A Study of Kuantan, Pahang Malaysia Terrestrial Background Radiation in Determination of Uranium & Potassium-40

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***Abstract*—Human is always exposed to radiation externally and internally from the early civilization due to various activities. Since Lynas Corporation which is the rare earth processing company is developed in the industrial area of Gebeng in Kuantan, and was said will lead to the production of higher amount of radiation, there is a need to do this study in order to measure the level of radiation exposure of the terrestrial area around Kuantan. This study will develop Kuantan district terrestrial background radiation baseline data and establish the preliminary data on Kuantan terrestrial based on the readings observed via gamma ray detector, analysis via Induced Couple Plasma-Mass Spectrophotometer for determination of uranium content and Atomic Absorption Spectrophotometer for determination of potassium-40. This study is carried out at ten**

different sampling locations around Kuantan at certain longitudes and latitudes taking the location of Lynas Corporation as the reference point by using Global Positioning System (GPS) navigation device. The dose rate measurements were done by in-situ method using a portable gamma ray detector at 1 m above the ground. The analysis result of soils by using ICP-MS shows that the highest uranium-238 concentration in soil sample is 345.51µg/g which is at the sampling point 3 in Bukit Tanah Merah. On the other hand, the lowest concentration of uranium-238 is 65.24µg/g at the first sampling point which is located at Lynas. The concentrations of potassium-40 are in the range 0.005 ppm to 0.014 ppm. The concentration of the potassium-40 in the samples is in the range of standard concentration of soil in Kuantan terrestrial. The isotopic ratio of potassium-40 is 0.0117% with limit of detection value is 0.4 ppm. The amount of radiation exposure is still in safe level for human and other living organisms in Kuantan.

Keywords—radiation, uranium, potassium-40

I. INTRODUCTION

From the beginning of the civilization, human is always exposed to radiation either externally or internally. The exposure to the radiation for an average person is about 80% of which are comes from natural sources of radiation. There are three major sources of naturally occurring radiation which are come from cosmic radiation, the earth's crust and human body. Naturally occurring radiation commonly known as terrestrial radiation while radiation from human body commonly referred as internal sources. Terrestrial radioactivity is a natural source of radiation that is come from the rocks, ground, building materials and drinking water supplies. Natural uranium is one of the contributors to terrestrial source. Mostly, the terrestrial environment radiation whether it is natural or human-made is related to the component of the soil [1]. Organisms have been continuously exposed to cosmic rays in the atmosphere and from naturally occurring radionuclides throughout the life on Earth [2]. There were ubiquitously distributed in all living and non-living components of the biosphere [2]. Uranium determination in environmental samples is a tool for evaluating human exposure. The isotopes of natural uranium exist as a prominent health concern despite having relatively low specific activity because their decay products contribute to radio toxicity and chemical toxicity [3]. Those elements are exist in soil, rocks, water and other samples and it can be incorporated into human food chain through various routes such as inhalation and many more. According to Ramli et al., (2005) there are some areas having extremely high background radiation levels around the world. Substantial attention has been given to the soil radiation, basically for the goals of creating baseline data for future radiation impact evaluation, radioactivity protection and exploration [1]. Systematic gamma spectrometric analysis showed that the high natural background radiation observed is caused by the presence of monazite in that area [1].

The objectives of study to establish preliminary data on Kuantan terrestrial and background radiation baseline data based on the readings observed via gamma ray detector, to investigate the significant amount of uranium content in soil sample around Kuantan State terrestrial using inductively coupled plasma mass spectrometry (ICPMS) and to quantify the concentration of Potassium-40 in soil samples via Atomic Absorption Spectroscopy (AAS).

II. PROCEDURES

A. Sampling

The study was carried out around Kuantan, Pahang. The terrestrial radioactivity were measured at certain longitudinal and latitudinal line grids sampling points, where possible readings were made at each crossing point of the grid referring to the Lynas centre as the reference point by using Global Positioning System (GPS). Every sampling point constitutes one main spot of measurement and additional four points separated five kilometres apart from the main spot. Data of similar measurement readings were plotted as the same contour line of radioactivity to produce 1 to 50 000 scale map [4]. There were 10 sampling points observed and the readings taken were the average value of triplicates. For each sampling plot, the soil samples and counting signals data were collected.

B. Total decay radiation

The locations for the measurement are selected using Kuantan map which Lynas Corp. is used as a reference point. The other locations are separated approximately five kilometres apart from the main spot. The coordinates based on the latitudes and longitudes of the location at which the samples were taken were recorded by using the GPS device. All 10 locations are shown and marked on the map and numbered. The average readings of triplicates recorded for each location. The radiation was read in sv/hr as the unit of measurement. In this study, the detector used was sodium iodide (NaI) crystal doped with thallium (Tl) as an activator. When the radiation hits the detector, the ionization process occurred. Then, the ion pulses collected will send the electronic pulse to the instrument. The detector was placed one meter above the ground during observation because the detectors are reasonably sensitive to contributions from higher energy gamma radiation due to other environmental radionuclides. The meter was pre-calibrated before taking it to the field. The readings were taken after 1 minute of detection and the read-out of the detector is the average value of the radiation measurement. The measurement process was carried out in a sunny weather.

C. Uranium detection

All equipment used for weighing was of laboratory standard type. The analytical balance used was brand Mettler Toledo of type New Classic MF with model JS 1203C (Switzerland). The oven used in drying the soil also within laboratory

standard. The instrument used for the analysis purpose was Inductively Coupled Plasma Mass Spectrometry. The laboratory apparatus used such as filter funnel, laboratory glassware, digestion vessel, mortar and pestle, sieve tube, were of standard laboratory type. Glassware used in all experiments was rinsed with deionised water before being used. Plastic bags and scoop were makes sure to be free from contaminants. The filter papers used were of standard laboratory type. The materials used were nitric acid:hydrochloric acid (65:37), deionised water and soil samples. Soil samples were taken at each location. The top soil was dug by using shovel approximately 5 to 10 cm depth for every location. The soil samples were then transferred into clean plastic bags and were labelled for its coordinate and sampling locations. The plastic bags containing sample were tied. The time and environmental condition were noted. The soil samples collected were kept in a save place before proceed with sample preparation for analysis purpose by using inductively coupled plasma mass spectrometry (ICP-MS). Each sample was homogenized prior to digestion. All unwanted compound or solid substances such as stone and nails were removed. Samples were dried in oven for 24 hours at 100°C to remove moisture content in the soils. The step followed by sieving the sample. For sieving, each sample was crushed ground separately by using clean mortar and pestle until fine powder was obtained. The purpose of crushing it into fine powder is to allow the samples to be digested easily. The sample was then sieved through -60 mesh sieve of 0.25 mm size. The portion of sample that did not pass through the sieve was discarded. The digestion of the soil samples was then carried out. The soil samples were digested prior to analysis using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The type of digestion chosen was microwave digestion. The digestion was performed by using standard digestion method. According to this method, the acid digestion of the sample was done in a closed vessel device using temperature control microwave heating in order for the metal determination by spectroscopic methods.For this method of digestion, the instrument that is used the microwave acid digestion apparatus that is Milestone Ethos or Start lab station with internal temperature sensor, 640-260 terminal with easy CONTROL software installed and HPR1000/10S high pressure segmented rotor. The fine dry soil sample was put into the TFM vessel and weight up to 0.5 g. The TFM vessel was then introduced into the HTC safety shield. 65% of nitric acid and 37% of hydrochloric acid were added. The acids were added drop by drop in order to wet the samples that were stayed at the inner of the wall vessel. Then, the solution was swirled gently to homogenize the sample with the acids. The vessel was then closed. The step proceed by introduced it into the rotor segment followed by tighten it using the torque wrench. The segment was then inserted into microwave cavity and the temperature sensor was connected. The microwave program was run to completion. The rotor was then cooled by the air or water until the solution reached room temperature. The vessel was then opened and the solution was then transferred into the marked flask. The suitable personal protective equipment such as thermal resistance glove was always makes sure to be used while working with microwave system. In order to analyze the uranium composition in the digested soil sample by using Inductively Coupled Plasma Mass Spectrometry, five different concentration of uranium calibration standard solution were prepared by using PerkinElmer Pure Plus Multi –Element Calibration Standard 1. The concentration of the PerkinElmer Pure Plus Multi – Element Calibration Standard 1 is 10 ppm (mg/L) which equal to 10 000 ppb (µg/L). The standard solution prepared in which were 10 ppb, 20 ppb and 30 ppb respectively. The samples were sent to the Laboratory of Faculty of Technology for analysis of uranium-238 composition of mass to charge ratio by using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) from PerkinElmer with model NexION 300X. All the concentration of samples to be analysed was make sure in ppb value since the high concentration of sample (usually in ppm) will clogged the instrument. The results obtained by the analysis were in the form of ppb or µg/L [5][6].

D. Potassium-40 detection

Each sample was transferred into a clean dry beaker and placed it in a drying oven set at 105°C for 12 to 15 hrs. After drying, the sample was cooled to room temperature. The dust sample was sieve trough a 600 µmmesh size stainless steel sieve and the sieved material were collected. The soil samples were then sieved again through a 2 mm sieve. The sample was mixed throughly to achieve homogeneity. From the samples, 0.5 g of sample was weighed in triplicates and transferred into a test tube digestion tube. 10 mL of concentrated nitric acid was added into each test tube for each sample (ten samples) and were heated to 95°C for 30-40 minutes in water bath (in fume hood). Another 2 mL of nitric acid and hydrochloric acid was added into the samples until no brown fumes were given off from the sample. The samples were allowed to cool down for 20 min. The samples were transferred to 50 mL capacity volumetric flasks. The sample was filtered if necessary. The digestion tube should be rinsed with deionized water and transfer the solution to the volumetric flask. The flask was made up with deionized water to obtain a total volume of 50 mL. The samples were kept in the refrigerator (4°C) until analysis by using Atomic Absorption Spectroscopy. The series of calibration solution (multi-element) from standard stock solution (1000 mg/L) were prepared. The concentration of standard solutions was diluted to 100 ppm. After that, the standard solution is prepared in 0.5 ppm, 1.0 ppm, 1.5 ppm, and 2.0 ppm. Finally, samples were analyzed by using Atomic Absorption Spectroscopy.

III. RESULT & DISCUSSION

The reading of the radiation using gamma- ray detector was made on 1 m above the ground in order to measure the gamma radiation dose rate from 10 different locations around Kuantan based on the coordinates with the aid of Garmin GPS device. The locations are selected using Kuantan map which

Lynas Corp. is used as a reference point. The readings taken from the detector is measured using the unit of Sv/hr. The table below shows the data collected during the measurement process. The mean value and standard deviation value are calculated and the range value of the readings is stated in the table. The following table shows the name of each location based on the sample location number.

Table 1.1: Counting signal collection based on sampling plot

Location	Coordinate (N)	Coordinate (E)	Radiation (Sv/hr)
1	04°00.396‘	103°22.553‘	0.154
2	03°57.883‘	103°23.009‘	0.098
3	03°59.122‘	103°23.473‘	0.155
4	03°59.498‘	103°24.703‘	0.188
5	04°01.177‘	103°24.275‘	0.143
6	04°04.500‘	103°23.350‘	0.125
7	03°53.006‘	103°21.894‘	0.110
8	03°50.090‘	103°20.988‘	0.140
9	03°48.771‘	103°22.236‘	0.128
10	03°51.553‘	103°19.240‘	0.167
		Mean	0.1408 ± 0.02
		Range	0.098-0.188

Theoretically, it was assumed that the counting signal would be highest at sampling plot 1^o and being decrease when getting far from the Lynas Centre. However, in this study the data shown were not similar to the theoretical facts and the highest counting signal was at sampling plot 4^o. This may due to the surrounding of location plot which contribute the radionuclides in terms of chemical production since it was nearer to the chemical factories in Gebeng, Pahang. From the results obtained, it can be deduced that the amount of radiation from this study in Kuantan is in the range of 0.098- 0.188 Sv/hr and the mean value of the surface dose rate are 0.1408 Sv/hr at 1 m above the ground of the study area which is considered as normal range as the global range set by United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR is 0.5-1.3 Sv/hr [7]. In order to obtain the linearity of the sample to be analyzed, Uranium standard solution was prepared by using deionized water. The linearity was studied by evaluating the calibration curve at four level of concentration including blank matrix. The different levels of concentration were as described in Table 1.2 below.

Table 1.2 Concentration of standard calibration of uranium varies with intensity

	µg/L	Intensity (y-axis)	x/ppb
	10.00	262433.600	9.774
	19.43	458159.300	19.888
	28.61	622474.3	28.3781
Σ	58.040	1343067.200	58.040
Mean	19.347	447689.067	19.347
Std Dev	9.305	180248.567	9.314

Good linearity was shown with value of R^2 to be 0.998. A residual plot was constructed in order to confirm the linearity.

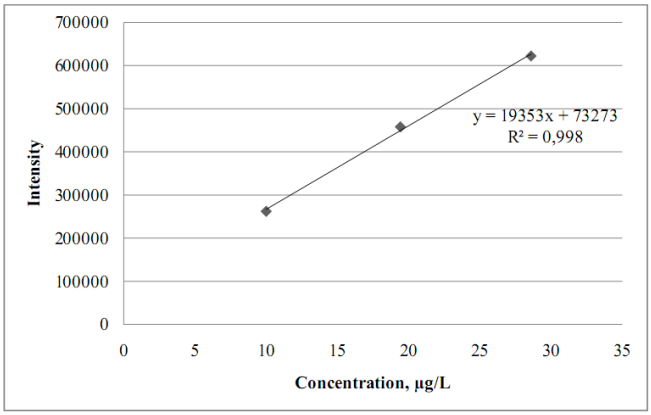

Figure 1.1: Standard calibartion curve of intensity versus concentration of uranium in ppb (µg/L)

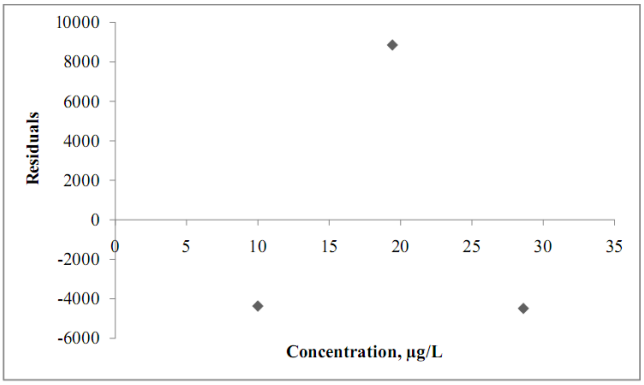

Figure 1.2: Standard calibration curve of residual plot versus concentration of uranium

The graph of intensity versus concentration of uranium was plotted in Figure 1.1. From the graph, there is a straight line with the correlation coefficient, r = 0.999. Hence, it can be concluded that there was strong positive linear relationship between intensity and concentration of uranium. However, the graph of residuals versus concentration of uranium in Figure 1.2 showed no correlation which means the residuals are not related to the concentration of uranium. Then, the hypothesis was tested whether all the location have the same population mean of uranium concentration or not by using one-way ANOVA. The result as followed (Table 1.3).

Table 1.3: One-way ANOVA for uranium concentration

	df	SS	MS	F	Significance F
Regression	1	64861401954	64861401954	551.12	0.03
Residual	1	117689553	117689553		
Total	2	64979091507			

Based on the ANOVA’s calculation above, it can be concluded that not all the population mean of uranium concentration are same for each location since *significance F*

(0.03) $< \alpha$ (0.05). The highest and lowest concentration of uranium at certain location was only depends on the type of soil. If the soil contains properties that can make the uranium to be stayed there, thus the amount of uranium will be higher. The highest uranium concentration in soil sample was at third sampling point in Bukit Tanah Merah (345.51 µg/g) while the lowest is located at first sampling point in Lynas (65.24µg/g). The limit of detection value was 1.68 ppb whereas limit of quantification was 21.05 ppb. The following data was the concentration of uranium-238 at 10 sampling plot.

Table 1.4: The variation in concentration of uranium-238 at different locations

Sampling Point	Sampling locations	Intensity (y-axis)	X (^{238}U concentration in soil/ppb)	X (x50 mL)
1	Lynas	34241	1.30	65.23
2	Taman Balok Makmur	76142.4	2.90	145.07
3	Bukit Tanah Merah	181343.7	6.91	345.51
4	Within Kem Bina Negara and Padang Hangus	39815.4	1.52	75.86
5	Kampung Gebeng	44549.4	1.70	84.88
6	Kampung Baging	81612.5	3.11	155.49
7	Perkampungan Batu Hitam	38592.1	1.47	73.53
8	Kampung Seri Pelindung	66695.3	2.54	127.07
9	Teluk Cempedak	107402.2	4.09	204.63
10	Pusat Pembangunan Kemahiran	60769	2.32	115.78
Σ		731163	27.86	1393.06
Mean		73116.3	2.79	139.31
σ			1.69	84.71

To study the radiation effect by potassium-40, graph of absorbance versus concentration of potassium was plotted shown in Figure 1.5 to determine the relationship between these two variables.

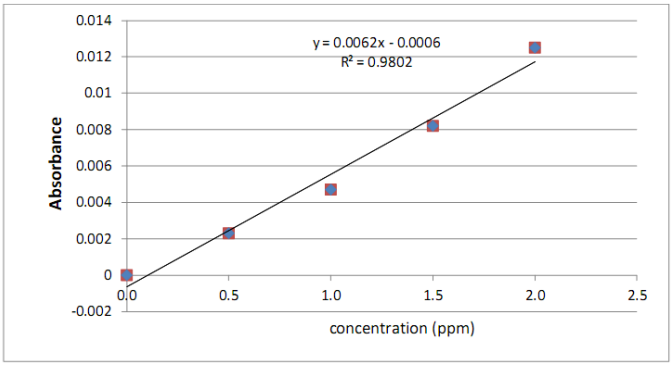

Figure 1.5: The calibration curve for potassium

The graph in Figure 1.3 shows a straight line with the value of correlation coefficient, r = 0.9900. We can conclude that, there is a strong positive linear correlation between concentration of potassium and absorbance. That's mean; the potassium concentration was proportional will be increase when the absorbance value is high. We decide to test whether our hypothesis that all the type of sample have the same population mean of concentrations or not. One-way ANOVA method has been chosen to solve this problem.

Table 1.5: One-way ANOVA table for potassium concentration

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
Between Groups	0.469981	9	0.05222	179.0402	4.03E-17	2.392814
Within Groups	0.005833	20	0.000292			
Total	0.475814	29				

By referring Table 1.5 above, since the *P-value* (4.03×10^{-17}) < α (0.05), then the hypothesis has been rejected. It's clear to conclude that not all the population mean of potassium concentration are the same for each location. The highest concentration of potassium was located at Lynas which is 1.146 mg/L because around this area, there are a lot of factories and Lynas is one of the industrial areas. The different type of soil affected the concentration of potassium-40. Two type of sample soil were collected which are clay type and sandy. The clay type of soil consist high concentration compared to sandy.

IV. CONCLUSION

Kuantan district terrestrial background radiation baseline data has been developed based on ten different locations selected around Kuantan. The highest reading of radiation recorded from this study is 0.188 Sv/hr and the lowest reading recorded is 0.098 Sv/hr. Further research, knowledge and information must be explored about the quantification of the potassium-40 or other alkaline metals in monitoring the quantification of matters that may harmful. According to the analysis of all 10 types of soil samples, almost all soil sample shows concentration of uranium-238 below this detection limit. The result of analysis is below the detection limit is due to contamination either from the wall of vessel or from the air.

V. ACKNOWLEDGEMENTS

Thanks to Research & Inovation Unit, Universiti Malaysia Pahang, Malaysia for the supported grant provided numbered RDU110365 and to the Central Laboratory Department, Universiti Malaysia Pahang.

REFERENCES

[1] Ramli, A. T., Wahab, A. Hussein, M. A. & Wood, A. K. 2005. Environmental 238U and 232Th concentration measurements in an area of high level natural background radiation at Palong, Johor, Malaysia. *Journal of Environmental Radioactivity, 80*, 287–304.

[2] Shenber, M. A. 1997. Measurement of Natural Radioactivity Levels in soil in Tripoli. *Appl. Radiat. Isot., 48*, 147-148.

[3] Rozmaric, M., Ivsic, A. G. & Grahek, Z. 2009. Determination of uranium and thorium in complex samples using chromatographic separation, ICP-MS and spectrophotometric detection. *Talanta, 80*, 352–362.

[4] Directorate of National Mapping, Malaysia (2004).

[5] Avivar, J., Ferrer, L., Casas, M. and Cerd, V. 2012. Fully automated lab-on-valve-multisyringe flow injection analysis-ICP-MS system: an effective tool for fast, sensitive and selective determination of thorium and uranium at environmental levels exploiting solid phase extraction. *J. Anal. At. Spectrom., 27*, 327-334.

[6] Baykara, O. & Dogru, M. 2006. Measurements of radon and uranium concentration in water and soil samples from East Anatolian Active Fault Systems (Turkey). *Radiation Measurements, 41*, 362–367.

[7] Saat, A., Kassim, N., Hamzah, Z., Farisz, A. 2010. Determination of surface radiation dose and concentrations of uranium and thorium in soil at UITM Perhilitan Research Station Kuala Keniam, Taman Negara, Pahang. *Journal of Nuclear and Related Technologies, 7(2)*, 49-54.