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Assessment of the natural radiological hazards in surface soil at high-background natural radiation areas in Northern Vietnam

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Abstract

Radiological hazard assessment is an important task. The natural radionuclides in 88 surface soil samples in/surrounding high-level natural radiation areas were measured using a high-purity germanium gamma-ray detector. The ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K activities varied from 60.4 to 655, 59.3 to 643, 71.2 to 886, and 252 to 745 Bq/kg respectively. The highest radionuclides concentration was found in rare earth element mines, followed by uranium and metallic mines. There was an equilibrium between ^{238}U and ^{226}Ra with ^{238}U/^{226}Ra ratios equal to 1.01. The radiation hazard indices in the study soil samples were far higher than the world average values.

Keywords Radiological hazard · Natural radionuclides · High-level natural radiation · REE mine · Uranium mine

Introduction

Natural radionuclides in soil, water, air, food, and even our bodies can originate from different sources such as weathering of the earth's crust, mining activity, fertilizer materials, and fallout from the atmosphere [1–15]. In which, the radionuclides in soil, including ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K are the main contributors to outdoor terrestrial radiation [16–19]. In addition, the radionuclides in the soil can transfer to the plants and accumulate in our bodies via ingestion [4, 9, 20–27]. Therefore, the investigation of natural radionuclides in the environment plays an important role in public health which can be used to assess the radiation population exposure and estimate the radiological hazard.

Investigations on natural radionuclides have received attention in the world and led to extensive studies in many countries, especially in/or surrounding high-level natural radiation areas. Recently, the natural radioactivity in rock and soil has been investigated worldwide [3, 7–9, 11, 12, 26, 28–41]. In general, the natural radioactivity concentrations in soil depend on the types of rock and their migration from rock to soil. Thus, the activity concentration of natural radionuclides in different surface soil areas needs to be examined, especially in/or surrounding high-level radiation areas.

In north Vietnam, there is a distribution of many mines which contain high content of radioactive elements such as rare earth mines in NamXe (NX), DongPao (DP) (Lai Chau), MuongHum (MH) (Lao Cai), YenPhu (YP) (Yen Bai); polymetallic mines (contain high uranium concentration) in NuiPhao (NP) (Thai Nguyen); uranium ore in BacYen (BY) (Son La); ThanhSon (TS) (Phu Tho), BinhDuong (NB) (Cao Bang). According to a recent report, these mines have a high radioactive background according to unpublished data from the Radioactive & Rare Minerals Division [42]. Therefore, in this study, the ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K concentrations in soil samples in these mines have been investigated. The study will provide baseline data for radiological hazard assessment when these mines will explore and exploit in the near future. Besides, current the radiological hazard indices are also evaluated based on the activity concentrations of these study radionuclides.

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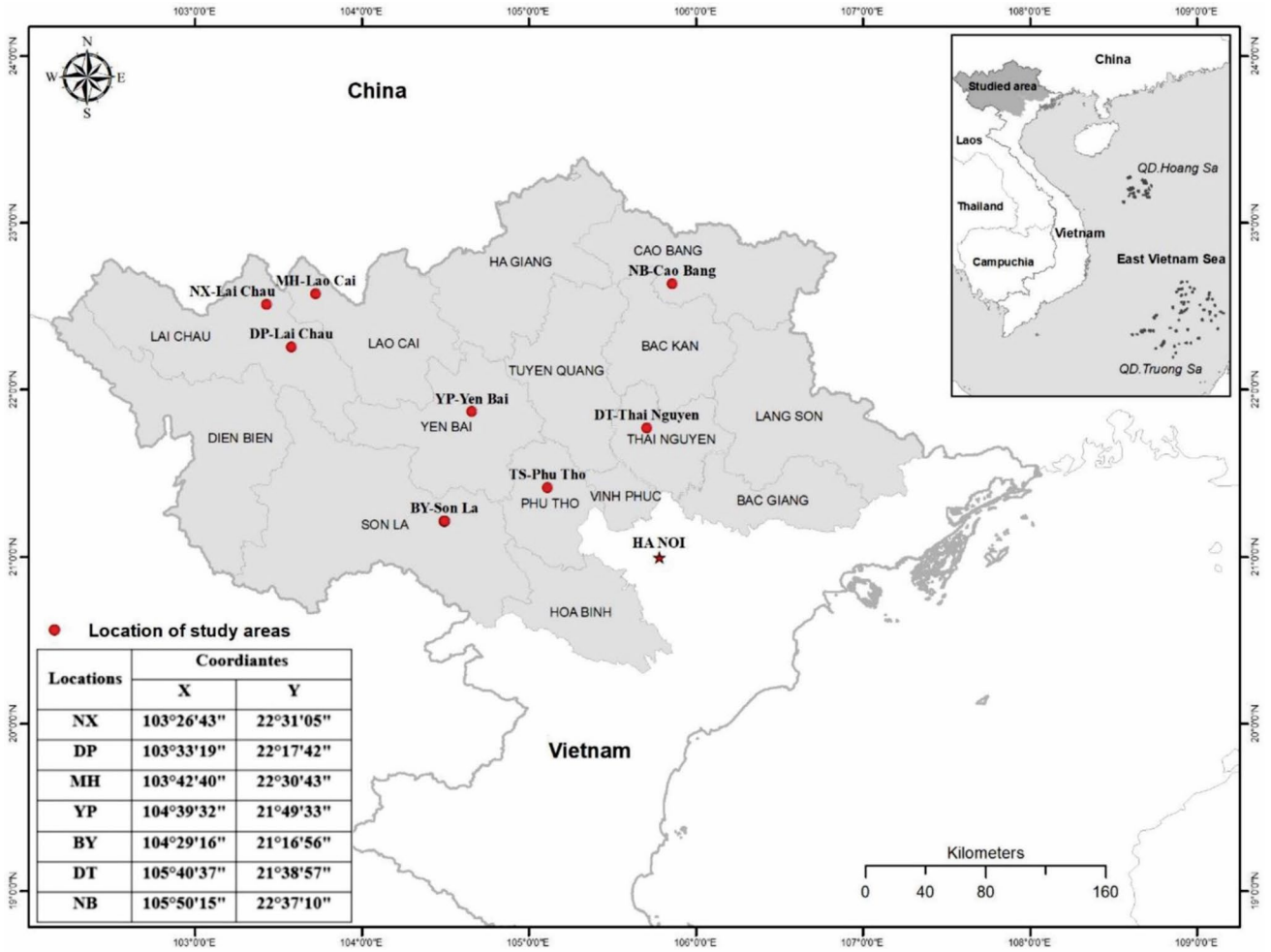
Materials

Study areas

There are eight different mines that contain high activity of natural radionuclides in Northern Vietnam. The study locations are presented in Fig. 1. In which, NX mine is one of the largest rare earth mines in Vietnam with probable reserves of about 7.7 million tons. DP mine ranks second with probable reserves of about 3.7 million tons and followed by MH with approximately 400,000 tons, YP with about 5000 tons. BY (Son La) is the location of the Cu-Ni deposit; TS (Phu Tho) and NB (Cao Bang) have uranium ore deposits while DT (Thai Nguyen) has the largest polymetallic mine in Vietnam. In each study area, eleven samples were selected including one test sample. The sampling points are random and dispersed within a geological formation, the formation of each studied mines. Namely, the NX area is located in the Vien Nam eruptive rock formation; DP in the Dong Giao Formation; MH in the Quaternary Formation; YP in the Dual Buy system; BY in the Suoi Be Formation; TS in the Sin Quyen Formation; NB in the Na Quan Formation; DT in the Phu Ngu Formation. The study samples were taken near the residential areas and distributed surrounding the mines to serve the purpose of the radiological hazard assessment of the health of the people. The sampling points were close to, or stood in the ore bodies of rare earth element or uranium mines in study areas.

Sample collection and preparation

Soil samples are the product of weathering process from bedrock. In each study location, eleven soil samples were collected with a total of eighty-eight soil samples at 8 study locations. The soil samples were taken at a depth of 0-30 cm from the surface. The soil was collected from five points



Locations	Coordiantes	
	X	Y
NX	103°26'43"	22°31'05"
DP	103°33'19"	22°17'42"
MH	103°42'40"	22°30'43"
YP	104°39'32"	21°49'33"
BY	104°29'16"	21°16'56"
DT	105°40'37"	21°38'57"
NB	105°50'15"	22°37'10"

Fig. 1 Location map with the coordinate of the center of study areas (map was modified from Hung et al. [43])

including four corners and the center of a square $1m^2$ meter with above depth. The soil sample was mixed inhomogeneous then one kilogram in weight was sampled. The selected samples were then removed from the stone, and tree roots and then put into plastic bags. The obtained samples were transported to the laboratory. The soil samples were dried at 105 °C to a constant weight. The dried soil samples also were then milled into powder. The powder was weighed and hermetically sealed in plastic containers for 30 days to obtain a secular equilibrium between the radium and its daughter radionuclides (following the modified sample-preparing procedure of [7, 44]. To avoid the contamination from system and material of containers, a blank sample) was also prepared which was used to subtract background and contaminated contribution.

Methods

Measurements of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K activity concentration in soil samples

The samples waited for the equilibrium status between radium and its daughter radionuclides. The activity concentration was measured using a high-resolution detector HPGe with a low background of ORTEC™ (detector energy resolution is 1.9 keV at the 1.33 MeV ^{60}Co gamma-ray peak). The analysis was performed using Gamma Vision software. To reduce the radiation in the laboratory, the detector is shielded by a 10-cm thick old-lead cylinder with a 1 mm cadmium and 1 mm copper inner lining. The soil samples were counted for two days to minimize the statistical counting error (less than 5% of uncertainty) and activity calculation, calibration, and quality control were carried out based on standard reference materials (RGU, RGTh, RGK, and IAEA-375).

The activity concentration of each sample was determined based on its respective gamma lines. The gamma lines of 609.3 keV, 1120.3 keV, and 1764.5 keV were used to determine the activity concentration of ^{226}Ra, while the lines of 911.2 keV, 969.0 keV, 583.0 keV were used for ^{232}Th, 1460 keV was used for ^{40}K, and 1001 keV was used for ^{238}U.

Evaluation of radiological hazard indices

Radium equivalent activity (Raeq) The radium equivalent (Ra_{eq}) is often used to determine the total radioactivity concentrations of a sample. The Ra_{eq} is calculated based on the estimation that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of ^{232}Th, and 130 Bq/kg of ^{40}K have the same gamma-ray dose rate. Therefore, the Ra_{eq} is calculated as the following equation [45]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

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

External and internal hazard index (hex and hin)

The external hazard index, H_{ex}, assesses the risk to health related to the emission of gamma radiation by different natural radionuclides. In other words, it represents the hazard of natural gamma radiation and is calculated as follows [46]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2)$$

The internal hazard index, $H_{in,}$ is used to estimate the internal exposure of living cells to radon and its progeny. It is calculated as the following equation [45].

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

The value of $H_{ex} = 1$ corresponds to the upper Ra_{eq} limit of 370 $Bq.kg^{-1}$.

Absorbed gamma dose rate

The gamma dose rate, D, is used to evaluate the exposure and absorption of radiation to the human body at 1 m above the ground containing naturally occurring radionuclides. The dose rate, D, has been evaluated using the conversion factors of 0.46 $nGy.h^{-1}$ for ^{226}Ra, 0.62 $nGy.h^{-1}$ for ^{232}Th, and 0.042 $nGy.h^{-1}$ for ^{40}K. Thus, it can be expressed as follows [47].

$$D\left(nGy.h^{-1}\right) = 0.46A_{Ra} + 0.62A_{Th} + 0.042A_K \quad (4)$$

where A_K, A_{Ra}, and A_{Th} is activity concentration of ^{40}K, ^{226}Ra, and ^{232}Th respectively.

Annual effective dose equivalent (AEDE)

The outdoor annual effective dose equivalent (AEDE) was calculated as the following equation:

$$AEDE\left(Sv.y^{-1}\right) = D\left(nGy.h^{-1}\right) \times DCF(Sv.Gy^{-1}) \times 10^{-3} \times OF \times T \quad (5)$$

where D is the absorbed gamma dose rate; DCF is an outdoor dose convention factor ($DCF = 0.7 \times 10^{-3}\ Sv.Gy^{-1}$); OF is an outdoor occupancy factor (OF = 0.2) [16]; T is the time factor (T = 8760 h).

The annual committed effective doses (CED) originating from the inhalation of radon or thoron outside dwellings can be calculated as the following equation:

$$\mathrm{CED}\left(\mathrm{mSv}.y^{-1}\right) = C \times F \times t \times \mathrm{K} \quad (6)$$

where C is the average radon or thoron concentration outside of the dwellings (Bq/m^3), F is the outside equilibrium factor for radon and its progenies or thoron and its progenies (F = 0.6 and F = 0.003 for radon and thoron respectively); t is annual spending hours outside (t = 1760 h); K = dose conversion factors (K = 9 nSv and K = 40 nSv for radon and thoron respectively) [47].

Excess lifetime cancer risk (ELCR)

Based on the values of AEDE, excess lifetime cancer risks (ELCR) were calculated using the following equations [48].

$$\mathrm{ELCR} = \mathrm{AEDE} \times \mathrm{Life\ Expectancy\ (LE)} \times \mathrm{Risk\ factor\ (RF)} \quad (7)$$

where LE is the life expectancy of Vietnamese people in North Vietnam and mountainous areas (71 years) (https://www.gso.gov.vn/default_en.aspx?tabid=774); RF is a fatal risk factor per Sievert which is equal to 0.057 Sv^{-1}[48].

Results and discussions

Soil samples

Activity concentration The range, average, skewness, and kurtosis values of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K concentrations in soil samples in the study area are listed in Table 1. As shown in this table, the activity concentration values of studied radionuclides in 8 locations mostly show light-tailed relative to a normal distribution (kurtosis < 3), except for ^{40}K in BX-Lao Cai (kurtosis > 3). In this study, the soil samples were taken from different locations which belong to three types of mine, namely rare earth element (REE) mines (NX-Lai Chau, MH-Lao Cai, DP-Lai Chau, YP-Yen Bai), uranium mines (TS-Phu Tho, NB-Cao Bang) and metal mines (BY-Son La, DT-Thai Nguyen). It can be seen that the activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K significantly vary from 60.4 to 655 Bq/kg, 59.3–643 Bq/kg, 71.2–886 Bq/kg, and 252–745 Bq/kg in respectively. The variation of the studied radionuclide concentrations is plotted in Fig. 2. There is a significant difference in the concentration of some radionuclides in study locations. The total concentration of the studied radionuclides significantly varied with the lowest value of 578 Bq/kg in DT-Thai Nguyen (metallic mine) and the highest one of 2438 Bq/kg in NX-Lai Chau (REE mine). It can be seen that the highest total activity was detected in REE mines, followed by uranium and metal mines. In particular, the highest concentrations of ^{226}Ra, ^{238}U, and ^{232}Th were observed in NX-Lai Chau (REE mine) with the value of 655, 643, and 886 Bq/kg respectively. The mine is the largest rare earth mine in Vietnam. In addition, illegal activities in the exploitation of ores by local people can release radionuclides into the surrounding soil. In the studied locations, for rare earth element (REE) mines such as NX-Lai Chau, DP-Lai Chau, and MH-Lao Cai, the concentrations of ^{232}Th are rather high since ^{232}Th is the main component of REE mines. For ^{40}K, the highest concentration was detected in DP-Lai Chau while the lowest one was found in TS-Phu Tho. The lowest concentrations of ^{226}Ra and ^{238}U were found in BY-Son La with the value of 60.4 and 59.3 Bq/kg respectively. These mines have low uranium concentrations. The lowest concentration of ^{232}Th was observed in DT-Thai Nguyen because the main ores in a polymetallic mine in this area are vonfram, fluorspar, copper, and bismuth. In general, the concentration of radionuclides significantly varies in each study area. It could be explained that the soil samples were taken from different points in, out, or close to the highly concentrated ore bodies of mines. Furthermore, the concentration of radionuclides in the soil also depends on the mineralogical composition and weathering process of the bedrock [49, 50]. For each studied location, the concentration of ^{40}K was the highest in DP-Lai Chau, BY-Son La, YP-Yen Bai, DT-Thai Nguyen, and NB-Cao Bang while the concentration of ^{232}Th was the highest in NX-Lai Chau, MH-Lao Cai, and TS-Phu Tho.

As shown in Table 1, the ratios of ^{238}U/^{226}Ra concentration in the soil slightly range from 0.97 to 1.07 with a mean value of 1.01. As reported in UNSCEAR (2000), the average value of ^{226}Ra/^{238}U in soils was about 1.03. This indicates the concentrations of ^{238}U and ^{226}Ra for soil samples in the studied area are almost equilibrium and similar to the reference value of UNSCEAR (2000). In another report by [51], the concentration of ^{238}U in surface soil can be decreased since the ^{238}U is leached from the surface soil due to rainfall and moved to the deeper soil layers. By contrast, ^{226}Ra is very immobile and cannot be dissolved by rainfall, thus its concentration in soil keeps constant. So the almost equilibrium of ^{238}U/^{226}Ra in this study shows that the leaching of ^{238}U from surface soil to deeper soil layer or removal by rainfall is insignificant.

The ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K concentrations in soil in this study are compared with previous studies in the world and presented in Table 2. In this table, the ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K concentrations in this study are higher than those in soil China, Armenia, and India [18, 29], [34, 52]. This can be due to the distribution of some rare earth elements and uranium mines in the study areas which contain high ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K concentrations. However, the concentration of ^{226}Ra in this study is lower than that in northern Iran [37], whereas the ^{40}K concentration is lower than that in northern Iran and northern India [30]. In comparison with other areas of Vietnam, the concentration of studied radionuclides in the soil in this study is significantly higher than in Ninh Son and Ho Chi Minh (Southern Vietnam), except for

Table 1 Concentrations of natural radionuclides in soil samples in Northern Vietnam

Types of mine	Locations	Value	Activity concentration (Bq/kg)					$^{238}U/^{226}Ra$
			^{226}Ra	^{238}U	^{232}Th	^{40}K	Total	
REE mine	NX-Lai Chau (n = 11)	Range	543–787	520–740	746–994	214–360		
		Average	655	643	886	254	2438	0.98
		Skewness	0.61	−0.43	−0.56	1.38		
		Kurtosis	2.44	0.65	−0.27	0.98		
	DP-Lai Chau (n = 11)	Range	150–213	151–204	177–270	413–1252		
		Average	180	175	217	745	1317	0.97
		Skewness	0.42	0.50	0.23	0.64		
		Kurtosis	−0.89	1.80	−1.18	−0.25		
	MH-Lao Cai (n = 11)	Range	359–492	347–555	553–1348	440–675		
		Average	428	436	848	601	2312	1.02
		Skewness	−0.02	0.79	0.85	−1.54		
		Kurtosis	−1.14	0.78	1.21	3.80		
	YP-Yen Bai (n = 11)	Range	25.3–148	23.1–139	53.4–229	209–501		
		Average	76.1	78.3	144	357	656	0.98
		Skewness	0.21	−0.05	−0.19	0.25		
		Kurtosis	−1.45	−1.78	−2.30	−2.10		
Metal mine	BY-Son La (n = 11)	Range	12.5–155	16.6–148	20.6–253	54.0–986		
		Average	60.4	59.3	114	528	761	1.03
		Skewness	0.91	0.98	0.32	0.05		
		Kurtosis	−0.13	−0.03	−1.77	−2.46		
	DT-Thai Nguyen (n = 11)	Range	53.9–132	59.3–146	53.3–100	122–393		
		Average	101	104	71.2	303	578	1.06
		Skewness	−0.46	0.13	0.60	−0.83		
		Kurtosis	−1.00	−1.68	0.51	0.46		
Uranium mine	TS-Phu Tho (n = 11)	Range	125–193	121–195	330–477	209–291		
		Average	154	159	387	252	952	1.03
		Skewness	0.58	−0.14	0.78	0.29		
		Kurtosis	−0.72	0.41	0.75	-0.87		
	NB-Cao Bang (n = 11)	Range	403–737	404–851	84.5–128	468–909		
		Average	594	636	103	724	2057	1.07
		Skewness	−0.72	−0.01	0.43	−0.43		
		Kurtosis	1.00	0.22	−0.82	−1.73		
Overall	Minimum value		60.4	59.3	71.2	252	578	0.97
	Maximum value		655	643	886	745	2438	1.07
	Average value		250	254	308	418	1384	1.01

^{40}K [35, 39]. The concentration of ^{40}K in Northern Vietnam is lower than that in Ninh Son and slightly higher than that in Ho Chi Minh, Vietnam.

Radiological hazards

The radiological hazard indices for the soil samples are plotted in Figs. 3, 4, 5, 6 and 7. The average R_{aeq} value for the soil in Northern Vietnam (812 Bq/kg) is about 2.5 times higher than the maximum allowable value of 370 Bq/kg [16] In particular, the significantly high values of R_{aeq} for soil samples are found in NX-Lai Chau, DP-Lai Chau, MH-Lao Cai, TS-Phu Tho, and NB-Cao Bang (Fig. 3) containing a high concentration of ^{226}Ra and ^{232}Th. The highest Ra_{eq} in NX-Lai Chau is about 5.2 times higher than the allowable value. The average values of Hex and Hin in the study areas are more than unity. In which, the H_{ex} and H_{in} values for the soil in NX-Lai Chau, MH-Lao Cai, NB-Cao Bang, DP-Lai Chau, and TS-Phu Tho are found to be higher than the desirable limit (> 1) (Fig. 4). By contrast, the H_{ex} and H_{in} values are found to be less than unity in some locations, including in BY-Son La, DT-Thai Nguyen and YP-Yen Bai (for H_{ex}) and BY-Son La and DT-Thai Nguyen (for H_{in}) (Fig. 4). Regarding the

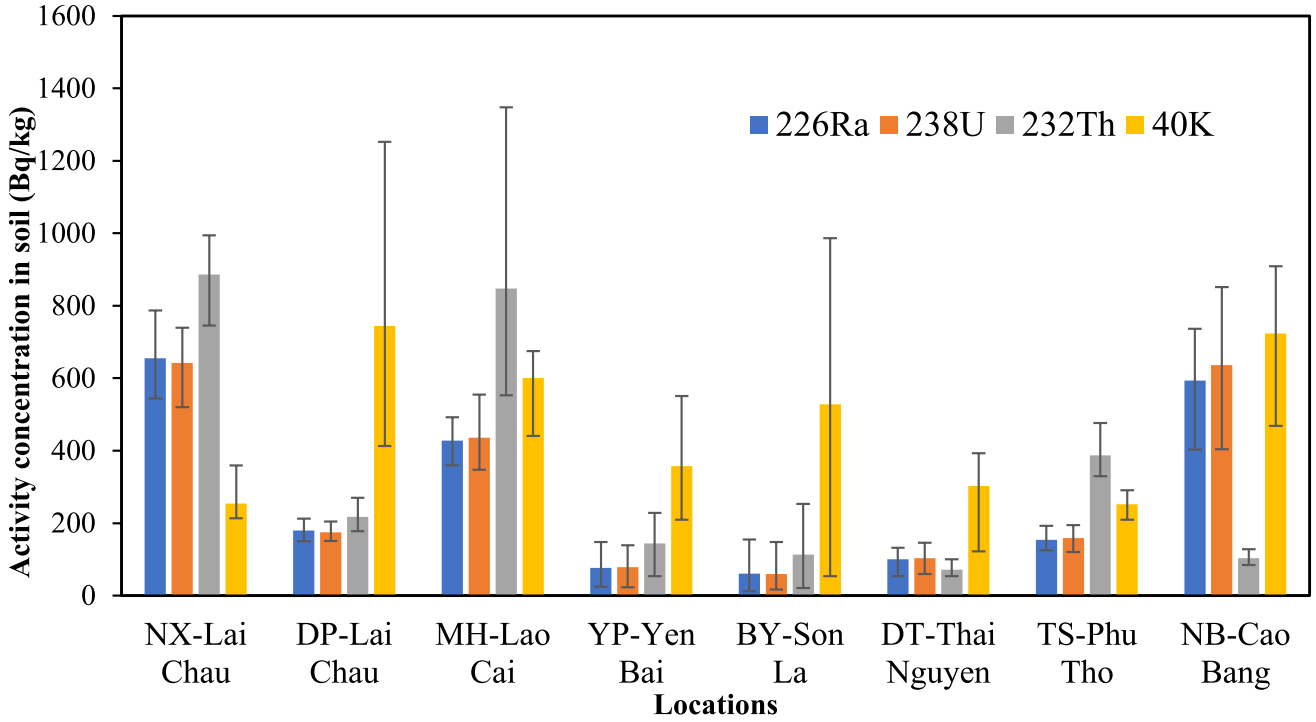


Fig. 2 Variation of min, max, and the average of natural radionuclide concentrations in soil samples in northern Vietnam

Table 2 Concentrations of natural radionuclides in soil samples in different countries

Countries	Activity concentration (Bq/kg)				References
	^{226}Ra	^{238}U	^{232}Th	^{40}K	
North Vietnam	60–655	59–643	72–886	254–745	This study
Ninh Son, Vietnam	25–193	13–161	30–206	493–1625	[39]
Ho Chi Minh, Vietnam	21–24	22–25	23–33	198 - 215	[35]
Northern Iran	4–24,284	–	ND – 107	7–853	[37]
China	6.9	11.9	9.6	39.6	[34]
	13–34.3	16.1–54	–	93.3–159	
Armenia	–	36.1–111	21.8–109	319–531	[52]
India	–	28.1–883	62.1–536	79.8–504	[29]
Punjab, India	25–48	–	25–48	356–598	[18]
Northern India	52	–	19	1627	[32]
Worldwide range	0–1000	0–1000	0.05–360	0–3200	[16]

absorbed gamma dose rate, the D values range from 104 to 861 (nGy.h^{-1}) with the mean of 364 (nGy.h^{-1}). The mean value of D is about 3.9 to 20 times higher than the range in the world which varies from 18 to 93 (nGy.h^{-1}) [16] The highest D of 861 (nGy.h^{-1}) was found in NX-Lai Chau while the lowest one was detected in DT-Thai Nguyen. Besides, the absorbed gamma dose rates (D) in all studied areas are higher than the worldwide value (Fig. 5). The values of AEDE vary from 127 to 1056 (μSv.y^{-1}) (Fig. 6) while the ELCR ranges from 0.52×10^{-3} to 4.27×10^{-3} (Fig. 7). The mean values of AEDE and ELCR are 446 (μSv.y^{-1}) and 1.8×10^{-3} in respectively. As reported, the average worldwide values of AEDE and ELCR are 70 and 0.29×10^{-3} respectively. It can be seen that in those study areas, these values are about 6.4 and 6.2 times higher than the average worldwide values.

The comparison of radiological indices for soil samples in this study with those reported from previous studies is listed in Table 3. As shown in this table, the radiological hazard indices of soil samples in Northern Vietnam are higher than those in most countries, except for Northern Iran. Furthermore, these values are also higher than the average value and range in the world as mentioned above.

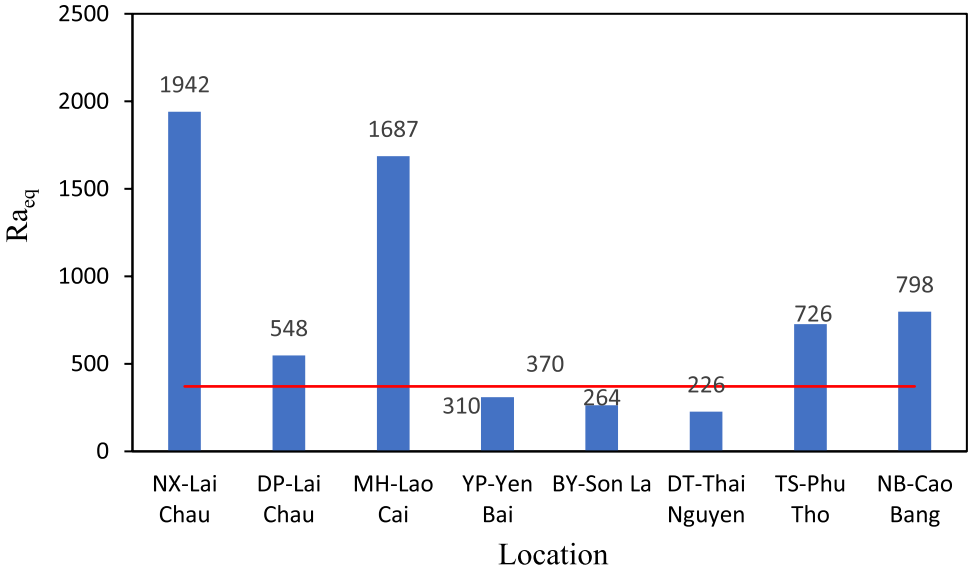


Fig. 3 Variation of Ra_{eq} in surface soil in eight study areas

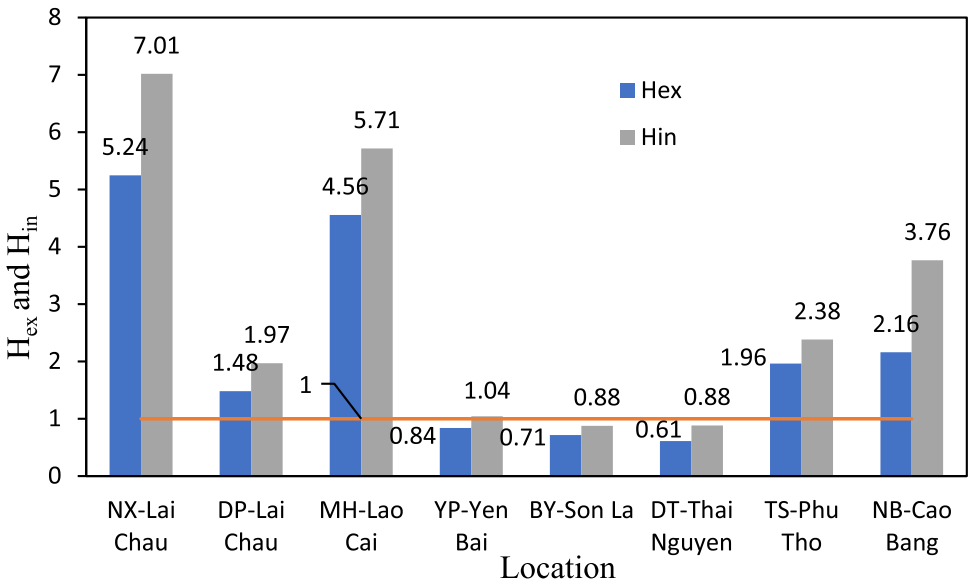


Fig. 4 Variation of H_{ex} and H_{in} in surface soil in eight study areas

Conclusions

The ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K activities in soil samples from different high-level natural radiation areas in northern Vietnam were extensively investigated. The radiation hazard assessment in the study areas was also evaluated. The concentrations of studied radionuclides significantly varied and depended on the position of soil samples (in, out, or close to the ore bodies), and the type of mines. Generally, the total concentration of studied radionuclides in REE mines was found to be the highest, followed by uranium and metal mines. The measurement results also show that the ^{238}U is almost in equilibrium with ^{226}Ra in all studied areas. Regarding the radiological hazards, the assessment of radiation hazard indices showed that all the natural hazard indices in those study areas were significantly higher than the average in the world. Based on this investigation, those areas should have to do further extra assessment in detail of radiological hazards for the population living in and surrounding those study areas.

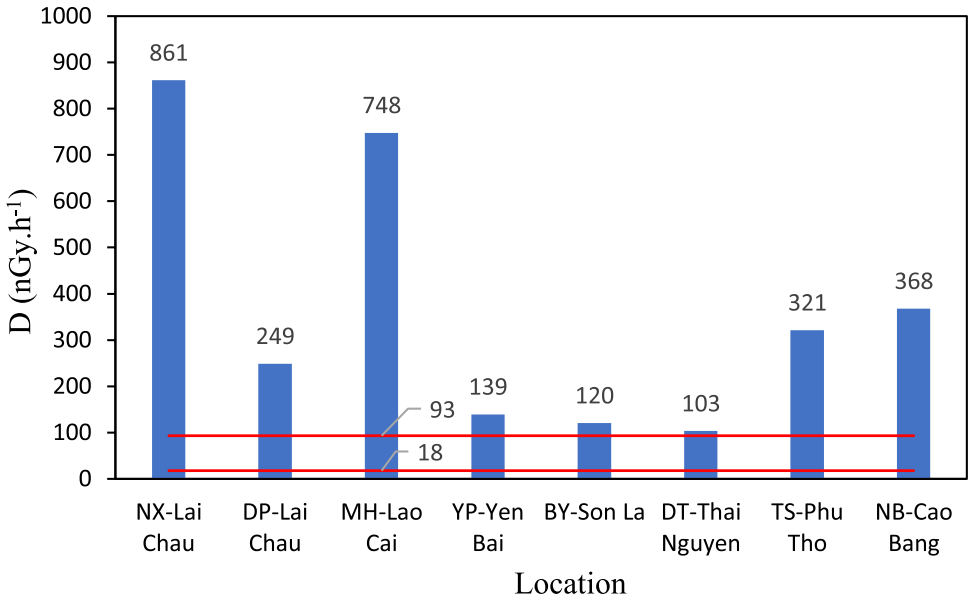


Fig. 5 Variation of absorbed gamma dose rate in surface soil in eight study areas

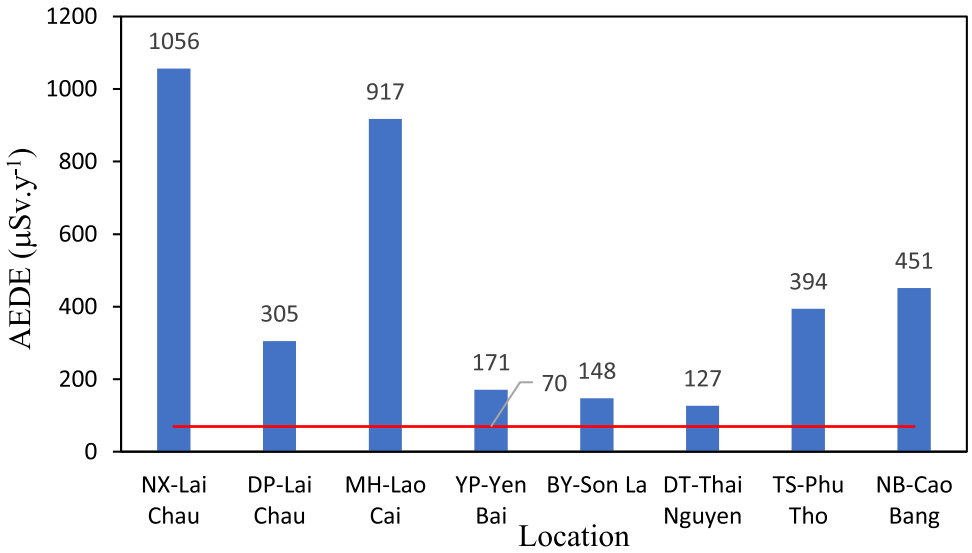


Fig. 6 Variation of AEDE in surface soil in eight study areas

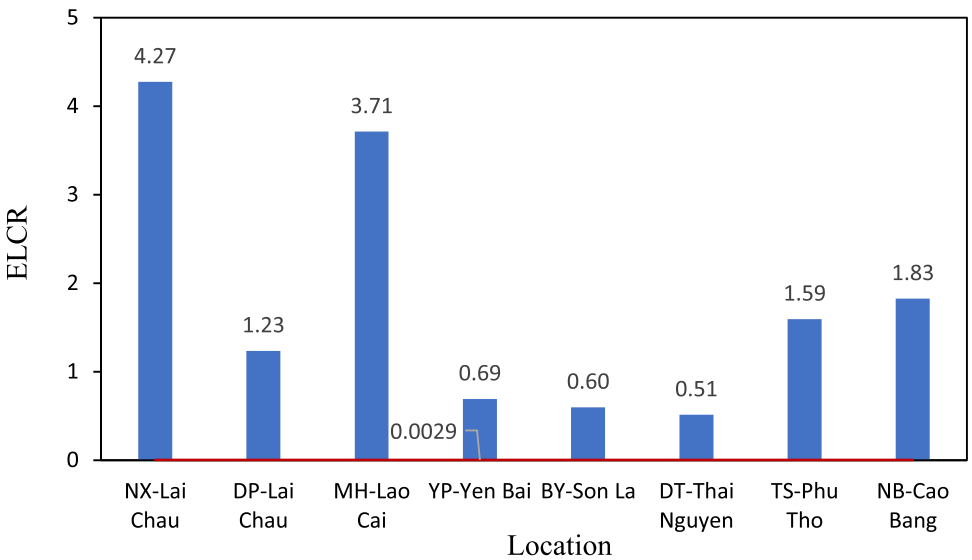


Fig. 7 Variation of ELCR in surface soil in eight study areas

Table 3 Radiation hazard indices for soil samples in different countries

Countries	R_{aeq} Bq/kg	H_{ex}	H_{in}	D nGy.h^{-1}	AEDE μSv.y^{-1}	ELCR ($\times 10^{-3}$)	References
Northern Vietnam	227–1942	0.61–5.24	0.67–6.07	104–861	127–1056	0.52–4.27	This study
Ninh Son, Vietnam	114–533	–	–	55–248	67–304	–	[39]
Ho Chi Minh, Vietnam	47 – 148	–	–	21–64	25–78	0.088–0.273	[35]
Northern Iran	5–24,300	0.01- 66	0.03–131	3–11,240	3–1300	0.01- 52	[37]
China	8.4–103	0.03–0.28	0.05–0.37	3.7–46.4	112–976	-	[34]
Kapan, Armenia	–	–	–	17.7–72.0	20–90	0.8 – 0.31	[52]
Kajaran, Armenia	–	-	–	48.6–180	60–220	0.21–0.77	
India	–	–	–	137–335	-	-	[29]
Punjab, India	–	–	–	52–69	60–80	0.21–0.28	[18]
Northern India	205	0.55	0.69	77–123	470–750	-	[32]
Worldwide	370	≤1	≤1	18–93	70	0.29	[16, 47]

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