Comparative study of natural radionuclides measurements and radiation dose assessment using vehicle-borne and laboratory gamma-ray spectroscopy techniques in Thailand

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Abstract. In this study, vehicle-borne and laboratory gamma-ray spectroscopy techniques were compared to estimate the activity concentration of natural radionuclides in Thailand soil. The activity concentrations of ^{238}U (^{226}Ra), ^{232}Th (^{228}Ra) and ^{40}K in soil from the 6 regions of Thailand (77 points) were simultaneously measured with in-situ portable NaI(Tl) (a 3-in × 3-in scintillation spectrometer). In parallel, soil samples collected from these sites were analyzed with the laboratory gamma-ray spectrometry technique (HPGe). A moderate correlation was observed between both in-situ and laboratory analysis. The absorbed dose rate in air due to ^{238}U, ^{232}Th and ^{40}K in soils was calculated using the Beck's Formula and the results were compared with measured values obtained by vehicle-borne at 1 m height above ground. The results of the calculated and measured dose rate show a good correlation of R^2=0.80. Moreover, ^{232}Th was found to be the main contributor to the absorbed dose rate in air at 1 m from surface soil from outdoor terrestrial radiation in the study areas with a mean value of 31%.

1. INTRODUCTION

Accidental, intentional or authorized releases from past and current human activities such as mineral exploration and mining, production of electrical energy and other applications can result in an effect of contaminated land. An ecosystem is contaminated varies from country to country, but at least is based on the estimation of the concentration of the contaminant present such as, a metal, metalloid or a specific radionuclide [1]. In general, the soil sampling has historically been used to evaluate the distributions of natural and artificial radionuclides of the fallout from nuclear accident in soil by gamma spectrometry [2-4]. In fact, the relatively large size, and high cost of soil sampling and laboratory analysis have limited the environmental soil monitoring. Moreover, detector calibration using traceable standards for different type of samples geometries and matrices and to long counting time for low levels of radionuclides are required. Therefore, many researchers have developed the in-situ gamma spectrometry measurements technique for quick and less costly radioactivity measurements [5,6]. In-situ gamma spectrometry can be easily used for determining natural and artificial radionuclides concentrations in the environment [7,8]. Likewise, it is useful in operational and emergency monitoring of nuclear facilities,

in radioactive contamination measurements and mapping, environmental and radiation safety studies [6]. Furthermore, the direct in-situ characterization of contaminated sites is an important alternative or complement to procedures involving the laboratory analysis of field samples. Direct in-situ characterization supports and improves the quality of the analytical data from a contaminated site.

In this study, two nuclear analytical techniques for determination of ^{238}U (^{226}Ra), ^{232}Th (^{228}Ra) and ^{40}K in soil, have been compared, namely, in-situ gamma ray spectrometry and laboratory gamma ray spectrometry. Measurement of these radionuclides are important for determining the public radiation dose from radionuclides existing in the soil [9]. The contributions on the terrestrial absorbed dose rate in air as gamma radiation from these primordial radionuclides are approximately 35% from ^{40}K, 25% from ^{238}U series and 40% from ^{232}Th series [10]. The global mean value of this dose according to the distribution of the population ranges from 50 to 59 nGy/h [11].

2. MATERIALS AND METHODS

2.1 Survey Area

The absorbed dose rates in air (nGy/h) from the natural radionuclides such as ^{40}K, ^{238}U series and ^{232}Th series were measured in 2017 - 2019 in the 6 regions of Thailand (The Eastern, Western, Southern, Central, Northern and Northeastern). The main part of Central Thailand is covered by Quaternary sediments which conceal a number of basins formed in response to dextral shear on the Mae Ping. The northern, western and southern peninsular regions of Thailand are dominated by Paleozoic rocks with small, deformed outliers of Mesozoic strata, and occasional small Tertiary basins. In Northeastern Thailand, an extensive outcrop of Mesozoic rocks occurs on the Khorat Plateau. The eastern region of Thailand is underlain by late Palaeozoic sedimentary or metasedimentary rocks, Triassic granites, and Quaternary sediments. Main cities and main roads with the high human population density were used to the extent possible, centered on residential areas.

2.2 Vehicle borne gamma-ray spectrometry

In this study, the car-borne technique used for in-situ measurement of natural radionuclide concentrations of ^{226}Ra, ^{228}Ra and ^{40}K, as well as the ambient dose rate. A 3-in × 3-in NaI(Tl) scintillation spectrometer (EMF-211, EMF Japan Co., Osaka, Japan) with a global positioning system was used for the present car-borne gamma spectrometry. For the normal survey, the measurements of the counts inside the car were carried out every 30-s along the route. Latitude and longitude at each measurement point were measured at the same time as the gamma-ray count rates (50 keV– 3.2 MeV) were recorded. Car speed was kept around 40 km/h. The photon peak of ^{40}K ($E\gamma$ = 1.464 MeV) and ^{208}Tl ($E\gamma$ = 2.615 MeV) was used for the gamma-ray energy calibration from the channel number and gamma-ray energy before the measurements. This survey system has been utilized for various environmental radiation research purposes in Japan. It has sufficient sensitivity and range to measure the natural radiation level as required for the measurements of environmental radiations [12].

2.3 Activity concentrations and the contribution of ^{238}U, ^{232}Th and ^{40}K to absorbed dose rate in air

In order to evaluate activity concentrations in soil, used for the evaluation of the conversion factor of absorbed dose rate in air were considered. The gamma-ray pulse height distributions (In-situ) were measure outside the car for 15 min at 77 locations (Figure 1). These observations were carried out on private land after obtaining specific permissions from the land owners and it was also confirmed that the field studies did not involve endangered or protected species. The NaI(Tl) scintillation spectrometer was positioned 1 m above the ground surface. Measured gamma-ray plus height distributions were then unfolded using a 22 × 22 response matrix for the estimation of absorbed dose rate in air. The detailed method has been reported by Minato [13]. The calculated absorbed dose rates in air using a 22 × 22 response matrix method were separated as natural radionuclides (^{40}K, ^{238}U series and ^{232}Th series). In this study, the energy bins were set to 1.39–1.54 for ^{40}K, 1.69–1.84 MeV and 2.10–2.31 MeV for ^{214}Bi (^{238}U series) and 2.51–2.72 MeV for ^{208}Tl. These energy intervals for the bins were given by Minato [13].

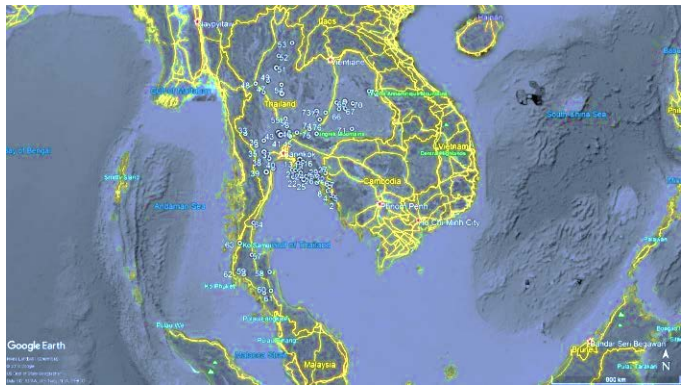


Figure 1: The soil sampling points and the measurement points for the gamma-ray pulse height distributions (In-situ).

2.4 Samples Preparation and Analysis

The surface soil samples from the 0–5-cm depth level weighing about 1 kg. were collected from the same point of the gamma-ray pulse height distributions measurement. They located in 33 provinces in the eastern, western, southern and central, northern and northeastern Thailand. Sampling sites on the selected routes were chosen relate to the center of each province as possible. 77 soil samples were oven dried at a temperature of 110 °C until constant weight. The dried samples were pulverized into a fine powder and sieved through a 250 µm mesh size sediment sieve. All homogenized samples were analyzed for activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K using gamma spectroscopy. The samples were packed into airtight plastic containers and sealed to prevent the escape of radon (^{222}Rn) and thoron (^{220}Rn) gases. Prior to measurement, the sample was stored for at least four weeks in order to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their radioactive progenies. Activity concentrations in the samples were measured using a high-

purity germanium (HPGe) detector (GEM25P4-76, ORTEC, USA) with a relative efficiency of 30 % in a low background configuration. Energy and efficiency calibrations of the detector were carried out using three different IAEA standard reference materials including IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1. Counting time for each sample was set at 24 h. By assuming secular equilibrium with their progenies in the ^{238}U and ^{232}Th decay chain, the activity concentration of ^{226}Ra was calculated using gamma-rays associated with decay products of ^{214}Bi (609.3 keV) and ^{214}Pb (351.9 keV). In the case of ^{228}Ra activity concentration, the gamma-ray lines of 911.2 keV from ^{228}Ac and 583.2 keV from ^{208}Tl were used. The activity concentration of ^{40}K was derived directly from the measured intensity of its photon peak at energy 1,460.8 keV [14].

2.5 The absorbed dose rate in air from soil sample

In secular equilibrium conditions, activity concentrations of ^{238}U and ^{226}Ra in uranium series and ^{228}Ra and ^{232}Th in thorium series remain the same. Thus, the absorbed dose rate in air can be evaluated from the activity concentrations of the ^{226}Ra, ^{228}Ra and ^{40}K measured in soil samples. If natural gamma sources uniformly distributed in the ground, the absorbed dose rate in air at 1 m above the ground can be calculated using the below equation based on the data provided by Saito and Jacob [15].

$$D_{air} = 0.462 \cdot A_U + 0.604 \cdot A_{Th} + 0.0417 \cdot A_K$$

Where; A_U, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K, respectively, in Bq/kg. The values of 0.462, 0.604 and 0.0417 are conversion factors (absorbed dose rate in air per unit activity per unit of soil mass, in units of nGy/h per Bq/kg) evaluated for ^{238}U-series, ^{232}Th-series and ^{40}K, respectively [16].

3. RESULTS AND DISCUSSION

3.1 Evaluation of natural radionuclides activity concentration

Activity concentrations of natural radionuclides of ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K in the eastern, western, southern, central, northern and north-eastern Thailand were measured using the in-situ measurement with car borne survey system and the laboratory soil-sampling-based gamma spectrometry technique as shown in Table 1 and Table 2, respectively. A comparison between the measured values of ^{238}U, ^{232}Th and ^{40}K with the two techniques of in-situ and laboratory gamma ray spectrometers are presented in Figure 2a – 2c. The mean activity concentration of ^{238}U, ^{232}Th and ^{40}K were 33 ± 1 Bq/kg, 26 ± 1 Bq/kg and 202 ± 6 Bq/kg respectively for the in-situ measurement. The mean activity concentration of ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K were 30 ± 2 Bq/kg, 44 ± 4 Bq/kg and 390 ± 8 Bq/kg respectively for the laboratory soil-sampling-based measurement. The median values of ^{238}U-series, ^{232}Th-series and ^{40}K in the earth's crust are 35, 30 and 400 Bq/kg, respectively [16]. The average value of ^{238}U, and ^{40}K from the 6 regions of Thailand measured in the soil samples is a bit lower than the worldwide median values, while as the average value of ^{232}Th is slightly higher. The high value of the activity concentration may correlate with the presence of the geological lineaments in the area (Granitic rocks,

Paleozoic rocks, Mesozoic rocks, and Tertiary rocks) [17]. Correspondingly, it is evident from the fact that thorium is higher than that of uranium in the Earth's crust [16].

Table 1: Activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq/kg) and their contributions to the absorbed dose rate in air by *in-situ* measurement.

Sampling region (no. of sample point)	Average contribution to dose rate (%)			Concentration (Bq/kg)						The outdoor absorbed gamma dose rates (nGy/h)	
				^{238}U		^{232}Th		^{40}K			
	^{238}U	^{232}Th	^{40}K	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)
The eastern (32)	36	43	21	9-77	34 (3)	4-62	29 (4)	23-3043	353 (29)	7-101	42 (7)
The western (9)	6	12	2	13-92	44 (2)	23-118	39 (3)	30-486	281 (16)	24-96	57 (14)
The southern (8)	3	9	1	15-75	41 (2)	15-79	35 (1)	36-304	170 (7)	15-90	49 (6)
The central (12)	13	13	5	10-73	31 (1)	7-59	21 (1)	27-442	194 (8)	22-70	38 (3)
The northern (3)	2	4	1	17-32	25 (1)	27-36	17 (1)	112-191	140 (8)	22-36	28 (2)
The northeastern (13)	46	41	12	16-35	23 (1)	8-21	14 (1)	15-261	71 (3)	15-33	24 (2)
Average	28	31	13	9-92	33 (1)	4-118	26 (1)	15-3043	202 (6)	7-101	40 (4)

Table 2: The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq/kg) and the outdoor absorbed gamma dose rate estimated for the soil samples.

Sampling region (no. of sample)	Concentration in soil (Bq/kg)						The outdoor absorbed gamma dose rates (nGy/h)	
	^{238}U		^{232}Th		^{40}K			
	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)
The eastern (32)	5-132	41 (4)	2-165	55 (5)	10-1270	380 (15)	6-191	68 (3)
The western (9)	13-92	31 (3)	23-118	51 (5)	47-1414	465 (20)	23-170	65 (5)
The southern (8)	11-77	40 (4)	20-126	63 (6)	30-965	448 (20)	18-124	75 (6)
The central (12)	6-54	30 (3)	1-81	45 (3)	67-1012	424 (21)	6-116	59 (3)
The northern (3)	17-32	25 (3)	27-36	28 (3)	248-534	465 (21)	36-46	48 (2)
The northeastern (13)	7-25	15 (3)	11-37	19 (2)	45-716	159 (9)	15-62	25 (1)
Average	5-132	30 (2)	1-165	44 (4)	10-1414	390 (8)	6-191	56 (2)
Worldwide median	16-110	35	11-64	30	140-850	400		51

3.2 Comparison between in-situ and laboratory activity concentration of ^{238}U, ^{232}Th and ^{40}K

Comparative analyses were done between *in-situ* and laboratory measurements. The ratio between the two measurements were 1.10, 0.59 and 0.52 for ^{238}U, ^{232}Th and ^{40}K respectively as shown in Table 3. The laboratory results obtained in this work were greater than the *in-situ* results for ^{232}Th and ^{40}K. The average % difference of 10%, 51% and 64% was obtained for ^{238}U, ^{232}Th and ^{40}K respectively. Moreover, a moderate correlation between these techniques can be observed in Figure 2a - 2c with linear regression lines of 70% for ^{228}U and ^{232}Th and 76% for ^{40}K. The deviation recorded between the *in-situ* and laboratory measurements in this work maybe due to the fact that the samples collected for the laboratory analysis only accounted for a very small volume of surface soil (0-5 cm.) as against the *in-situ* measurements with gamma contributions from the top 30 cm layer soil [18]and surface area up to hundreds of m^3 [19]. Moreover, the water content, and soil density affect the attenuation of gamma within the soil. In regards to the water content of the soil, attenuation coefficients show a difference of less than 3% for soil with water contents of 0% and 25% [20]. Therefore, high moisture in soil would underestimate the specific activity of radionuclides in the soil by *in-situ* measurement. On the other hand, Corbacho and Baeza[22] reported that there was an evidence of a slight increase of natural

radionuclides in the top soil layer (0-5 cm) at the areas nearby the ash pond due to the deposition of fly-ash whereas a depth soil layer was a homogeneous distribution of natural radionuclides. Likewise, the use of fertilizers on agricultural lands may lead to an increase of natural radioactivity in farm soil [21].

Table 3: In-situ and laboratory measurements of average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq/kg) and their respective ratios for comparison

In-situ measurements (Bq/kg) (S.D.)			Laboratory measurements (Bq/kg) (S.D.)			Ratio (In situ/Laboratory)		
^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K
33 (1)	26 (2)	202 (6)	30 (2)	44 (4)	390 (8)	1.10	0.59	0.52

3.3 Comparison between in situ and laboratory outdoor absorbed dose rate in air

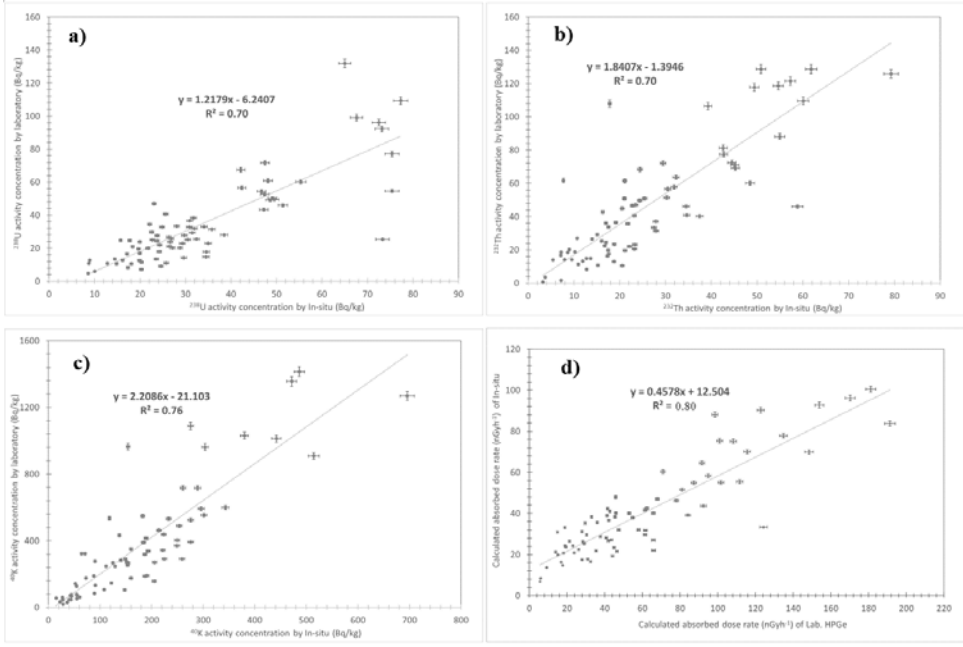
The results of the calculated and measured dose rate show a good correlation of R^2=0.80 (Figure 2d). The average absorbed dose rates in air estimated for the soil samples varied from 25 to 68 nGy/h with a mean of 56 ± 2 nGy/h. The average absorbed dose rates in air from direct measurement using *in-situ* technique varied from 24 to 57 nGy/h with a mean of 40 ± 4 nGy/h. The average percent difference was 33%. Additionally, the uncertainty in the absorbed dose rate in air using the laboratory gamma spectrometry was about 4% while as the uncertainty using the *in-situ* gamma ray was about 10%. The *in-situ* measurement uncertainty was 2.5 times higher than laboratory gamma spectrometry. This may cause by the non-uniform distribution of radioactivity in soil, the largest source of *in-situ* uncertainty was likely to be because a uniform reference calibration does not accurately represent the true efficiency. Moreover, the ratio of the average absorbed dose rate in air inferred from activity concentrations of radionuclides in soil to the average absorbed dose rate in air from *in-situ* measurement using car borne survey system in this study is 1.5. A factor of 1.5 is likely due to shielding of the soil moisture and the climate condition during the measurement time. The difference between the two methods could be further explained that the *in-situ* gamma spectrometry gives representative source concentration in the horizontal plane. In practice, an infinite half space can be taken as a large area of open ground with a radius up to 70 m where there are few obstructions [12] whereas the gamma spectrometry in laboratory measures radioactivity in soil samples collected from the top 5 cm of a 1 m^2 area providing a dry mass of approximately 200 g. It is important to acknowledge this measurement error when employing the two methods.

4. CONCLUSIONS

The use of a vehicle-borne measurement system is an effective practical way to quickly obtain database on the radiation background levels in the 6 regions of Thailand. The average absorbed gamma dose rate from vehicle-borne survey at 1 m above the ground surface, in Thailand was 40±2 nGy/h. The highest average absorbed dose rates in air at 1 m above the ground surface from vehicle-borne survey were observed at the eastern region of Thailand. Moreover, the average values of activity concentrations of ^{238}U, ^{232}Th and ^{40}K in the soil samples collected from these regions are 30 ± 1 Bq/kg, 44 ± 2 Bq/kg and 390 ±

8 Bq/kg, respectively. In addition, the values of activity concentration of ^{232}Th seem to exceed the corresponding world median values.

Figure 2: The relation of measured activity concentration of a) ^{238}U, b) ^{232}Th, c) ^{40}K and d) the absorbed dose rate in air with the two techniques of in-situ and laboratory gamma ray spectrometers



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