Natural radioactivity in the soil of Thoulakhom district in Vientiane province, Laos

Sonexay Xayheungsy, Le Hong Khiem

Abstract—Results of the first investigation of the activity concentration of the surface soil samples collected at various locations of Thoulakhom district of Vientiane province of Laos People's Democratic Republic (PDR) are presented in this work. The activity concentration of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the soil samples were determined by gamma spectrometer using a high-energy resolution semiconductor detector HPGe. The activity concentrations of the natural radionuclides were in the range from 11.28 to 31.46 with the mean of 21.76 Bq.kg^{-1} for ^{226}Ra, from 7.13 to 44.47 with the mean of 21.85 Bq.kg^{-1} for ^{232}Th and from 8.96 to 581.52 with the mean of 112.89 Bq.kg^{-1} for ^{40}K. These mean values of the activity concentration were lower than the average worldwide ones, which were 33, 45 and 420 Bq.kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The results indicate dthat the radiation hazard from natural ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in all investigated soil samples taken from area under investigation in this work was not significant.

Index Terms—gamma ray spectrometry, natural radionuclide, activity concentration, radiation hazard indices.

1 INTRODUCTION

The natural radionuclides are present everywhere with different concentrations. They were created together with the creation of the earth. Among the natural radionuclides, ^{238}U, ^{232}Th, and ^{40}K are most significant. The isotopes ^{238}U and ^{232}Th are radioactive and they undergo radioactive decay into many other radioactive isotopes until they are stable. As radium and its

daughter products produce 98.5 % of the radiological effects of the uranium series, the contribution from the ^{238}U is used to be replaced with the decay product ^{226}Ra.

The natural radionuclides emit gamma radiation which continuously affect human health. Therefore, the investigation of their concentrations in the living environments of human beings is an important task in order to estimate the radiation hazard from these natural radionuclides to human.

It is possible to say that the earth's crust including soil is one of the environment having the largest presence of the natural radionuclides. The natural radionuclides in the soil not only affect the health of the people living beneath it but also in building materials which are made from it. Therefore, in order to limit the risks caused by the natural radioactive isotopes in the earth, it is necessary to measure the activity concentration of the radionuclides in it. Due to the importance of this issue, the survey of the activity concentration of natural radionuclides in the surface soil has been attracted the interest of many researchers in the world [1-4]. In Lao PDR, this issue so far has not been interested.

In this work, we present the first results of measuring the activity concentration of the natural radionuclides of the soil samples taken in Thoulakhom district of the capital Vientiane of Lao PDR by using high-resolution gamma-ray spectroscopy. It is necessary to stress that this is the first ever investigation of the natural radioactivity in the soil of Lao PDR. Such kind of investigation will be carried out continuously in the future in different provinces of Lao PDR in order to provide the data on natural radioactivity of soil in these regions. These data will help the evaluation the background exposure levels in the

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investigated areas. Furthermore, they will help the authorities for better planning land use as well as to reduce the level of influence of natural radioactivity to people health of Lao PDR.

2 EXPERIMENTAL METHODS

Sample collection and preparation

In fig. 1, the locations for collecting the soil samples in Thoulakhom district of Vientiane province of Lao PDR are presented. The total number of the locations was 10 and they were indicated by the circles with the dots on the map. They are marked as P1, P2...P10. A soil auger had been used for obtaining the soil samples at a depth of about 0.6 meters from the surface layer at the designed locations. At each location, about 5 kg wet weight per sample had been collected and they were sent to the laboratory at the Center

for Nuclear physics of the Institute of physics of Vietnam Academy of Science and Technology for measurement of the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides.

All measurements carried in our laboratory were done with 0.2 mm sieved soil. For this purpose, firstly, soil sample moisture was removed by drying at a temperature of 100 ºC for about 24 hours in an oven. After removing moisture, the samples were crushed and sieved by using a sieve to pass a mesh of 0.2 mm. The homogenised samples were filled into the cylindrical plastic beakers with the diameter of 10 cm, which were then hermetically sealed with the aid of PVC tape to prevent the escape of airborne ^{222}Rn and ^{220}Rn from the samples. The samples were weighed and stored for more than 30 days for reaching secular equilibrium between ^{226}Ra and its short-lived daughters.
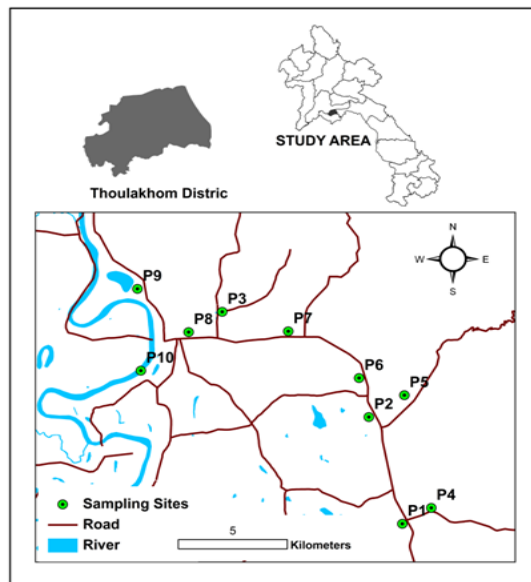

Fig. 1. The map of Lao PDR showing the Thoulakhom district and the soil sampling locations were indicated ass P1, P2… P10.

Measurement of the activity concentration of the soil samples

A gamma-ray spectrometer was used for measuring the activity concentration of the soil samples. The gamma-ray spectrometer is connected with a coaxial cylinder HPGe detector model GEM20P4-70 made by Ortec company. The relative detection efficiency of the detector is

about 20% relative to a 3" x 3" NaI(Tl) scintillation detector. The energy resolution of the detector at 1.332 MeV was 1.8 keV. The electronic system of the spectrometer contained a detector high voltage power supply and signal processing modules. The latter included a main spectroscopy amplifier model 572A made by Ortec company and a computer based PCA-MR 8192 ACCUSPEC multi-channel analyzer. For

data acquisition, storage, display, and on-line analysis of the acquired gamma-spectra, an advanced multi-channel analyzer emulation software MAESTRO-32 has been used. For spectral off-line analysis, the software, GENIE-2K software had been used.

In order to reduce the number of background gamma radiation present at the laboratory site, the HPGe detector was placed in a low-level Canberra Model 747 lead shield having a lead thickness of 10 cm. It prevented prevent high background counts due to external sources, thus reducing counting times and improving the lower limit of detection. The 1 mm tin and 1.6 mm copper graded liner prevented interference by x-rays from lead. Energy calibration of the detector was carried out by using two different sources ^{60}Co and [226]Ra, which emitted gamma-rays of energy ranged between 186.21 keV and 2447.86 keV.

The activity concentration of [40]K was determined directly by its own gamma-ray at 1460.8 keV (10.7%), while the specific activities of [226]Ra and [232]Th were calculated based on the weighted mean values of their respective decay products in equilibrium. The specific radioactivity of [226]Ra was determined using the 295.22 keV (18.5 %), 351.93 keV (35.6 %) gamma-rays from [214]Pb and 609.31 keV (45.49 %), 768.36 keV (4.89 %), 1120.14 keV (15.0 %), 1764.43 keV (15.28 %) from [214]Bi. The activity concentration of [232]Th was determined using the 583.187 keV (85.0 %), the 2614.511 keV (99.79 %) from [208]Tl and 911.12 keV (25.8 %) from [228]Ac. The value written inside the parentheses following gamma-ray energy indicated the absolute emission probability of the gamma decay. In order to obtain a good statistics, gamma spectra of the soil samples was measured in long enough time. In our case, each sample was measured for about 72000 seconds. Measurements with an empty sample container under the same measuring condition of the sample containers filled with soil were also carried out to determine the ambient background in the laboratory site.

The activity concentrations in each soil sample were determined by the relative method using the standard samples made from the IAEA-RGU-1,

IAEA-RGTh-1 and IAEA-RGK-1 reference materials [5]. These materials were obtained from the International Atomic Energy Agency (IAEA), for which the activity concentration of the interested radionuclides were known. The densities of the reference and investigated soil samples were similar. Furthermore, the geometry of the containers of the soil samples was identical to that of the reference materials (IAEA-RGU-1, IAEA-RGTh-1, and IAEA-RGK-1). By applying the relative method for the activity concentration determination, many corrections could be avoided.

3 RESULTS AND DISCUSSION

Calculation of the activity concentrations

The following equation had been used for calculating the activity concentration of [40]K, [226]Ra and [232]Th radionuclides in the soil samples:

$$A_m = \frac{C_m}{C_s} \times \frac{M_S}{M_m} \times A_s \times \frac{1-e^{-0.693t_m/T_{1/2,i}}}{1-e^{-0.693t_s/T_{1/2i}}} \tag{1}$$

where:

A_m was the activity concentration of radionuclide in the soil sample given in Bq.kg[-1];

A_s was the activity concentration of radionuclide in the standard given in Bq.kg[-1];

C_m was the count rate (counts/second) obtained under the corresponding peak of the soil sample;

C_s was the count rate (counts/second) obtained under the corresponding peak of standard sample;

M_s was mass of the standard sample in kg;

M_m was mass of the soil sample in kg;

t_m was the measuring live time for the soil sample in second;

The mean activity concentration in Bq.kg[-1] of the natural radionuclides of [226]Ra, [232]Th, and [40]K measured in soil samples obtained from 10 locations of the Thoulakhom district of Vientiane province were listed in Table 1. In the last row of table 1, the average worldwide activity concentrations of [226]Ra, [232]Th, and [40]K radionuclides were listed for comparison. In this table, the activity concentrations of the natural

radionuclides in Bq.kg^{-1} range from 11.28±0.90 to 31.46±1.19 for ^{226}Ra, from 7.13±1.04 to 44.47±1.70 for ^{232}Th and from 8.96±2.60 to 581.52±8.09 for ^{40}K.

The mean activity concentrations were 23.14 Bq.kg^{-1} for ^{226}Ra, 27.13 Bq.kg^{-1} for ^{232}Th and 226.26 Bq.kg^{-1} for ^{40}K. These average values were within those reported from literature in other countries in the world and were presented in UNSCEAR 2000 report [7] which were listed in the last row of the table, namely 33 Bq.kg^{-1} for ^{226}Ra, 45 Bq.kg^{-1} for ^{232}Th and 420 Bq.kg^{-1} for ^{40}K. However, the activity concentrations of ^{40}K of Ban Nam Ang (581.52±8.09 Bq.kg^{-1}) and Ban Keun (468.59±7.60 Bq.kg^{-1}) were a bit higher than the worldwide average value (420 Bq.kg^{-1}).

Table 1. Mean activity concentrations of 10 soil samples collected from the different locations of the Thoulakhom district of Vientiane province, Lao PDR

Sample location	Location name	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	^{40}K (Bq.kg^{-1})
1P	Ban Dong	11.28±0.90	7.43±1.05	40.52±3.88
2P	Ban PhaThao	25.94±1.13	29.56±1.52	137.13±5.39
3P	Ban Nam Ang	30.06±1.17	44.47±1.70	581.52±8.09
4P	Ban Nanokkhoum	20.43±1.06	14.47±1.25	81.38±4.68
5P	Ban Phonmouang	15.73±0.99	15.10±1.27	68.63±4.47
6P	Ban NaKang	13.25±0.95	7.13±1.04	8.96±2.60
7P	Ban Naxanglek	29.01±1.16	37.77±1.62	88.31±4.78
8P	Ban Keun	31.46±1.19	44.42±1.70	468.59±7.60
9P	Ban Hatnoi	28.61±1.16	39.58±1.64	415.23±7.34
10P	Ban Boungphao	25.62±1.13	31.39±1.54	372.28±7.12
Total		21.76±0.34	21.85±0.43	112.89±1.48
Worldwide Mean [7]		33	45	420

Estimation of radiation hazard indices

By using the obtained activity concentrations of ^{226}Ra and ^{232}Th and ^{40}K radionuclides of the investigated soil samples, the radiological hazard indices of all samples were also calculated including absorbed dose rates (D), radium equivalent activity (Ra$_{eq}$), external hazard index (H$_{ex}$), internal hazard index (H$_{in}$) and representative level index (I$_\gamma$). Table 2 presented the average values of absorbed dose rate, radium equivalent activity, external hazard index, internal hazard index and gamma-index of all investigated soil samples.

The absorbed dose rate D in the air at 1 m above the ground surface due to the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K could be calculated using the formula given below [6]:

$$D(nGy.h^{-1}) = 0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_K \quad (2)$$

Where D was the absorbed dose rate in nGy.h^1 and A$_{Ra}$, A$_{Th}$ and A$_K$ are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq.kg^{-1} of the soil samples. It should be mentioned that for calculating the absorbed dose rate D by using above equation, the conversion factor recommended by UNSCEAR was adopted namely, 0.0417 nGy.h^{-1} for ^{40}K, 0.462 nGy.h^{-1} for ^{226}Ra and 0.604 nGy.h^{-1} for ^{232}Th [7].

From table 2, the average values of calculated absorbed dose rates in the samples under investigation were ranged between 10.80±0.77 to 65.00±1.21 nGy.h^{-1} with the mean of 30.26±0.31 nGy.h^{-1} and found to be comparable to the worldwide average of 55 nGy.h^{-1}. However, the absorbed dose rates in Ban Nam Ang and Ban Keun were a bit higher than this value.

The radium equivalent activity Ra$_{eq}$

Radium equivalent activity was an index that had been introduced to compare the activity concentration of materials containing different amounts of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides and to represent their activity concentrations by a single quantity, which took into account the radiation hazards associated with them. It was calculated as the weighted sum of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations, based on the assumption that 10 Bq.kg^{-1} of ^{226}Ra, 7 Bq.kg^{-1} of ^{232}Th and 130 Bq.kg^{-1} of ^{40}K produce the same gamma dose rate. The formula to calculate was as follows:

$$Ra_{eq}(Bq.kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (3)$$

where A$_{Ra}$, A$_{Th}$, and A$_K$ were the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. The values of the radium equivalent activity of all soil samples as listed in Table 2 showed that it ranged from 24.13±1.78 to 138.43±2.77 Bq.kg^{-1} with the mean of 66.05±0.72 Bq.kg^{-1}. The recommended safety limit for this quantity was 370 Bq·kg^{-1} [7].

External hazard index H$_{ex}$ and internal hazard index H$_{in}$

Natural occurring radionuclides in terrestrial soils produced an external radiation field. Human beings were exposed to this radiation field continuously. The external hazard index (H$_{ex}$) and internal hazard index (H$_{in}$) were used as radiological indicators to estimate the radiological implications of the natural radionuclides in the surface soil on the people who were living on it. The external hazard index and the internal hazard index were calculated by the following formulae:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \quad (4)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \quad (5)$$

Where, A$_{Ra}$, A$_{Th}$, and A$_K$ were the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively. The value of these hazard indices mould be less than unity to the radiation hazard insignificant i.e.; the area was safe to the human for living. According to our calculated values of these indices listed in table 2, the values of the external hazard index ranged from 0.07 to 0.37 Bq.kg^{-1} with the mean of 0.20±0.12 while the values of internal hazard index ranged from 0.10 to 0..46 with the mean of 0.26±0.13 Bq.kg^{-1}. All of these values were lower than unity.

Representative level index I$_\gamma$

Representative level index (I$_\gamma$) was used to estimate the level of gamma radiation associated with different concentrations of some specific radionuclides. According to [8], it was defined as follows:

$$I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500} \quad (6)$$

It should be noted that the safety value for this index was less than unity. The values of the representative level index presented in table 2 were between 0.17 and 1.03 with the mean of 0.52±0.32. All the values were less than unity except the value of the sample taken in Ban Nam Ang was a bit higher (1.03 Bq.kg^{-1}).

Table 2. The average values of absorbed dose rate, radium equivalent activity, external hazard index, internal hazard index and gamma-index of all investigated soil samples

Sample location	Location name	Absorbed dose rate (nGy.h^{-1})	Ra$_{eq}$ (Bq.kg^{-1})	H$_{ex}$	H$_{in}$	I$_\gamma$
1P	Ban Dong	11.39±0.78	25.02±1.78	0.07	0.1	0.18
2P	Ban PhaThao	35.56±1.21	78.78±2.48	0.21	0.28	0.56
3P	Ban Nam Ang	65.00±1.88	138.43±2.77	0.37	0.46	1.03
4P	Ban Nanokkhoum	21.57±0.92	47.39±2.11	0.13	0.18	0.34

Sample location	Location name	Absorbed dose rate (nGy.h^{-1})	Ra$_{eq}$ (Bq.kg^{-1})	H$_{ex}$	H$_{in}$	Iγ
5P	Ban Phonmouang	19.25±0.91	42.60±2.10	0.12	0.16	0.3
6P	Ban NaKang	10.08±0.77	24.13±1.78	0.07	0.1	0.17
7P	Ban Naxanglek	39.90±1.13	89.82±2.62	0.24	0.32	0.63
8P	Ban Keun	60.91±1.21	131.07±2.77	0.35	0.44	0.97
9P	Ban Hatnoi	54.44±1.17	117.18±2.68	0.32	0.39	0.86
10P	Ban Boungphao	46.32±1.11	99.17±2.54	0.27	0.34	0.73
Mean±SD		30.26±20.14	66.05±42.78	0.20±0.12	0.26±0.13	0.52±0.32
Worldwide recommended value		55	<370	<1	<1	<1

4 CONCLUSION

This is the first ever investigation of natural radioactivity of soil in Lao PDR. In this study, the activity concentrations of natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples taken from 10 locations in Thoulakhom District in Vientiane province of Lao PDR were investigated by the help of the high resolution gamma-ray spectrometer using HPGe detector. The activity concentrations of the natural radioactive isotopes were in the range from 11.28 Bq.kg^{-1} to 331.46 Bq.kg^{-1} with the mean of 21.76 Bq.kg^{-1} for ^{226}Ra, from 7.13 Bq.kg^{-1} to 44.47 Bq.kg^{-1} with the mean of 21.85 Bq.kg^{-1} for ^{232}Th and from 8.96 Bq.kg^{-1} to 581.52 Bq.kg^{-1} with the mean of 112.89 Bq.kg^{-1} for ^{40}K. These values were within the worldwide range. The obtained activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were then used to deduce the values of different radiation hazard quantities such as absorbed dose rate in air (D), radium equivalent activity (Ra$_{eq}$), external hazard index (H$_{ex}$), internal hazard index (H$_{in}$) and representative level index (I$_\gamma$). Most of the values of these quantities were found to be less than the worldwide mean values. It meant that the area under investigation was safe to the human for living. In some locations, the absorbed dose rate in air (D) and the radium equivalent activity were found to be a bit higher. Further investigation will be carried out in other areas of Lao PDR.

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Phóng xạ tự nhiên trong đất tại huyện Thoulakhom của tỉnh Vientian, Lào

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Tóm tắt—Bài báo này trình bày các kết quả khảo sát lần đầu về hoạt độ phóng xạ riêng của các mẫu đất lớp bề mặt được lấy tại các vị trí khác nhau tại huyện Thoulakhom của tính Vientiane của Cộng hòa Dân chủ Nhân dân Lào. Hoạt độ phóng xạ riêng của các đồng vị phóng xạ tự nhiên ^{226}Ra, ^{232}Th và ^{40}K trong các mẫu đất được xác định bằng phổ kế gamma sử dụng detector bán dẫn siêu tinh khiết HPGe với độ phân giải năng lượng tốt. Hoạt độ phóng xạ riêng của các đồng vị phóng xạ tự nhiên tương ứng có giá trị trong khoảng từ 11,28 đến 31,46 với trung bình 21,76 Bq.kg^{-1} đối với của ^{226}Ra, từ 7,13 đến 44,47 với trung bình 21,85 Bq.kg^{-1} đối với ^{232}Th và từ 8,96 đến 581,52 với trung bình 112,89 Bq.kg^{-1} với ^{40}K. Các kết quả cho thấy phóng xạ tự nhiên ^{226}Ra, ^{232}Th và ^{40}K trong các mẫu đất lấy tại các vị trí khảo sát không gây ra nguy hiểm về phương diện phóng xạ.

Từ khóa—phổ kế gamma, đồng vị phóng xạ tự nhiên, hoạt độ riêng, các chỉ số liều lượng bức xạ