report by Yii *et al.* (2016). All samples were individually counted using high purity germanium (HPGe) gamma spectrometry system with the p-type detector of 25% relative efficiency for 50 000 seconds. It was calibrated using customized gamma multi-nuclides standard sources comprising of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co in the same counting geometry. A container with the same geometry filled with inert materials counted during the weekend was used to determine the background counts. All measurements were corrected to the density and reference date.

The activities of ^{226}Ra, ^{228}Ra, ^{232}Th, and ^{238}U were calculated through their progeny energy peaks i.e. ^{214}Pb and ^{214}Bi for ^{226}Ra, and

^{238}U; ^{228}Ac for ^{228}Ra; and ^{212}Pb, ^{228}Ac, and ^{208}Tl for ^{232}Th. Meanwhile, ^{40}K was calculated via directly its energy peak (Yang *et al.* 2005; El-Reefy *et al.* 2006). All activity calculations were corrected to the density and sampling date. The minimum detectable activities (MDA) for ^{226}Ra, ^{228}Ra, ^{232}Th, and ^{238}U were 1 Bq/kg after considering the sample size and counting time and ^{40}K was 5 Bq/kg (Yii *et al.* 2016).

Calculation of Radiological Index

The radiological index consists of several parameters. Among these are:

(i) Radium equivalent activity Ra_{eq} concentration index.

Radium equivalent activity concentration index with a symbol of Ra_{eq} is among well known and most widely used to estimate radiation hazard index or radiation hazard parameter or radiological index. In this estimation, the specific activities of radium, thorium, and potassium in different combinations in soil samples were compared, where Ra_{eq} is defined as bellows (Beretka & Mathew 1985):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.07A_{K}$$

Where, C_{Ra}, C_{Th} and C_K are the specific activity concentrations for ^{226}Ra (^{238}U), ^{232}Th and ^{40}K (in Bq/kg), respectively. This equation is based on the estimate that 1 Bq/kg of ^{226}Ra (^{238}U), 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K generate the same γ- ray dosage rate (Stranden 1979; Yang *et al.* 2005; Ahmed 2005). For the non-hazardous, the calculated Ra_{eq} should not exceed a maximum value of 370 Bq/kg as reported by UNSCEAR (1982).

(ii) External radiation hazard, H_{ex}:

To estimate the additional radiological hazard on the people which are daily exposed to the natural gamma radiation from soil, the external radiation hazard, H_{ex} was calculated using the following equation as reported earlier by Yang *et al.* (2005) and Nabil *et al.* (2010):

$$H_{ex} = (A_U/370) + (A_{Th}/259) + (A_K/4810) \leq 1$$

Where, A_U, A_{Th} and A_K are the activity concentrations for ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in Bq/kg, respectively. The values of the indices should be ≤ 1 (Krieger 1981).

(iii) The total air absorbed dosage rate

The total air absorbed dosage rate, D (nGy/hr) due to the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg is estimated using equation given by UNSCEAR (2000), Venkidasamy *et al.* (2011), UNSCEAR (2008) and, Sheela and Shanthi (2016);

$$D = 0.462A_U + 0.604A_{Th} + 0.0417A_K$$

Where, A_U, A_{Th} and A_K are the activity concentrations for ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in Bq/kg, respectively. Sheela and Shanthi (2016) derived this equation for calculating the total air absorbed dosage rate in air at the height of 1.0 m above from the ground that measured the activity concentrations for uniform distribution of naturally occurring radionuclides in the environmental materials.

RESULTS AND DISCUSSION

Radioactivity Level of ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K in Soil

The results of the activity concentrations (Bq/kg) of ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K in soil samples at a different location in the study area are presented in *Figure 2*. The activity concentration of ^{226}Ra in soil samples ranges from 8.7 Bq/kg to 95.4 Bq/kg. For ^{228}Ra found to be in the range of 6.6 Bq/kg to 134.0 Bq/kg. The activity concentration of ^{238}U and ^{232}Th varied from 8.7 Bq/kg to 106.0 Bq/kg and 6.2 Bq/kg to 130.0 Bq/kg, respectively. Meanwhile, the activity concentration of ^{40}K in soil found to vary from 10.6 Bq/kg to 1160.0 Bq/kg. These results showed a broad range of radioactivity level where some locations appeared to be higher. This noticeable difference may be attributed to the geochemical composition and origins of soil types in particular study areas (Dabayneh *et al.* 2008; Thabayneh & Jazzar 2012). Moreover, the higher values of the activity concentrations belong to soil samples which may be attributed to soil types which were probably radioactive-rich granite, phosphate, sandstone, and quartzite (UNSCEAR 1993; UNSCEAR 2002).

The average activity concentrations of ^{226}Ra measured in the present study were comparable to the value reported for offsite Lynas Plant sampling stations during the pre-operational as well as the value reported for worldwide and other countries such as China, Japan, India as summarized in *Table 1*. Thorium-232 (^{232}Th) found to be higher average values compared to Thailand, China, Japan and world values. The high variability of result attributed mainly to the typical concentration of the radioactive materials in soil, especially in Malaysian soil (Ismail 2009). Meanwhile, activity concentration of ^{40}K in the soil at the present study found to be far lower than other countries. Observation revealed that the measured activity concentration of ^{40}K significantly exceeded other natural radionuclides. This shows that ^{40}K is a more abundance radionuclide compared to other radionuclides in the soils (Thabayneh & Jazzar 2012). Furthermore, due to ^{40}K is a highly mobile and easy to dissolve radionuclide, its variation might be due to spreading of widely used industrial materials (fertilizer, chemicals, etc.) at Gebeng industrial area (Yii *et al.* 2016). Another result showed the activity concentrations of ^{232}Th (^{228}Ra) were higher than ^{238}U (^{226}Ra) at all studied locations. This was mainly due to the decay property that may play an important role in rapid generating of ^{228}Ra (half-life: 5.75 years) from its parent of ^{232}Th compared to ^{226}Ra with long half-life of 1602 years which decay from ^{232}U (Moore *et al.* 1995; Krest *et al.* 1999).

Radiological Index

The radium equivalent activity concentration index, Ra_{eq}, the external radiation hazard, H_{ex} and the total air absorbed dosage rate, *D* are summarized in *Figure 3*. The radium equivalent activity concentration index, Ra_{eq} and the external radiation hazard, H_{ex} are radiological index, defined as the radiation hazard index which is used to assess the radiation hazard of the gamma ray due to the present of NORM radionuclides (Yii *et al.* 2016). Their estimations varied from 19.0 – 335.9 Bq/kg (mean: 139.6 Bq/kg) and 0.05 – 0.94 (mean: 0.39), respectively. The value of Ra_{eq} found to be not exceeding a maximum value of 370 Bq/kg as reported by UNSCEAR (1982). Meanwhile, the value of the H_{ex} indices was less than unity (the values of the H_{ex} should be ≤ 1) (Krieger 1981). Thus, these findings reflected negligible radiation hazard in the vicinity of Lynas plants negligible. In other words, there were no additional radiation level and no radiological hazard that may effect the people living in the surrounding areas.

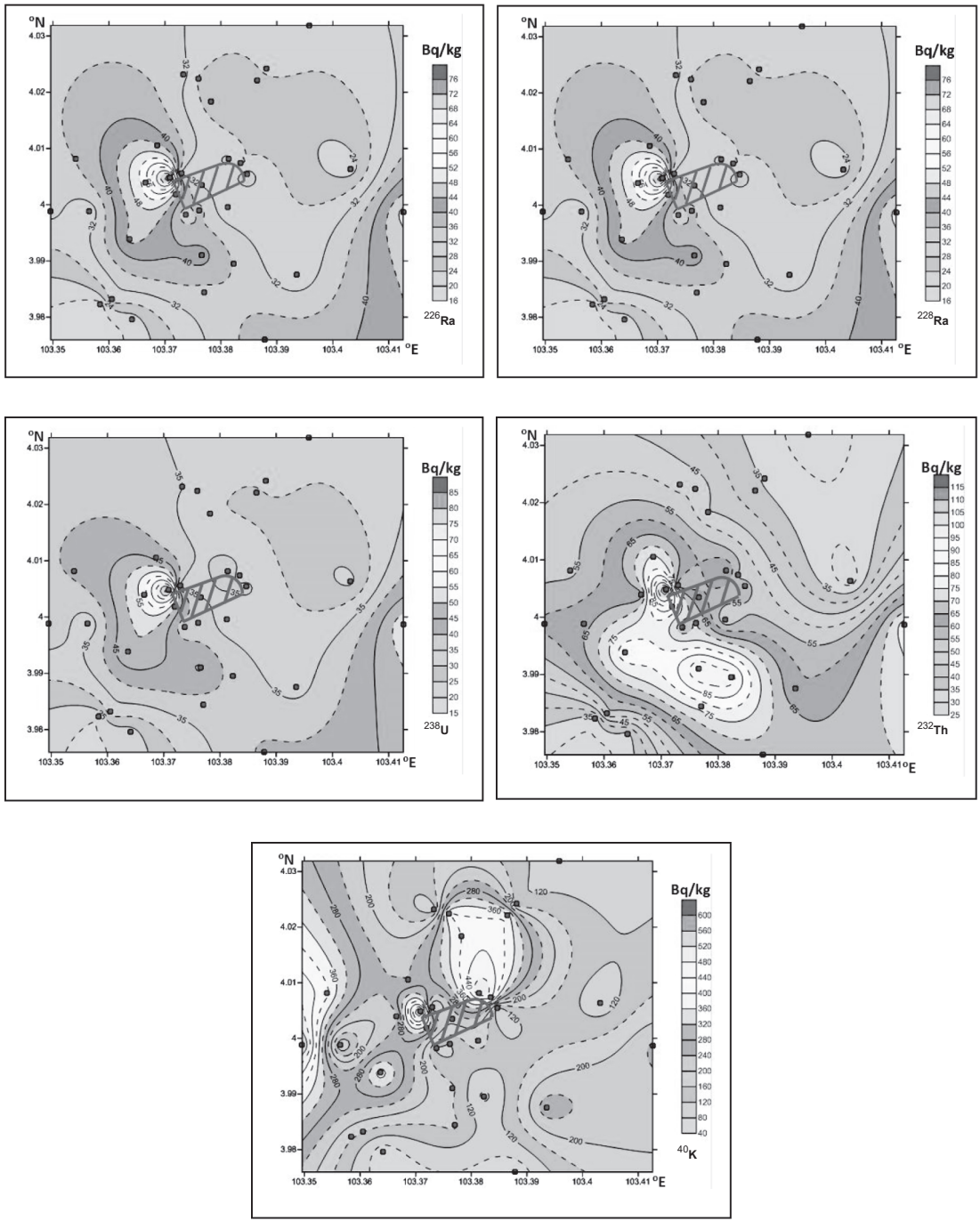

Figure 2. Soil radioactivity level of ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th *and* ^{40}K *in the vicinity of Lynas plant.*

Table 1. Comparison of radioactivity concentrations for ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K in soil.

Location	Level	Activity concentration of radionuclide (Bq/kg)				
		^{226}Ra	^{228}Ra	^{238}U	^{232}Th	^{40}K
This study	Minimum	9	7	9	6	11
	Maximum	95	134	106	130	1160
	Average	35	62	38	60	245
Onsite Lynas plant (pre-operational) (Ismail 2009)	Minimum	13	25	–	–	–
	Maximum	56	178	–	–	–
	Average	26	64	–	–	–
Offsite Lynas plant (pre-operational) (Ismail 2009)	Minimum	12	10	–	–	–
	Maximum	57	83	–	–	–
	Average	32	69	–	–	–
Pahang (Ismail 2009)	Average	71	88	–	–	–
Peninsular Malaysia (Ismail 2009)	Average	74	98	–	–	–
Malaysia (UNSCEAR 2000)	Minimum	38	–	49	63	170
	Maximum	94	–	86	110	430
	Average	67	–	66	82	310
Malaysia (Mohsen *et al.* 2007)	Minimum	41	–	–	105	75
	Maximum	90	–	–	516	848
	Average	–	–	–	–	–
Phuket Island (Chanyotha 2011)	Minimum	20	–	–	25	36
	Maximum	390	–	–	530	2610
	Average	94	–	–	190	720

Table 1 Cont. Comparison of radioactivity concentrations for ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K in soil.

Location	Level	Activity concentration of radionuclide (Bq/kg)				
		^{226}Ra	^{228}Ra	^{238}U	^{232}Th	^{40}K
Thailand (UNSCEAR 2000)	Minimum	11	–	–	7	7
	Maximum	78	–	–	120	712
	Average	48	–	–	51	230
China (UNSCEAR 2000)	Minimum	2	–	–	1	9
	Maximum	440	–	–	360	1800
	Average	32	–	–	41	440
Hong Kong (UNSCEAR 2000)	Minimum	20	–	–	16	81
	Maximum	110	–	–	200	1100
	Average	59	–	–	95	530
Japan (UNSCEAR 2000)	Minimum	6	–	–	2	15
	Maximum	98	–	–	88	990
	Average	33	–	–	28	310
India (UNSCEAR 2000)	Minimum	7	–	–	14	38
	Maximum	81	–	–	160	760
	Average	29	–	–	64	400
Worldwide (UNSCEAR 2000)	Average	35	–	–	30	400
World average (Yii *et al.* 2016)	Average	–	–	45	33	420

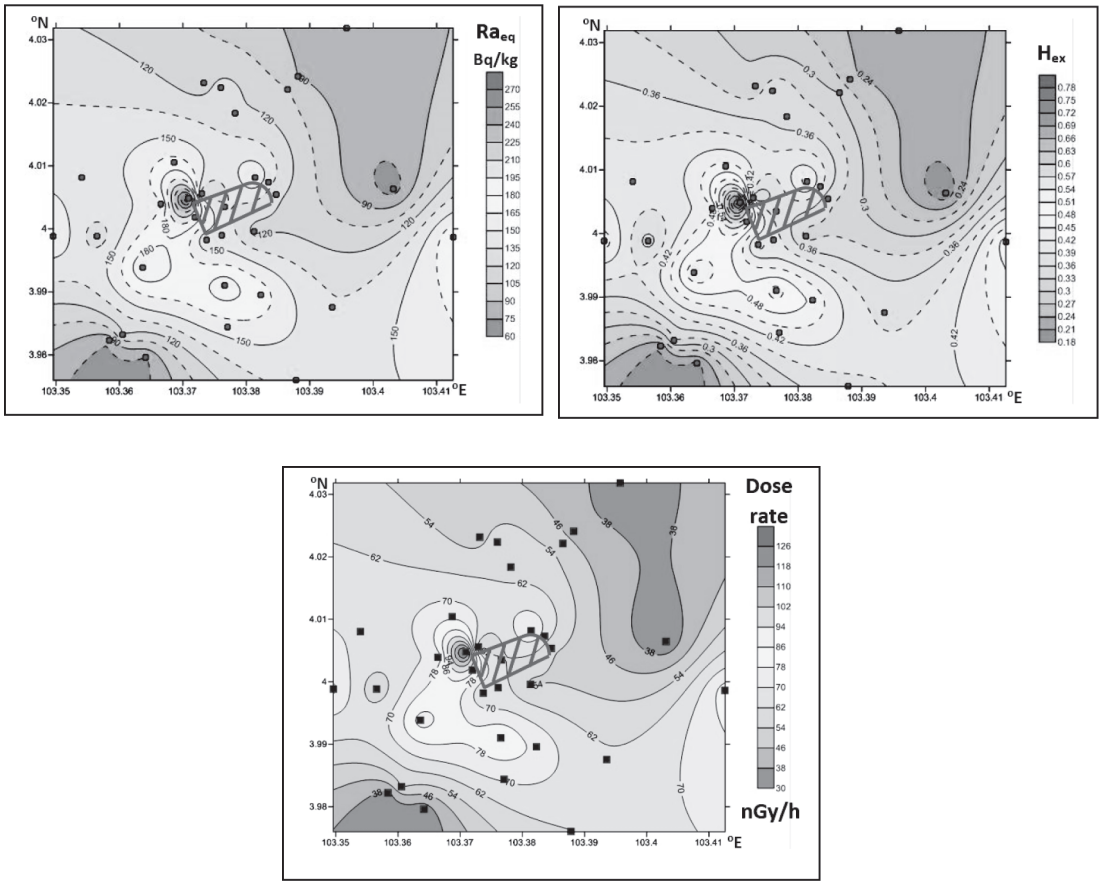

Figure 3. Profile of radiological index in the vicinity of Lynas plant.

The total air dosage rate for the outdoor environment calculated from the absorbed dosage rate in the study area was found to be varying from 8.6 – 157.1 nGy/h with a mean value of 64.2 nGy/h. This was slightly higher than the estimated average global terrestrial radiation of 55 nGy/h (range: 28 – 120 nGy/h), 57 nGy/h (range: 18 – 93 nGy/h) and 62 nGy/h reported by UNSCEAR (1993), UNSCEAR (2000),] and Hien *et al.* (2002), respectively. However, this value is much lower compared to the value recorded in Ulu Tiram with a range of 96 – 409 nGy/h (mean: 200 nGy/h) (Abdul-Rahman 2007), 55 – 130 nGy/h (mean: 92 nGy/h) for Peninsular Malaysia (UNSCEAR 2000), and 2 – 100 nGy/h (mean: 77 nGy/h) for Thailand (UNSCEAR 2000). The radiological index or radiation hazard parameters at some locations in the study area slightly exceeded the UNSCEAR average range but lower than Peninsular Malaysia might be due to natural occurrence of a relatively high activity concentration of natural radionuclides in the soil at particular locations. Usually, they were attributed to soil types which probably contained natural radioactive-rich minerals such as granite, monazite, quartzite, etc.

CONCLUSIONS

The activity concentration level of ^{226}Ra, ^{228}Ra, ^{238}U, ^{232}Th and ^{40}K measured in the 31 soil samples collected from the vicinity of Lynas rare-earth processing plant were the broad range where some small locations appeared to be higher. These relatively higher values and noticeable difference in some locations might be attributed to the geochemical composition and origins of soil types in particular study areas. Thus, the impacts of these natural radionuclides and the corresponding additional external radiation if any, exposure to the public were almost negligible. Consequently, the measured levels of these natural radionuclides confirmed that there were no hazard effects on the people residing in the vicinity of the Lynas Plant. Apart from that, the finding of this study could contribute to the setting up of a reference level for studies about natural radioactivity in the soil samples of this region in future.

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