Natural radioactivity and radiological hazards in soil samples in Savannakhet province, Laos

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Abstract

This paper presents the evaluation of natural radioactivity and radiological hazards of the terrestrial naturally occurring radionuclides of ²²⁶Ra, ²³²Th and ⁴⁰K in Savannakhet province, Laos. The activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K are in the range of 6.6–73.6, 3.8–113.8 and 13.6–906.4 Bq kg⁻¹, with the average values of 22.4 \pm 2.1, 30.8 \pm 2.9 and 211.6 ± 16.5 Bq kg⁻¹, respectively. The average radium equivalent activity Ra_{eq} is calculated as 82.8 \pm 9.7 Bq kg⁻¹, which is smaller than the safety limit of 370 Bq kg⁻¹. Radiological hazard indices have also been evaluated in comparison with the world average values.

Keywords Natural radioactivity · Activity concentration · Radiological hazard · Soil sample

Introduction

Two main sources of the exposure of general public to natural radiation are cosmic ray and radioactive nuclides existing in the earth's crust $[1, 2]$ $[1, 2]$ $[1, 2]$. The average dose rate of cosmic ray at sea level is about 30 nGy h^{-1} , while the world average dose rate of terrestrial natural occurring radionuclides is about 59 nGy $h^{-1}[1, 3]$ $h^{-1}[1, 3]$ $h^{-1}[1, 3]$ $h^{-1}[1, 3]$. Natural radioactivity in soils comes mainly from radionuclides in the decay series of uranium (238) , thorium (232) Th) and potassium (40) K). The levels of natural radioactivity are also dependent on the geological and geographical structure of soils $[1, 2]$ $[1, 2]$ $[1, 2]$ $[1, 2]$. ²²⁶Ra subseries contributes about 98.5% of the total external gamma dose

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induced by the whole 238 U decay series, and therefore, it is usually referred to as 226Ra series instead of 238U series [\[4](#page-12-3)]. As reported in UNSCEAR report [[1\]](#page-12-0), the world average radionuclide concentrations in soils are 35 Bq kg⁻¹ for ²²⁶Ra, 30 Bq kg−¹ for 232Th and 400 Bq kg−¹ for 40K, respectively [[1\]](#page-12-0). Although the world average values of natural radionuclides in soils are low, the variation between different locations could be up to 1000 Bq kg⁻¹ for ²³⁸U, 360 Bq kg⁻¹ for ^{[2](#page-12-1)32}Th and 3200 Bq kg⁻¹ for ⁴⁰K [2]. Several worldwide regions with higher background radiation were notifed in China, Iran, India, Italy, France, Switzerland, Australia and Brazil [[1\]](#page-12-0). Therefore, the data of radioactivity concentrations in a specifc area should be connected with its population distribution to evaluate the health effect to human livings.

Majority of the external gamma dose rate above typical soils (95%) arises from primordial radionuclides incorporated in soils [[1](#page-12-0)]. The soil layer upper 30 cm contributes predominantly to the natural terrestrial radiation exposure [[5\]](#page-12-4). Soils are also the sources of spreading radionuclides to water, air, sediments and biological systems. Thus, soils are important matrices for evaluating the radiological exposure of the humans and biota, and examining the environmental radiological contamination. It means that measurement of natural radioactivity in soils is necessary to determine the change of the natural background activity with time in case of radioactive release, which is essential for environmental protection [[6](#page-12-5)]. There has been increasing interest in

mapping the natural radioactivity concentrations and radium equivalent activity in soils and establishing baseline data in many countries [\[4](#page-12-3), [6–](#page-12-5)[15\]](#page-12-6). Consequently, radiological hazard parameters can also be evaluated based on the radioactivity concentrations of natural occurring radioactive materials $[1-3]$ $[1-3]$. Therefore, these efforts are considerably important for assessing the public dose rates and the performance of epidemiological studies.

Assessment of the natural radionuclide concentrations in soil samples in Laos for establishing a baseline data is of high important. Several efforts have been made to survey and evaluate the terrestrial natural occurring radioactivity in soils and building materials in Laos. Leuangtakoun et al. [[16\]](#page-12-7) assessed the natural radioactivity in surface soils in Bolikhamxay province, Laos. The natural radioactivity and radiological hazards in building materials in Laos were investigated in Ref. [[17,](#page-12-8) [18\]](#page-12-9).

The present work aims at evaluating the natural radionuclide concentrations of ²³²Th, ²²⁶Ra and ⁴⁰K in soil samples collected widely in Savannakhet province, Laos for establishing a baseline data in this region. The results were then used to analyze radiological hazard indices such as absorbed gamma dose rate in air (*D*), annual effective dose equivalent ($AEDE$), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and internal hazard index (H_{in}) . The radioactivity concentrations of several radionuclides and radiological hazard indices in Savannakhet province have also been evaluated in comparison with neighboring and worldwide regions.

Materials and methods

Sampling area

Laos is a landlocked Southeast Asian country lying between latitude from 14.117◦ to 23.684◦ N and longitude from 100.413° to 108.832° E. It shares the borders with Myanmar and China to the northwest, Vietnam to the east, Cambodia to the southwest, and Thailand to the west and southwest.

Fig. 1 Map of Savannakhet Province, Laos and the locations of soil samples

Laos has an abundance of natural resources and environmental riches with forest covering half of the country. The climate is tropical and affected by the monsoon pattern. Savannakhet is the largest province located in the southern part of Laos with the area of $21,774 \text{ km}^2$ and the population of 970,000. The province lies on the latitude of 16° 33′ 54.18″ N and the longitude of 104° 45′ 9.83″ E as shown in Fig. [1.](#page-1-0) It shares the borders with Khammuane province to the north, Quang Tri and Thua Thien-Hue provinces of Vietnam to the east, Salavan province to the south, and Nakhon Phanom and Mukdahan provinces of Thailand to the west. The capital of Savannakhet province, also known as Kaysone Phomvihane or Muang Khanthabouly, is one of the two notable cities of Laos. Savannakhet province is administratively divided into 15 districts as displayed in Fig. [1.](#page-1-0) Sepone district is the larg‑ est mining location of copper and gold, and the most significant mining interest in Laos. Other mining locations include Vilabuly and Champhone districts. The province is also one

of the main tobacco producing areas, and is an important trading post between Thailand and Vietnam.

Sample collection and preparation

Soil samples at 80 locations distributed widely in Savan– nakhet province, Laos were collected during November and December, 2018. This time period was also the dry season in Laos with the outdoor temperature of about 30–40 ◦C. The sample locations are close to populated agriculture felds and tourist areas. The sampling locations are denoted as S1–S80 as depicted in Fig. [1.](#page-1-0) At the sampling sites, soil samples were collected from the surface layer with the depth of about $5-30$ cm $[10, 11, 19]$ $[10, 11, 19]$ $[10, 11, 19]$ $[10, 11, 19]$ $[10, 11, 19]$ $[10, 11, 19]$. Five topsoil samples were collected at the four corners and the center of a square with the side of 60 cm. The soils were then mixed up, and the amount of 1–2 kg was taken using a quartile method. After removing organic materials and pieces of stone, the samples

Fig. 2 Average activity concentrations of ²²⁶Ra in soil samples in 15 districts of Savannakhet province, Laos

were air dried at room temperature for about 24–48 h at laboratory. Then, the samples were dried in an electric oven at the temperature of 110 ◦C for about 6 h. In other related works, samples were dried for about $10-12$ h at the tem-perature of 100–[11](#page-12-11)0 ℃ to obtain constant masses [11, [20](#page-12-13)]. In the present work with relatively dried soil samples, the drying duration of about 6 h is considerably adequate. The samples were crushed and served with a mesh having holes with the diameter of 0.2 mm. The homogenized samples were weighted and placed in a cylindrical polyethylene box having the diameter of 7.5 cm and the height of 3.0 cm. The samples were stored in a period of four weeks for attaining secular equilibrium between ²²⁶Ra with ²¹⁴Bi and ²¹⁴Pb.

Analysis method

The soil samples were measured using a low background gamma spectroscopy of ORTEC P-type coaxial high purity

Germanium (HPGe). The gamma spectroscopy was calibrated using the IAEA RGU–1, RGTh–1 and RGK–1 reference materials to construct the detector efficiency curve as a function of gamma energy [\[21\]](#page-12-14). The detector was then used to measure the IAEA–375 soil reference material. The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were obtained with the deviation less than 3% compared to the reported values. Each soil sample was measured during a period of 60,000–86,400 s to ensure that the 1σ statistic errors of important photopeaks are less than 5%. Similar measurement duration of 60,000 was also applied in Ref. [[6](#page-12-5)]. In particular, to evaluate the activity concentration of ^{226}Ra , it is determined based on the photopeaks of 295.57 keV and 351.9 keV emitted from $2^{14}Pb$ and the photopeaks of 609.3 keV and 1120.3 keV emitted from 214 Bi. The activity concentration of 232 Th was determined based on the photopeaks of 338.6 keV and 911.1 keV of ²²⁸Ac and the peak of 583.19 keV of 208Tl. Whereas, the activity concentration

Fig. 3 Average activity concentrations of ²³²Th in soil samples in 15 districts of Savannakhet province

of 40K was determined directly from its gamma line of 1460 keV. The activity concentration of a certain radionuclide is calculated as follows [[13](#page-12-15)]:

$$
A(\text{Bq kg}^{-1}) = \frac{n}{\epsilon \times I_{\text{eff}} \times m_{\text{s}}},\tag{1}
$$

where *A* is the activity concentration of the radionuclide in Bq kg−¹ ; *n* is the net gamma counting rate (cps) for a peak at a given energy; ϵ is the detector efficiency of a specific gamma-ray, I_{eff} is the emission probability of the photon, and m_s is the mass of a soil sample. The 2σ standard deviations of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K are calculated from the errors of the net gamma counting rates, the detector efficiency, the branching ratios and the mass of soil samples.

In order to assess the radiological hazards associated with natural occurring radioactivity materials, radium equivalent activity Ra_{eq} , absorbed gamma dose rate D , annual effective dose equivalent *AEDE*, external hazard index H_{ex} and internal hazard index H_{in} have been evaluated from the activity

concentrations of ^{226}Ra , ^{232}Th and ^{40}K . Since the radioactivity levels of 2^{26} Ra and 2^{32} Th decay series and 40 K in soils are non-uniform, the Ra_{eq} is commonly used to determine the total radioactivity of a sample. The Ra_{eq} is evaluated based on an estimation that 10 Bq kg⁻¹ of ²²⁶Ra, 7 Bq kg⁻¹ of ²³²Th and 130 Bq kg−¹ of 40K produce the same gamma ray dose rate. Thus, the Ra_{eq} is calculated as follows [\[22](#page-12-16), [23](#page-12-17)]:

$$
Ra_{\text{eq}} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}}
$$
 (2)

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of 226 Ra, 232 Th and 40 K, respectively.

The calculated absorbed gamma dose rate, denoted as *D*, at about 1 m above the ground surface has been evaluated using the conversion factors of 0.46 nGy h^{-1} for ²²⁶Ra, 0.62 nGy h−¹ for 232Th and 0.042 nGy h−¹ for 40K. Hence, the absorbed gamma dose rate, *D*, can be expressed in as follows [[24\]](#page-12-18):

$$
D(nGy h^{-1}) = 0.46A_{Ra} + 0.62A_{Th} + 0.042A_K.
$$
 (3)

Fig. 4 Average activity concentrations of ⁴⁰K in soil samples in 15 districts of Savannakhet province

The outdoor annual efective dose equivalent, *AEDE*, was calculated using following equation [[1\]](#page-12-0):

$$
AEDE(\text{mSv y}^{-1}) = D \times DCF \times OF \times T \tag{4}
$$

where *D* is the absorbed gamma dose rate obtained in Eq. ([3\)](#page-4-0); *DCF* is a dose conversion factor; *OF* is an outdoor occupancy factor and T is the time factor (8760 h). The values of *DCF* and *OF* are 0.7 Sv Gy−¹ and 0.2, respectively, as taken from UNSCEAR 2000 [[1\]](#page-12-0). The outdoor occupancy factor $OF = 0.2$ is originally from considering people spending about 20% of their time outdoor [[25](#page-12-19)].

The external hazard index, H_{ex} , representing the hazard of natural gamma radiation is calculated as [[3,](#page-12-2) [23,](#page-12-17) [24](#page-12-18)]:

$$
H_{\text{ex}} = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810}
$$
 (5)

The internal hazard index, H_{in} , is calculated as [\[23](#page-12-17)]:

$$
H_{\rm in} = \frac{A_{\rm Ra}}{185} + \frac{A_{\rm Th}}{259} + \frac{A_{\rm K}}{4810}.
$$
 (6)

The radiation hazard is insignificant when the H_{ex} and H_{in} are less than unity. The value of $H_{ex} = 1$ corresponds to the upper Ra_{eq} limit of 370 Bq kg⁻¹.

Results and discussion

Activity concentrations

Fig. [2](#page-2-0) shows the average 226 Ra activity concentrations in soil samples in 15 districts of Savannakhet province. The activity concentrations of 226 Ra in soils are vary in the range from 6.6 ± 1.5 to 73.6 ± 7.7 Bq kg⁻¹. The smallest activity concentration of 6.7 ± 1.5 Bq kg⁻¹ is obtained with sample S7 in Outhoompone district, whereas the highest value of 73.6 \pm 7.7 Bq kg⁻¹ is obtained with sample S80 at Nong district. The average radioactivity level of 226 Ra in Savannakhet province is about 22.4 ± 2.1 Bq kg⁻¹. Comparing to the world average value of 35 Bq kg⁻¹, the average radioactivity concentration of 226 Ra is smaller by a factor of 0.64, but the highest value in this region is about two times greater than the world average value $[1]$ $[1]$ $[1]$. It can also be seen from Fig. [2](#page-2-0) that three regions in Savannakhet province having greater radioactivity concentrations of 226 Ra than others are Nong, Vilabuly and Sepone districts. The highest average 226Ra activity concentration of 46.3 Bq kg−¹ is obtained at Nong district.

Figures [3](#page-3-0) and [4](#page-4-1) display the average 232 Th and 40 K activity concentrations in soil samples in 15 districts of Savannakhet province. The values of 232 Th activity concentrations

Table 1 Activity concentrations of radionuclides 226Ra, 232Th and 40 K in soil samples in Savannakhet province, Laos

in the soil samples vary in the range from 3.8 ± 1.5 to 113.8 ± 3.2 Bq kg⁻¹. The smallest value of 3.8 ± 1.5 Bq kg⁻¹ corresponds to sample S24 collected in Xayphoothong district, while the highest value of 113.8 ± 3.2 Bq kg⁻¹ is obtained with sample S57 in Vilabuly district. The highest average value of 61.7 Bq kg−¹ is obtained at Nong district as shown in Fig. [3](#page-3-0). It is noticed that the average radioactivity concentration of 232Th in Savannakhet province is about 30.8 ± 2.9 Bq kg⁻¹ which is approximate the world average value of 30 Bq kg⁻¹[[1\]](#page-12-0). However, the highest concentration of 232Th in Savannakhet province is greater than the world average value by a factor of 3.8.

The activity concentration of 40 K in Savannakhet province is in the range from 13.6 ± 3.3 to 906.4 ± 31.4 Bq kg⁻¹. The lowest value of 13.6 \pm 3.3 Bq kg⁻¹ is obtained with sample S13 in Khanthabuly district, and the highest value of 906.4 \pm 31.4 Bq kg⁻¹ is obtained with sample S80 in Nong district. Nong district also corresponds to the highest aver-age value of [4](#page-4-1)88.4 Bq kg⁻¹ as shown in Fig. 4. The average 40 K activity concentration is 211.6 \pm 16.5 Bq kg⁻¹, which is about half of the world average value (400 Bq kg^{-1}) [\[1](#page-12-0)]. However, the highest value in Nong district is greater than the world average value by a factor of 2.3.

Table [1](#page-5-0) presents the average activity concentrations of radionuclides ^{226}Ra , ^{232}Th and ^{40}K in soil samples in 15 districts of Savannakhet province in comparison with the world average values. It is noticed that the activity concentrations of the radionuclides are higher at Sepone, Vilabuly and Nong districts compared to other regions in Savannakhet province. The three districts are also known as locations of mining interest. From Figs. [2,](#page-2-0) [3](#page-3-0) and [4,](#page-4-1) one can also see the higher activity concentrations of natural occurring radioactive materials in the three districts than that in the others. The activity concentrations of $226Ra$ and $232Th$ in the three

Table 2 Comparison of activity concentrations of radionuclides in Savannakhet province, Laos and other worldwide regions

a Average activity concentration

^bRange of activity concentration

districts are greater than the average value by a factor of 1.5–2.1. Whereas, the activity concentration of 40K obtained in the three districts is greater than the average value by a factor of 1.7–2.4.

Table [2](#page-6-0) shows the comparison of the average activity concentrations of natural radionuclides obtained in soil samples in Savannakhet province, Laos with other worldwide regions. Comparing with neighboring regions such as Hue and Da Nang provinces of Vietnam, the average activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Hue province are 57.0, 47.8 and 309.0 Bq kg−¹ , while the values in Da Nang province are 51.0, 58.2 and 366.2 Bq kg⁻¹, respectively, which are greater than that obtained in Savannakhet province [\[11](#page-12-11)]. Comparing with other worldwide regions as listed in Table [2,](#page-6-0) one can see that in general, Savannakhet province is among the regions with relatively lower activity concentrations of natural radionuclides in soils than others.

Radium equivalent activity

The maximum value of Ra_{eq} in soils should be less than the limit of 370 Bq kg⁻¹ to ensure the external dose less than 1.5 mGy h−¹ as recommended by UNSCEAR reports [[1,](#page-12-0) [3](#page-12-2)]. Figure [5](#page-7-0) shows the calculated average Ra_{eq} in the soil samples in 15 districts of Savannakhet province. The Ra_{eq} values vary in a wide range from 17.5 to 299.9 Bq kg⁻¹ with the average value of 82.3 Bq kg−¹ . One can see that similar to the radioactivity concentrations, three districts having greater average values of Ra_{eq} than others are Nong, Vilabuly and Sepone. The highest Ra_{eq} value of 299.9 Bq kg⁻¹ obtained with sample S80 at Nong district is still smaller than the limit of 370 Bq kg^{-1} , while the average Ra_{eq} is much smaller than the limit value by a factor of 0.22 [\[1](#page-12-0)].

Fig. 5 Average radium equivalent activity Ra_{eq} in 15 districts of Savannakhet province

Fig. 6 Calculated average absorbed gamma dose rate *D* (nGy h[−]¹) in 15 districts of Savannakhet province

Absorbed gamma dose rate

Fig. [6](#page-8-0) shows the average *D* values in 15 districts of Savan– nakhet province. The calculated values of *D* due to the terrestrial gamma radiation are obtained in the range of 8.1–137.5 nGy h⁻¹ with the highest value of 137.5 nGy h⁻¹ occurring at Nong district. The average value of *D* in Savannakhet province is 37.4 nGy h⁻¹, which is smaller than the world average value of 59 nGy h⁻¹, but the highest value of *D* is greater than the world average value by a factor of 2.3.

Annual efective dose equivalent

The *AEDE* values are obtained in the range from 0.01 to 0.17 mSv y⁻¹. The highest *AEDE* value corresponds to sample S80 at Nong district, where the highest activity concentrations of radionuclides are obtained. Figure [7](#page-9-0) show the average values of *AEDE* in 15 districts of Savannakhet province, which vary in the range of 0.026–0.093. The average value of *AEDE* in total is 0.05 mSv y⁻¹ which is lower than the world average value of 0.07 mSv y^{-1} [[1\]](#page-12-0). These values

are less than the *AEDE* limit of 1 mSv y−¹ for an individual and 20 mSv y−¹ for radiation workers as recommended by International Commission on Radiation Protection [[26\]](#page-12-24).

External and internal radiological hazard indices

Figures [8](#page-10-0) and [9](#page-11-0) show the calculated average H_{ex} and H_{in} obtained from the activity concentrations in soil samples in 15 districts of Savannakhet province. The H_{ex} values in the soil samples are within the range from 0.05 to 0.81. This means that the highest value of H_{ex} is less than unity. The average values of H_{ex} in 15 districts (0.121–0.444) as dis-played in Fig. [8](#page-10-0) and the average H_{ex} in total of 0.22 ± 0.03 are much less than unity. As shown in Fig. [9,](#page-11-0) the average H_{in} values in 15 districts and the average H_{in} of 0.28 \pm 0.03 in Savannakhet province are much less than unity in most of the area. There are two samples with higher values of *H*in (0.94 and 1.01) found in Vilabuly and Nong districts, but only one sample (S80 at Nong district) has the H_{in} greater than unity. The results of H_{ex} and H_{in} imply that there is

Fig. 7 Calculated average annual external effective dose rate *AEDE* (mSv y⁻¹) in 15 districts of Savannakhet province

no signifcant radiological hazard to human health in this region.

Table [3](#page-11-1) shows comparison of the average radiological hazard indices obtained in Savannakhet province with that reported for worldwide regions. In general, the radiological hazard indices obtained in Savannakhet province are relatively lower than that of other regions in Southeast Asian countries as well as world average values. The average Ra_{eq} values in neighboring regions such as Hue and Da Nang provinces, Vietnam (149 and 162 Bq kg⁻¹, respectively) are comparable with the highest values obtained in Savannakhet province (131 and 160 Bq kg^{-1} in Sepone and Nong districts, respectively) [[11\]](#page-12-11).

Conclusions

Measurement of the radioactivity concentrations of 80 soil samples collected widely in Savannakhet province, Laos is conducted using a HPGe gamma spectrometer for evaluating a baseline data of radioactivity concentrations and radiological hazards in the area. The activity concentrations in soil samples are in the range from 6.6 to 73.6 Bq kg⁻¹ for of ²²⁶Ra, from 3.8 to 113.8 Bq kg⁻¹ for ²³²Th and from 13.6 to 906.4 Bq kg⁻¹ for ⁴⁰K, respectively. The average activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K are 22.4 \pm 2.1, 30.8 ± 2.9 and 211.6 ± 16.5 Bq kg⁻¹, respectively. The values of activity concentrations are relatively low compared to the

Fig. 8 Calculated average external hazard index H_{ex} in 15 districts of Savannakhet province

world average values and that of other worldwide regions. The average activity concentration of 232 Th is approximate the world average value (30 Bq kg⁻¹), but the value of ⁴⁰K is about half of the world average value $(400 Bq kg⁻¹)$. The average Ra_{eq} is 82.8 \pm 9.7 Bq kg⁻¹, which is much less than the safety limit of 370 Bq kg−¹ . The highest concentrations of all three nuclides were found at Nong district but the highest value of Ra_{eq} (300 Bq kg⁻¹) is still less than the safety limit value. The results of radiological hazard indices such as absorbed gamma dose, annual efective dose equivalent, external and internal radiation hazard indices indicate no signifcant efect to human health.

Fig. 9 Calculated average internal hazard index H_{in} in 15 districts of Savannakhet province

a Average value

^bRange of the values

Compliance with ethical standards

Conflict of interest The authors declare that they have no confict of interest regarding the publication of this paper.

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