



Relationship between physicochemical parameters with radioactive concentration in soil and annual effective dose

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Abstract

This study aims to clarify the relationship between soil physicochemical properties and the concentration of natural radionuclides. Our results show that principal component analysis indicated a positive correlation between activity concentration and sand content, and a negative correlation between activity concentration and clay content, while other factors showed little information about clear correlations with concentration. The calculated total average dose and total cancer risk exceeds permissible limits for the public, indicating a significant potential health impact on human health. The results show that the area around the mine exhibits a potential radiation hazard.

Keywords Soil physicochemical · Natural radionuclides · Annual effective dose · Principal component analysis

Introduction

Soil plays an important role in managing and influencing the movement and distribution of natural radionuclides in the environment [1]. Radionuclides such as ^{226}Ra , ^{232}Th , and ^{40}K are naturally occurring radioactive materials that can pose significant risks to human health and ecosystems, particularly in high radiation background areas. The world average values of these three isotopes, according to the UNSCEAR (2000) report, are 30, 35, and 400 Bq/kg for ^{226}Ra , ^{232}Th , and ^{40}K , respectively. Therefore, research on natural radioactivity in soil and the associated hazard indices has been receiving considerable attention worldwide. In Vietnam, studies on this topic have been conducted by many scientists and organizations to assess radioactivity levels and identify influencing factors [2–8]. Many studies in mining areas have been carried out to evaluate radioactivity levels and the impact of mining activities on the environment. Results often indicate that the concentration of radionuclides

are higher in these mining areas compared to other regions and global averages [2, 8, 9]. Several studies have examined the relationship between natural radioactivity levels in soil and public health issues. These studies may help to identify potential risks and propose preventive measures [2, 6, 7, 10].

The concentration and mobility of naturally occurring radioactive substances in soil are influenced by various physicochemical properties, including soil pH, organic carbon (OC) content, soil texture, and cation exchange capacity. Understanding the relationship between soil physicochemical properties and the concentration of radionuclides is essential for predicting the distribution and potential impact of radioactivity on health. For example, soil pH can affect the solubility and bioavailability of radioactive substances, while organic matter can bind with radioactive substances and influence their mobility [11, 12]. Additionally, soil texture affects water retention and movement, thereby influencing the transport of radioactive substances through the soil profile [13]. Tsuey-Lin et al. (2011) investigated the relationship between the physicochemical properties of soil and its radioactive activity. The findings indicate a negative correlation between radioactive activity and both pH and conductivity and suggests that the transport of radionuclides in the soil is influenced by sorption to soil particles and possibly the migration of these particles [14]. Prakash (2018) reported that this finding aligns closely with Narayana's study in 2010, which similarly found no clear correlations between

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OC content and the concentrations of ^{226}Ra , ^{232}Th and ^{40}K [15, 16]. Weak to negligible correlations were observed between pH and the activities of ^{226}Ra and ^{232}Th [16]. Several studies have also highlighted the influence of soil pH and organic matter content (OM) on the concentration of natural radioactive nuclides [15, 17, 18].

By determining the levels of radioactive nuclides (^{226}Ra , ^{232}Th , and ^{40}K) in soil samples collected from high radiation background areas in Lao Cai, Vietnam, and examining the relationship between the concentration of these nuclides and soil physicochemical parameters, we aim to provide valuable insights for environmental protection and public health. Understanding the mechanisms behind the distribution of radioactive substances in soil can inform remediation strategies and land management practices, ultimately reducing exposure risks for residents in affected areas.

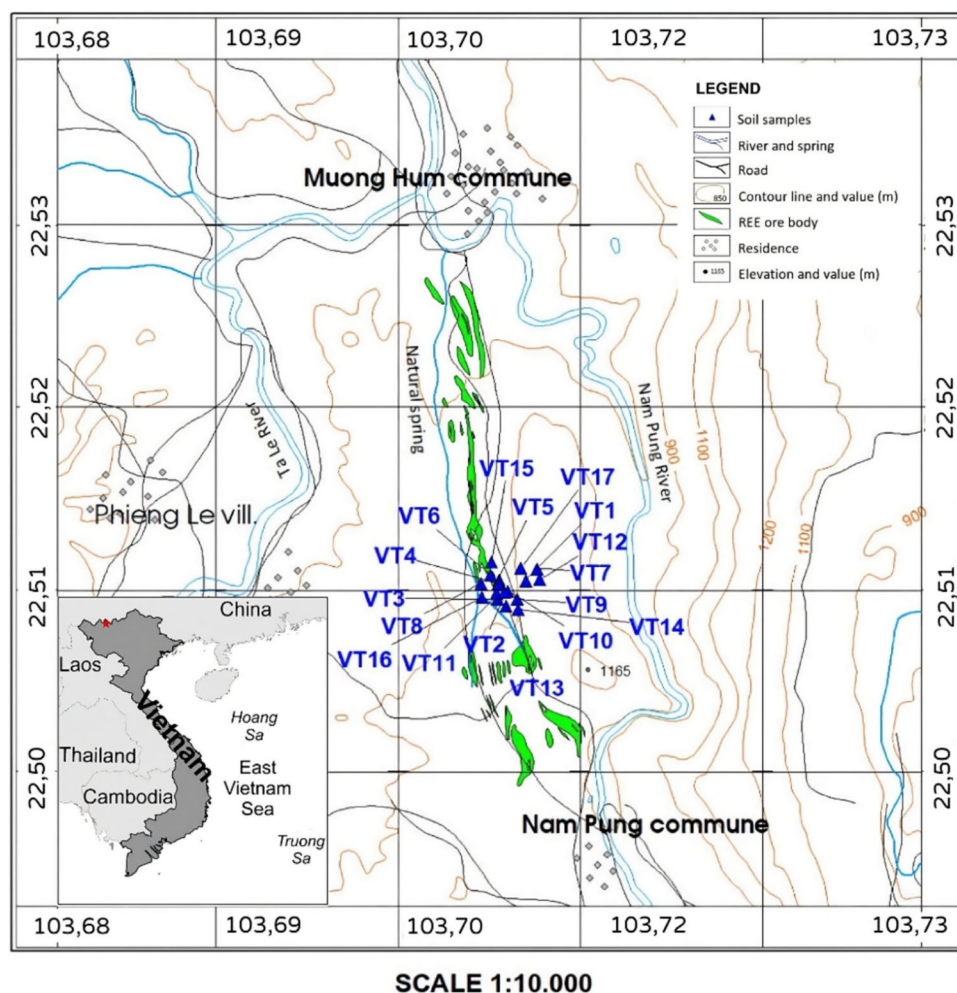
Materials and methods

Geographical location and geological characteristic

The study area is located in Nam Pung commune, an area with high radiation background due to the influence of rare earth (REE) mines in Bat Xat, Lao Cai which is a province located in the Northwest region of Vietnam. Sampling diagram shown in Fig. 1. The geographical and geological characteristics of this area have been mentioned in several reports, highlighting that the mineral composition of rare earth ore in this mine mainly includes thorite, euxenite, bastnäsite, samarskite, magnetite, ilmenite, ilmenorutile, orthite, barite... etc. Among these, the average content of ThO_2 is 0.157%, and U_3O_8 is 0.016% [2, 19].

Nam Pung has a land area of about 63.17 km² and is located in a mountainous area, the terrain is mainly hills, mountains, and valleys. The soil belongs to the group of Haplic Acrisols (Hfa) with dark gray, gray-yellow or red-yellow colour, light to medium mechanical composition, granular, porous structure,

Fig. 1 Sampling diagram



parent rock is in the process of strong weathering. Due to the presence of high mountainous terrain and large fragmentation, the climate of this region is subtropical and temperate, suitable for the growth of many types of fruit trees and temperate vegetables.

Description and processing of the sample

Soil samples in this study were collected from 17 locations in residential areas for evaluation. The soil samples were taken using a thin steel core (length: 70 cm, diameter: 110 mm). At each sampling point, we collected five individual samples from the topsoil layer (0–30 cm), then mixed them thoroughly and took about 3 kg of soil, which was labeled for identification. After removing gravel, stone fragments, and tree roots, they were packed in plastic bags and transported to the laboratory. The sampling procedures follow IAEA guidelines and ISO 18400–101:2017 standard [20]. These samples were dried at 105 °C for 8 h and then milled to a diameter of less than 0.2 mm using a sieve [5]. The fine soil was spread evenly on a flat surface to form a square and then divided into four equal parts. Soil from two opposite quarters was collected, mixed thoroughly, and about 300 g of the sample is collected. The samples were then sealed in a Marinelli glass container and left to stabilize for 30 days. The samples were then sealed in a Marinelli container and left to stabilize for 30 days to reach secular equilibrium.

Radioactivity analysis

The concentrations in studied samples were measured using a gamma spectrometer system consist of a Broad Energy Germanium (BE6530 model) detector manufactured by Canberra and an accompanying electronic part consisting of a pre-amplifier, an amplifier, a multi-channel analyzer (MCA) and computer-based software. The BEGe detector can detect gammas in a wide energy range (from 3 keV to 3 MeV) with a resolution of 2.0 keV at the energy of 1332 keV. The Genie 2000 computer-based software is used to control the 8192 channel Lynx MCA and to acquire, process, and display the gamma spectra. In addition to its capabilities for collection and processing, it also provides a variety of features such as peak search, peak fitting, background subtraction and energy calibration. The gamma emission peaks used to determine the nuclide ^{226}Ra , ^{228}Ra and ^{40}K are listed in Table 1 [21–23].

The concentration, A_X (Bq kg $^{-1}$), of a radionuclide X is written as T.-H. Bui et al., 2023:

$$A_X = \frac{n(E_\gamma, X)}{\varepsilon(E_\gamma) \times I(E_\gamma, X) \times m} \quad (1)$$

where $n(E_\gamma, X)$, $\varepsilon(E_\gamma)$, $I(E_\gamma, X)$ and m are respectively the net counting rate (cps) for a peak, the detector efficiency,

Table 1 Gamma emission characteristics of measured radionuclides

Radionuclide determined	Radionuclide measured	Energy— E_γ (keV)	Intensity— I_γ (%)
^{226}Ra	^{214}Bi	609.31	45.49
		1120.30	14.91
		1764.49	15.31
^{228}Ra	^{228}Ac	911.20	25.80
^{228}Ra	^{208}Tl	583.0	30.55
		2614.50	22.6
^{40}K	^{40}K	1460	10.66

the gamma emission probability at the energy E_γ and mass of sample.

The measurement of radioactive nuclides, self-absorption effects and summation peaks effects have been calibrated by us using standard samples. With the standard and measured samples having the same geometry and composition, effects such as self-absorption and summation peaks can be neglected. Additionally, with the selected configurations, the small sample mass (300 g) was measured on a BeGe spectrometer, so all analyzed spectra have a dead time of less than 0.5%, allowing the summation peaks effect to be ignored. Regarding the self-absorption effect in the sample, the gamma emission peaks used for analysis have high energy and the sample thickness is small, so the self-absorption effect in the sample can also be neglected.

Soil properties analysis

In this work, soil physicochemical properties such as soil structure, organic carbon and soil pH were investigated to better understand the influence of these factors on the concentration of radionuclides. Robinson pipette method is a technique used to determine the sand, silt and clay content of soil samples [24]. The setting velocity of particles (affected by their size and density) helps differentiate between sand, silt and clay fractions. The humic acid and fulvic acid content (organic carbon) in soil was determined by the Walkey-Black method based on the oxidation properties of organic matter using potassium dichromate solution ($\text{K}_2\text{Cr}_2\text{O}_7$) [25]. Finally, soil pH was determined by mixing a sample of soil with a specific concentration of potassium chloride (KCl) solution [26].

Radiological hazard indices

The gamma absorbed dose rate in the air calculated according to UNSCREAR (2000) [27]:

$$D_{in} (n\text{Gyh}^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.081A_K \quad (2)$$

$$D_{out}(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (3)$$

where D_{in} , D_{out} are the indoor and outdoor gamma absorbed dose rate which calculated at 1 m above the ground; A_{Ra} , A_{Th} , A_K are the activity concentrations in $Bq\ kg^{-1}$ of ^{226}Ra , ^{232}Th and ^{40}K (in the soil, respectively).

Corresponding to the absorbed dose rate in air, the indoor (E_{in}) and outdoor (E_{out}) annual effective dose are calculated as follows [28–31]:

$$E_{in}(\mu Sv.h^{-1}) = D_{in}(nGyh^{-1}) \times 4.91 \quad (4)$$

$$E_{out}(\mu Sv.h^{-1}) = D_{out}(nGyh^{-1}) \times 1.23 \quad (5)$$

The value of excess lifetime cancer risks—ELCR is calculated by multiplying E_{in} and E_{out} with the life expectancy (LE) (about 70 years) and a fatal risk factor per sievert (RF) ($0.055\ Sv^{-1}$) as follows [28, 29, 32]:

$$ELCR_{in} = E_{in} \times LE \times RF \quad (6)$$

$$ELCR_{out} = E_{out} \times LE \times RF \quad (7)$$

Principal component analysis (PCA)

Principal Component Analysis is a widely-used technique for dimensionality reduction, which identifies the principal components of the data. It is one of the most widely used methods in machine learning and statistics for reducing the number of dimensions of data, while retaining the most important information present in the original dataset. In this work, PCA is conducted using XLSTAT 2024 by Lumivero, a statistical tool integrated with Microsoft Excel, to investigate the influence of each soil physicochemical parameters, such as soil pH, organic carbon and soil texture (sand, silt and clay) on the concentration of natural radionuclides in the soil. The results are described in the form of Biplot.

Results and discussion

The soil physicochemical properties

Table 1 presents the results of the analysis of physicochemical properties in soil samples. Soil pH values range (average) from 4.2 (VT9)—6.8 (VT16), with an average of 5.6. Compared to other regions in Vietnam, the average soil pH in this work is lower than that of cultivated soil in Tien Le, Hanoi (6.6) [3], but higher than paddy soil in Hoa Vang district, Da Nang (located in the South Central Coast of Vietnam) (4.0) [33] and Red River Delta in Northern Vietnam (4.7) [34]. In general, agricultural soils in many different

regions of Vietnam tend to be acidic. The Organic carbon (OC) range from 0.1% to 0.4%, averaging 0.2% which is lower than that in Hoa Vang, Da Nang (1.5%) and Red River Delta in northern, Vietnam (1.6%) by a factor around 7 [33, 34]. The average fraction of sand, silt and clay in the soil are 33.09%, 45.16% and 21.75% respectively. According to the particle size analysis for paddy soil in Da Nang and clay soil in Northern Vietnam, the differences regarding particle size distribution were not significant.

Activity concentration in soil

The radioactive activity values of soil samples in Nam Pung are presented in Table 2. Overall, the study area exhibits a high level of radioactive background and the activity concentration of natural radionuclides in the soil fluctuates in a broad range depending on the location relative to the rare earth mine area, soil parameters and other influencing factors. The average (median) activity concentration of ^{226}Ra , ^{232}Th and ^{40}K are $62.2 \pm 2.55\ Bq\ kg^{-1}$ ($59.2 \pm 1.4\ Bq\ kg^{-1}$), $79.2 \pm 3.31\ Bq\ kg^{-1}$ ($61.8 \pm 2.6\ Bq\ kg^{-1}$) and $407 \pm 14\ Bq\ kg^{-1}$ ($390 \pm 15\ Bq\ kg^{-1}$) respectively. The high activity concentrations of ^{226}Ra are observed in several sampling sites, with VT10 exhibiting the highest value, along with other sites such as VT3, VT5, VT7, VT10 and VT13. The results show that locations with high ^{232}Th radioactivity will have high ^{226}Ra radioactivity. Elevated radioactivity levels in soil can indeed be related to the geochemical composition of the soil. These areas have higher radioactivity than other areas because they are located near the mine area in Bat Xat. The mineral composition of rare earth ores in Bat Xat, typical for placer ore types, includes thorite, euxenite, bastnäsite, samarskite, magnetite, ilmenite, ilmenorutile, orthite, barite ... etc. Thorium content ranges from 0.106 to 0.188% ThO_2 , average 0.157% ThO_2 ; uranium from 0.012 to 0.028% U_3O_8 , average 0.016% U_3O_8 [19, 35].

A comparison with studies on soil radioactivity in other locations in Vietnam shows that the concentrations of ^{226}Ra and ^{232}Th are nearly three times higher than those in Hanoi (23.7 and 28.9 $Bq\ kg^{-1}$) and Ho Chi Minh City (22.8 and 28.9 $Bq\ kg^{-1}$) [7, 36]. The concentration of ^{40}K in this study is very close to the world average value (400 $Bq\ kg^{-1}$), but larger than that in the cultivated soil in Hanoi, Vietnam (235 $Bq\ kg^{-1}$) [3] and the urban soil in Ho Chi Minh, Vietnam (213 $Bq\ kg^{-1}$) [7] by a factor of 2. Countries like South Korea, Pakistan and India have lower levels of ^{226}Ra and ^{232}Th activity compared to the values observed in this study, but higher levels of ^{40}K [37–39]. Conversely, an area with high background radiation in China has higher concentrations of ^{226}Ra and ^{232}Th (66 $Bq\ kg^{-1}$ and 109 $Bq\ kg^{-1}$ respectively) than that in this study, while ^{40}K level is lower (211 $Bq\ kg^{-1}$). The research results from areas near mines such as uranium mines in Central-West of Spain [40] and

Table 2 Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in soil and the soil physicochemical properties

Symbol	Activity (Bq kg^{-1})			Physiochemical				
	^{226}Ra	^{232}Th	^{40}K	pH	OC (%)	Sand (%)	Silt (%)	Clay (%)
VT1	31.3 ± 2.5	73.6 ± 5.0	405 ± 14	4.5	0.44	52.90	32.70	14.40
VT2	42.9 ± 2.8	52.1 ± 2.7	211 ± 11	6.0	0.16	26.80	47.90	25.30
VT3	89.9 ± 1.5	125 ± 3	396 ± 16	5.3	0.32	18.50	50.20	31.30
VT4	28.4 ± 2.0	58.1 ± 3.5	182 ± 8	6.3	0.22	29.80	36.40	33.80
VT5	86.6 ± 6.1	104 ± 7	290 ± 16	5.1	0.23	38.20	43.30	18.50
VT6	24.5 ± 1.6	28.9 ± 1.9	390 ± 15	6.1	0.16	39.70	51.20	9.10
VT7	85.4 ± 2.5	91.0 ± 3.1	364 ± 14	6.6	0.10	29.70	50.20	20.10
VT8	42.9 ± 2.8	52.1 ± 2.7	211 ± 11	6.0	0.16	26.80	47.90	25.30
VT9	22.4 ± 1.4	24.6 ± 1.9	108 ± 7.6	4.2	0.34	17.20	38.80	44.00
VT10	130 ± 3	169 ± 7	520 ± 16	4.5	0.20	52.30	31.50	16.20
VT11	96.6 ± 1.7	192 ± 4	906 ± 18	5.4	0.30	51.30	34.70	14.00
VT12	79.4 ± 2.4	49.2 ± 2.6	437 ± 13	5.4	0.29	19.40	54.80	25.80
VT13	109 ± 2	124 ± 2	670 ± 18	5.9	0.20	35.20	50.10	14.70
VT14	33.1 ± 2.7	47.9 ± 3.3	162 ± 10	6.2	0.22	25.00	54.50	20.50
VT15	30.1 ± 2.6	20.8 ± 1.5	193 ± 11	6.2	0.12	39.50	44.70	15.80
VT16	59.2 ± 1.4	61.8 ± 2.6	789 ± 20	6.8	0.12	25.40	56.40	18.20
VT17	65.5 ± 4.5	72.7 ± 2.6	690 ± 19	5.5	0.17	34.80	42.50	22.70
Mean	62.2 ± 2.5	79.2 ± 3.3	407 ± 14	5.6	0.22	33.09	45.16	21.75
Median	59.2 ± 1.4	61.8 ± 2.6	390 ± 15	5.9	0.20	29.80	47.90	20.10
Min	22.4 ± 1.4	20.8 ± 1.5	108 7.6	4.2	0.10	17.20	31.50	9.10
Max	130 3	192 4	906 18	6.8	0.44	52.90	56.40	44.00

Mean: The average value of all data points in the dataset

Median: The middle value when the data are arranged in order

Min: The lowest value in the dataset

Max: The highest value in the dataset

Egypt [41], tin mines in Nigeria [42] and Malaysia [43] or gold mines area in Bolikhamxay, Laos [44] all show that the activity concentration of natural radionuclides are significantly higher than the values reported by UNSCEAR (2000).

The findings in Table 2, indicating that the average radioactive activity in soil samples decreases in the order $^{40}\text{K} > ^{232}\text{Th} > ^{226}\text{Ra}$, are consistent with the known geochemical concentration of these radionuclides. Potassium is a common element in soil and rocks, constituting about 2.4% of the Earth's crust by weight [45]. The results show that the fluctuations of the three radionuclides in soil samples depend on many factors. Soils derived from different parent materials (igneous, sedimentary, or metamorphic rocks) will have varying concentrations of these radionuclides. Regional geology, including the presence of uranium, thorium, and potassium-rich rocks, affects soil radioactivity.

Principal component analysis

In the present work, the Principal Component Analysis (PCA) method was used to reveal the influence of soil physicochemical properties on the concentration of each

radionuclide. This analysis underscores how soil characteristics significantly impact the concentration of radioactive nuclides, with distinct relationships observed between different radionuclides and specific soil texture. Figure 2 shows that the soil physicochemical factor does not very much in the coordinate system formed by first factor (F1) and second factor (F2). The results show that the dependence of ^{226}Ra , ^{232}Th and ^{40}K on the soil properties is clearly demonstrated. The F1 and F2 axes are the first two principal components, responsible for explaining most of the variance in the data, accounting for approximately 73–78%. Among them, F1 plays the major role, contributing the largest proportion (41–46%), reflecting the factor with the strongest influence on the distribution of the data. Meanwhile, F2 contributes less (31–34%), but still plays an important role in clarifying the variation in the data. Clay and OC have an inverse relationship with sand and pH, indicating that soils rich in clay and organic carbon tend to have less sand and lower pH, and vice versa. Silt plays an intermediary role but alters the relationship between different PCA plots. Clay and organic-rich soils usually have better water retention capacity, leading to a lower pH (more acidic soils). In contrast, sandy soils tend

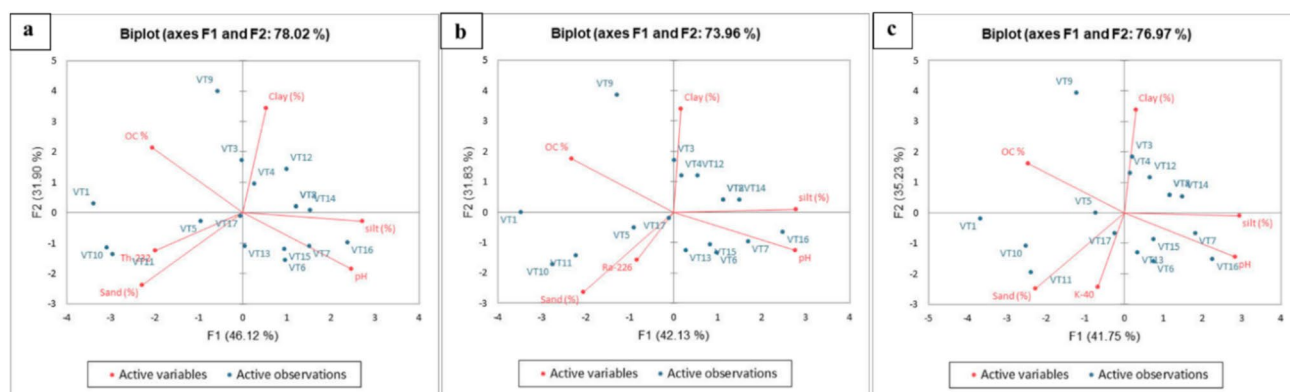


Fig. 2 Loading Component plot between the soil physicochemical properties and concentration of natural radionuclides

to have high permeability, less water and nutrient retention, resulting in a higher pH (more alkaline soils) [16, 46]. Silt, which has a grain size smaller than sand but larger than clay, has a moderate ability to retain water and nutrients. Therefore, when silt content changes, it can alter the relationship between the other factors.

For ^{232}Th , the F1 explained 46.12% of the total variance and is primarily influenced by soil pH, OC, sand and silt with squared cosines of 0.621, 0.434, 0.531 and 0.749 respectively. The F2 explained 31.90% of the total variance and is mainly associated with clay (square cosine of 0.829). F3 is mainly effected by ^{232}Th with square cosin of 0.480 (Supplement Table S1). Figure 2a indicates that ^{232}Th has a strong correlation with sand and soil pH. Pearson correlation analysis (Table 3) shows a strong positive linear relationship with sand (correlation coefficient 0.5) and a moderate negative relationship with soil pH (correlation coefficient -0.29). This implies that areas with high sand content and low pH may contain more ^{232}Th and vice versa.

For ^{226}Ra , F1 is primarily influenced by soil pH, organic carbon (OC) content and silt content (squared cosines of the variables > 0.53). F2 is mainly contributed by clay content

(0.860) and sand (0.515) content. Together, these factors explained over 73.96% of the total variance. ^{226}Ra has a significant influence on F3 with a square cosine of 0.729 (Supplement Table S1). Figure 2b shows that ^{226}Ra is also related to sand, but less strongly compared to ^{232}Th . The Pearson correlation coefficient between ^{226}Ra and sand is 0.26, indicating a weak positive linear correlation between them.

For ^{40}K , two main components explained 76.97% of the total variance. The first principal component (F1) is primarily influenced by pH, sand, OC and silt with squared cosines of 0.707, 0.530, 0.457 and 0.764 respectively. The second principal component (F2) represents the remaining variance with squared cosines corresponding to ^{40}K (0.442), sand (0.464) and clay (0.856).

Overall, through the PCA chart, it can be seen that the first two principal components are linear combinations of the original variables and are arranged in descending order of variance. The presence of the three radioactive nuclides is mainly influenced by physicochemical factors such as sand, soil pH, organic carbon (OC), and clay. The soil structure, such as the proportion of sand, clay, and silt particles, determines the soil's ability to retain water and essential elements [46]. Soils with a high clay content have a large surface

Table 3 Correlation between radionuclides and soil physicochemistry

	^{226}Ra	^{232}Th	^{40}K	pH	OC (%)	Sand (%)	Silt (%)	Clay (%)
^{226}Ra	1.00							
^{232}Th	0.85	1.00						
^{40}K	0.59	0.63	1.00					
pH	-0.19	-0.29	0.04	1.00				
OC (%)	-0.04	0.20	-0.02	-0.77	1.00			
Sand (%)	0.26	0.50	0.39	-0.26	0.12	1.00		
Silt (%)	-0.09	-0.40	-0.04	0.64	-0.45	-0.66	1.00	
Clay (%)	-0.27	-0.30	-0.49	-0.25	0.26	-0.72	-0.05	1.00

The bolded value 0.85 indicates a strong positive linear correlation close to 1 between the two isotopes ^{226}Ra and ^{232}Th .

Table 4 Radiological hazards index in soil samples

Symbol	Radiological hazards index					
	D_{in} (nGy h ⁻¹)	D_{out} (nGy h ⁻¹)	E_{in} (mSv y ⁻¹)	E_{out} (mSv y ⁻¹)	ELCR _{in} (10 ⁻³)	ELCR _{out} (10 ⁻³)
VT1	143	75.8	0.70	0.17	2.45	0.61
VT2	114	60.1	0.56	0.14	2.15	0.49
VT3	253	134	1.24	0.31	4.77	1.09
VT4	105	55.8	0.51	0.13	1.98	0.45
VT5	217	115	1.07	0.27	4.10	0.94
VT6	86.0	45.1	0.42	0.11	1.62	0.37
VT7	208	110	1.02	0.26	3.93	0.90
VT8	114	60.1	0.56	0.14	2.15	0.49
VT9	56.4	29.7	0.28	0.07	1.07	0.24
VT10	348	184	1.71	0.43	6.57	1.50
VT11	373	198	1.83	0.46	7.05	1.61
VT12	163	84.6	0.80	0.20	3.07	0.70
VT13	290	153	1.42	0.36	5.49	1.25
VT14	96.3	51.0	0.47	0.12	1.82	0.41
VT15	66.3	34.5	0.32	0.08	1.25	0.29
VT16	186	97.6	0.91	0.23	3.52	0.80
VT17	196	103	0.96	0.24	3.70	0.84
Mean	177	93.5	0.87	0.22	3.32	0.76
Standard Error	21.8	11.6	0.11	0.03	0.41	0.09
Minimum	56.5	29.7	0.28	0.07	1.07	0.24
Maximum	373	198	1.83	0.46	7.05	1.61
World's average [27]	84	59	0.41	0.07	1.45	0.29

Mean: The average value of all data points in the dataset

Standard Error: A measure of the accuracy of the sample mean as an estimate of the population mean. It reflects how much the sample mean is expected to vary due to random sampling. A smaller standard error indicates a more reliable estimate

Min: The lowest value in the dataset

Max: The highest value in the dataset

World's Average: The global average value of the radiological hazards index, based on data compiled from multiple countries or regions by UNSCEAR (2000). It is commonly used as a reference point for comparing local or regional values

area, which helps retain more nutrients and organic matter, thereby affecting the movement of elements. Sandy soils typically have poor retention of elements and may lead to leaching of radioactive elements. In general, clay-rich areas tend to retain more radioactive elements than sandy areas, due to their larger surface area and higher adsorption capacity. However, the actual levels of radioactivity also depend on the geological origin and the presence of radioactive minerals. Research by Korobova (2014) indicates that radioactive nuclides are retained due to the aggregation of fine particles in the soil, forming water-resistant clusters that are less prone to dispersion or dissolution in water [13]. Kumar (2023) contradicts the current study by indicating that clay and OC exhibit significant positive correlations with ²²⁶Ra concentration, with clay playing a primary role in enhancing

radioactive content in soil [47]. Similarly, Vineethkumar (2024) and Dragovic (2011) observed strong negative correlations between the activities of radioactive nuclides and sand content, and positive correlations with clay content [17, 18]. Contrary to the common assumption that clay-rich soils retain more radioactive substances due to their high surface area and adsorption capacity, our study, through PCA analysis, revealed that areas with higher clay content and lower sand content actually exhibit lower levels of radioactivity. This anomaly may be attributed to the geological origin of the study area (located near a rare earth mine—[47]), or to weathering and leaching processes that remove radioactive materials from the surface layers. The rare earth deposit in the study is a lateritic eluvial–deluvial type, formed through the tropical weathering of REE-enriched granitoid rocks.

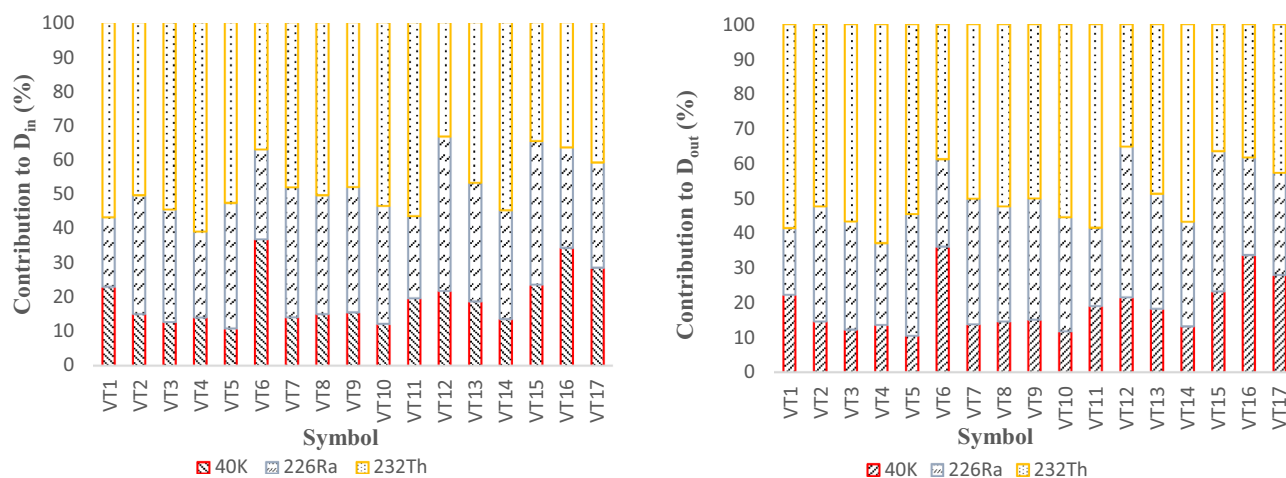


Fig. 3 The fraction contribution of ^{226}Ra , ^{232}Th and ^{40}K to the total absorbed gamma dose rate in the air

Rare earth elements released from minerals can be adsorbed onto clay particles within the soil horizon, with a portion of the material transported downslope to form deluvial accumulations.¹

Annual effective dose

The values of D_{in} , D_{out} , E_{in} , E_{out} , ELCR_{in} and ELCR_{out} are presented in Table 4. Specifically, the highest values of E_{in} (D_{in}) belong to sample VT11, with the values of 1.83 mSv y^{-1} (373 nGy h^{-1}), while the lowest values are observed in VT9 with 0.28 mSv y^{-1} (56.4 nGy h^{-1}). The average levels of E_{in} and D_{in} exceed the world average values (0.41 mSv y^{-1} and 84 nGy h^{-1}) [27] by a factor of 2. For the outdoor values, the lowest and highest values of E_{out} (D_{out}) are 0.07 mSv y^{-1} (29.7 nGy h^{-1}) in VT9 sampling and 0.46 mSv y^{-1} (198 nGy h^{-1}) in VT11 sampling. Compared to the global average (in seen Table 1), the E_{out} and D_{out} value are 3 and 1.5 times higher respectively. The results show that the gamma absorbed dose rate and annual effective dose values, both indoor and outdoor indicate, the potential for radiation exposure among the general population due to the activity concentration of natural radionuclides in in the study area.

The ELCR for indoor and outdoor exposure ranged (average) from 1.07×10^{-3} to 7.05×10^{-3} (3.32×10^{-3}) and 0.24×10^{-3} to 1.61×10^{-3} (0.76×10^{-3}). In practice, the overall ELCR index includes both the inhalation risk (due to radon) and ingestion risk, which are crucial factors in assessing the level of radiation hazard. However, in the current study, our focus is on evaluating the potential cancer risk associated with exposure to gamma radiation. Radon will be considered and incorporated into future studies. Similar to gamma dose rate and annual effective dose, the excess

lifetime cancer risk achieved its highest value in the VT11 site and its lowest in the VT9 site. Furthermore, the large D and E (in and out) values lead to ELCR values that are also 2 to 3 times larger than the global average for both indoor and outdoor exposure. Therefore, the ELCR values calculated in present work indicate that natural radiation may pose a risk of developing a mortal cancer [28]. A proportional relationship has been found between the received effective dose of radiation and the risk of developing cancer. For effective doses of 1 mSv y^{-1} , 10 mSv y^{-1} , 100 mSv y^{-1} , 1000 mSv y^{-1} , the excess lifetime cancer risk also increases by 0.004%, 0.04%, 0.4% and 4% respectively [48]. When compared to other studied worldwide, the values obtained for indoor and outdoor excess lifetime cancer risk in this work are higher than those reported in Iraq and Pakistan (1.63×10^{-3} and 0.54×10^{-3}) [28, 49].

The data provided in Fig. 3 emphasizes the varying contributions of different radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) to the indoor (D_{in}) and outdoor (D_{out}) gamma absorbed dose rates, as well as the indoor (E_{in}) and outdoor (E_{out}) effective dose rates. Depending on the location, the contributions of each radionuclide are different, the average contribution ratio of ^{232}Th is the highest with the value of 47.95%, followed by ^{226}Ra (32.68%) and ^{40}K (19.37%), accepted for site VT6, VT12, VT15 and VT16. When comparing the findings to the world average values determined by UNSCEAR, where the contributions to total variance of ^{232}Th , ^{226}Ra , and ^{40}K are 36%, 32%, and 32% respectively, it shows that the contribution of the Th series is higher than that of the U series [27]. Because the study area is situated near a rare earth mine containing a high thorium content ($0.157\% \text{ ThO}_2$), nearly 10 times greater than uranium ($0.016\% \text{ UO}_2$) [19, 35]. Notably, the total of annual effective dose average (indoor and outdoor) calculated is 1.09 mSv y^{-1} , more than twice as large as the global average effective dose from

¹ Geological division for radioactive and rare elements Vietnam.

external terrestrial radiation (0.48 mSv y^{-1}) [50, 51] and exceeding the effective dose of 1 mSv y^{-1} for the public [52]. It should be noted that the "effective dose of 1 mSv y^{-1} " is the natural radiation exposure by people living for a year in an area of low background radiation, which includes ingestion, inhalation of nuclides such as radon and cosmic radiation as well. There are actually 8 locations where the effective dose exceeds 1 mSv y^{-1} related to locations with high radionuclide activities of ^{226}Ra and ^{232}Th , including VT3 (1.55 mSv y^{-1}), VT5 (1.34 mSv y^{-1}), VT7 (1.28 mSv y^{-1}), VT10 (2.14 mSv y^{-1}), VT11 (2.29 mSv y^{-1}), VT13 (1.78 mSv y^{-1}), VT16 (1.14 mSv y^{-1}) and VT7 (1.2 mSv y^{-1}). This indicates that people living in the study area and surrounding areas may face enhanced risks from external radiation exposure.

Conclusion

The results of the average activity concentrations of radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in the area surrounding the mine were 62.2 ± 2.55 , 79.2 ± 3.31 , and $407 \pm 14 \text{ Bq kg}^{-1}$, respectively. The concentrations of ^{226}Ra and ^{232}Th are twice the global average, while the concentration of ^{40}K only shows a slight increase. The radionuclide concentrations exhibit a positive correlation with the clay content, a negative correlation with the sand content, and little or no correlation with other physicochemical properties of the soil. Principal component analysis (PCA) shows that for ^{232}Th , component F1 accounts for 46.12% of the total variance and is primarily influenced by soil pH, organic carbon (OC) content, sand, and silt, while component F2 accounts for 31.90% of the total variance and is mainly related to clay content. For ^{226}Ra , F1 is primarily influenced by soil pH, OC content, and silt, while F2 is mainly influenced by sand and clay content. For ^{40}K , component F1 contributes 41.75% of the variance and is primarily influenced by pH, sand, OC, and silt. The results of the PCA and Pearson correlation analysis indicate that the concentration of radioactive substances is influenced by all the physico-chemical factors of the soil, with a strong positive correlation with sand, a negative correlation with clay, and little or no correlation with the remaining physico-chemical properties of the soil. These findings contradict previous studies, raising the question of whether there are other factors that influence the changes in the physico-chemical properties of the soil, and consequently affect the concentration of radionuclides. Radiological hazards assessments show that the radiation exposure risk in the community is primarily due to the activity concentrations of natural radionuclides in the study area, with ^{232}Th being the most influential nuclide, accounting for nearly 50% of the total contribution from radionuclides. Based on these results, the study recommends that detailed

assessments of radiation exposure levels for the population in the study area should be conducted.

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Author contributions NTN, BTH, VNB, BVL, NVD, TVK designed the ideas for the study, planned for the experiment, derived the models and analyses the data, wrote the manuscript. All authors provided critical feedback, discussion and helped shape the research and analysis for the manuscript.

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Declarations

Conflict of interest The authors declare that they have no competing interests.

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