DOI: 10.31276/VJSTE.64(4).32-35

Study on the dispersion of radon (^{222}Rn) in geological objects in Bat Xat district, Lao Cai province, Northern Vietnam

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Received 4 January 2021; accepted 1 April 2021

Abstract:

The Bat Xat district in the Lao Cai province of Vietnam has enormous potential for resources of copper and rare earth minerals. In fact, exploration and exploitation activities for these minerals have brought about positive economic benefits to this area in recent years. However, this activity also brings about negative impacts on the environment and the surrounding population. The mineral deposits in this area contain a significant content of radioactive substances like uranium and thorium that radiate radon (^{222}Rn) and thoron (^{220}Rn). Therefore, the aim of this study is to investigate the distribution of radon as a function of depth of geological objects in the study areas. Results of this study showed that radon concentration has an exponential distribution with soil depth with R^2 ranging from 0.87 to 0.97. It was found that radon concentration in sand was higher than that in clay and soils mixed with gravels. It was also shown that the density and moisture content of the soil is inversely proportional to the diffusion length of radon in the soil.

Keywords: **dispersion, Lao Cai, RAD-7, radon concentration, radon in soil.**

Classification numbers: **2.1, 4.3**

Introduction

The radioactive decay of uranium and thorium chains and their progenies are widely distributed on Earth's surface and exist in all soil, rock, and mineral types at different concentrations. These radioisotopes affect living organisms. Once released into the environment, radioisotopes can enter the body through food, water, and air. In particular, radon exists in the form of a noble gas, so its dispensability into the environments could be on a wider scale than once thought. According to research from scientists around the world, radon is one of the leading carcinogens causing lung cancer [1-4]. Approximately 59% of the annual effective radiation dose that people receive is derived from radon. In the air, at least 70% of radon is released from the soil [5-8].

Radon migrates to the soil surface by different mechanisms and then emanates into the atmosphere [9]. The diffusion of radon in soil is a complex process. This process depends on many factors such as physical, chemical, geological, climate, and uranium content in the soil.

Measurement of the emission of radon from soil into the air has been used to assess the risk of radon exposure to the population. Further, the distribution of radon concentrations in soil has been successfully used in predicting Earth's kinetic phenomena such as earthquakes, karst caves, geological faults, and eruptions [10, 11].

The aim of this study is to investigate the dispersion of radon concentrations within geological objects into the environment in the Bat Xat district, Lao Cai province of Vietnam.

The Bat Xat district is delimited by the coordinates 22°51'÷22°55' N latitude and 103°68'÷103°74' E longitude. It is a complex terrain area as the central part contains low mountainous terrain surrounded by two sides of high mountains that are strongly dissected. The elevation change from 500-2,000 m creates many walls that are separated by river systems. The population distribution in the area is uneven. Most residents are concentrated in Muong Hum, including the Kinh and Day ethnic groups. Most of the Dao and H'mong converge into scattered villages in the high part of the terrain in the area and a few Dao people live in the low and fairly flat valley of Pieng Lao. In Sin Chai village, the Ha Nhi live right on the ore bodies of the radioactive Muong Hum rare earth mine and their houses resemble cottages with only one door; thus, homes are not ventilated and the accumulation of radon gas remains a threat to local residents [12].

Materials and methods

Geological characteristics of the study area

According to the survey results of the Geological Division for Radioactive and Rare Elements [12, 13], the geological features of the study area include the following formations: 1) Suoi Chieng (PP*sc1*), for which the main components are two-mica-granite shale, quartz-feldspar-mica schist, and thin layers of quartzite; 2) Sin Quyen (PP-MP*sq*), which contains metamorphic sedimentary formations, and the main components of biotite schist, mica schist, amphibolite, and quartzite; 3) Cha Pa Formation (NP*cp*),

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which is distributed in a small area in the southeast corner of the study area. The main components of the NPsp are quartz-sericite schist and marble; 4) Ban Nguon formation (D*1bn*) for which the main lithological compositions are shale, sericite, carbonate shale, quartzite, calcareous sandstone, occasionally scapolite, cordierite, and biotite horn; 5) Ban Pap formation (D*1-2bp*) with a petrographic composition including a thin layer of limestone, clay limestone, dark gray limestone, fine to medium grain, rock with block structure or thin layers, silicate limestone, silicic shale, and in some places alternating layers of quartz - sericite - chlorite schist; and 6) the quaternary formation (Q) in which the main compositions are sand, gravel, and clay (Fig. 1) [12, 13].

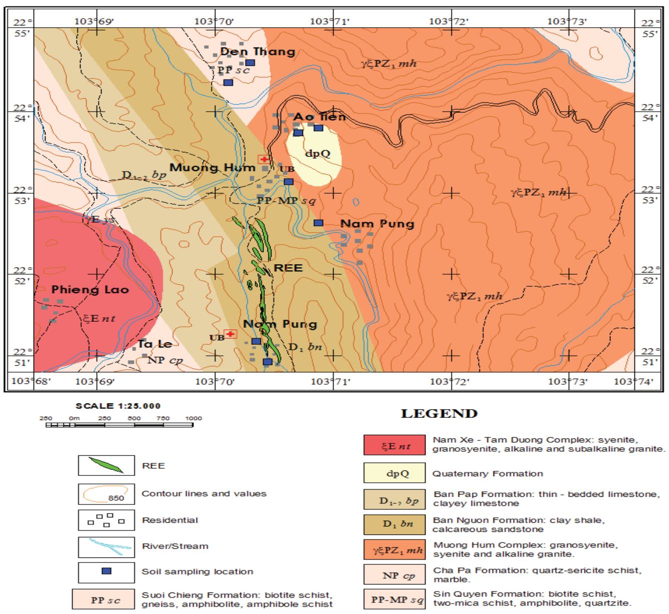

Fig. 1. Geological characteristics of the study area.

In the study area, there are copper and rare earth minerals that contain radioactive substances like uranium and thorium at high levels. The Muong Hum rare earth mine contains highly radioactive substances with uranium content ranging from 0.005 to 0.265% U_3O_8 and thorium content ranging from 0.106 to 0.188% ThO_2. The exploration and reserve assessment has been completed and will be exploited and processed in the near future [13]. The process of exploiting and processing radioactive minerals, excavating ores, exploiting land, collecting ore, crushing, and enriching strongly dispersing radioactive elements into the surrounding environment. There is a concentration of radioactive radon gas that affects the health of people living in rare earth mines and surrounding areas.

Therefore, in this study, an investigation of the diffusion of radon within the soil depth of different geological formations was conducted. The survey was carried out at 8 different locations in the Bat Xat district of Lao Cai province. These sites were chosen because they are typical geological formations in the region and where residents and farming land with food crops reside. The marks of 4 locations on the map in Fig. 1 (blue colour points) are described in Table 1.

Table 1. Survey locations of radon diffusion in soil.

No.	Geological object	Location	Symbol	X	Y
1	Suoi Chieng formation (PP_{sc1})	Muong Hum	SC.1	103°70'76"	22°54'19"
2		Muong Hum	SC.2	103°70'39"	22°54'20"
3	Sin Quyen formation (PP-MPsq)	Nam Pung	SQ.1	103°71'27"	22°53'10"
4		Nam Pung	SQ.2	103°71'52"	22°52'65"
5	Ban Nguon formation (D_1bn)	Den Sang	BN.1	103°71'11"	22°51'10"
6		Den Sang	BN.2	103°71'01"	22°51'33"
7	Quaternary formation (Q)	Sang Ma Sao	DT.1	103°71'54"	22°53'70"
8		Sang Ma Sao	DT.2	103°71'34"	22°53'64'

Methods

Determination of radon distribution in soil: The radon concentration was determined at each location using punching equipment at depths ranging from 30 cm to 120 cm. Measurements were carried out in the field using the dedicated radon measuring system RAD-7 shown in Fig. 2. The device consists of a hemispherical chamber for air coated with a conductive layer. A silicon semiconductor detector was installed at the centre of the chamber. The measuring system consists of a gas pump that was connected to a gas receiver made with holes that allow gas to be taken in from underground. At the beginning of the measurement, the gas pump takes the gas containing radon at the position where the gas receiver is inserted into the RAD-7 chamber. Decaying radon produces the radionuclides ^{218}Po and ^{214}Po inside the chamber. Under an electric field, ^{218}Po and ^{214}Po are led to the detector in the centre of the hemisphere. Then, the RAD-7 converts the alpha energy imparted from these radionuclides into electrical signals [14-16]. The time taken for each measurement was 30 min.

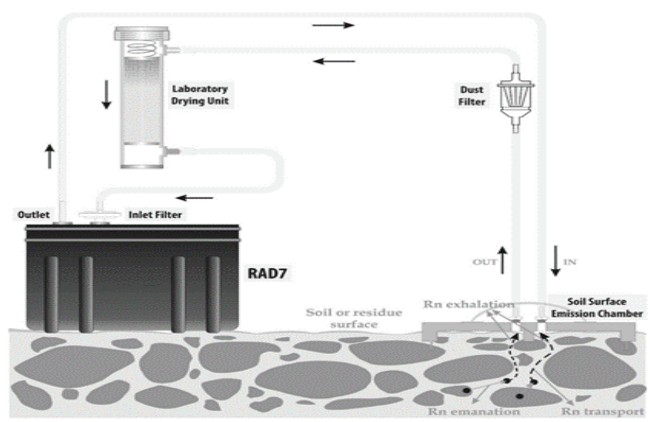

Fig. 2. A schematic layout of the installation used for radon distribution within soil depth.

In order to obtain the radon concentration as a function of soil depths, the diffusion of radon in soil was assumed to obey an exponential law as follows [5, 7]:

$$D\frac{d^2C(z)}{d^2z} - \lambda C(z) + \lambda C_\infty = 0 \qquad (1)$$

where C(z)(Bq.m^{-3}) is the radon concentration in soil pores at depth z (cm); C_∞(Bq.m^{-3}) is the radon concentration at infinite soil depth; D is the radon diffusion coefficient in soil (m^2.s^{-1}); λ is the decay constant of radon (s^{-1}).

The solution of Eq.(1) is

$$C(z) = C_\infty + Be^{-\sqrt{\frac{\lambda}{D}}z} + Ae^{\sqrt{\frac{\lambda}{D}}z} \quad (2)$$

where A and B are constants; the radon concentration at an infinite depth of soil (C_∞) is constant for a single homogeneous soil layer and is determined by the following equation:

$$\frac{dC}{dz} = 0 \ (z = \infty) \quad (3)$$

Applying boundary conditions to Eq.(2), one obtains an expression for C(z) as follows:

$$C(z) = C_\infty + Be^{-\sqrt{\frac{\lambda}{D}}z} = C_\infty + Be^{-\frac{z}{L}} \quad (4)$$

where L (cm) is the diffusion length of radon in soil.

Radon concentration in soil at each site was determined at different depths (C(z)). The variation of radon concentration (C(z)) by depth (z) was fit according to the exponential law (Eq.4) and the coefficients C, B, and L were derived from this simulation in Table 3.

Determination of soil properties: Soil samples collected in the field were brought to the laboratory to determine some soil properties such as density, water content, and particle size.

The samples were weighed for mass determination, then dried for 6 h at 300°C. Soil moisture and soil density are determined by the following Eqs. (5) and (6):

$$w = \frac{m_w - m_d}{m_d} (\%) \quad (5)$$

$$\rho = \frac{m_d}{V} (g/cm^3) \quad (6)$$

in which w (%) is the moisture content of the sample; m_w(g) is the wet weight of the soil sample; m_d (g) is the dry weight of the soil sample; and V (cm^3) is the volume of the soil sample.

Soil particle size, moisture content, and soil density were determined at the laboratory of the Soil and Fertilizers Research Institute. Soil particle size directly influences radon emission and radon distribution in soil. If uranium is evenly distributed within the spaces in between soil grains, then the particle size is smaller and the radon emission is higher and vice versa [10]. However, when the soil particle size is greater than 0.1 mm, the emission is low and nearly equivalent to other particle sizes [10].

In this study, we use the following QA/QC procedures: sampling (sampling location, date), sample transportation (keeping sample storage and storage requirements), handling and laboratory sample preparation, analysis, control and calibration of analytical instruments, and sample control analysis.

Results and discussion

The profile of radon concentrations by soil depth is presented in Table 2 and depicted in Figs. 3 and 4. For each geological formation (object), the profile was experimentally determined at two points.

Table 2. Radon concentrations within soil depth.

Depth (cm)	Radon concentrations ($Bq.m^{-3}$)							
	Suoi Chieng formation		*Sin Quyen formation*		*Ban Nguon formation*		*Quaternary formation*	
	SC.1	*SC.2*	*SQ.1*	*SQ.2*	*BN.1*	*BN.2*	*DT.1*	*DT.2*
20	259	313	498	641	399	433	367	531
40	726	802	1201	986	969	1151	1094	1301
60	1078	1139	1648	1256	1432	1636	1694	1895
80	1344	1372	1931	1468	1808	1964	2190	2353
100	1545	1533	2111	1634	2115	2185	2600	2706
120	1696	1644	2225	1764	2365	2334	2939	2979
140	1810	1721	2297	1865	2568	2436	3219	3189

With the results of Table 2, the diffusion expressions (Eq. 4) with C_∞, B, and L parameters for different geological formations are derived and presented in Table 3. In Table 3, the soil density and soil moisture in each formation are also presented.

Table 3. Diffusive equation constants of radon for different geological formations in Bat Xat along with soil density and moisture of the formation.

No.	Geological object	Diffusive equation	Diffusive length, L (cm)	Soil density, ρ ($g.cm^{-3}$)	Soil moisture, w (%)
1	Suoi Chieng formation ($PPPP_{sc1}$)	$C(z)=2{,}161-2{,}521e^{-z/71}$	71	2.64	15.9
2		$C(z)=1{,}892-2{,}287e^{-z/54}$	54	2.66	15.1
3	Sin Quyen formation (PP-MPsq)	$C(z)=2{,}423-3{,}132e^{-z/44}$	44	2.54	17.8
4		$C(z)=2{,}134-2{,}032e^{-z/82}$	82	2.61	16.9
5	Ban Nguon formation (D_1bn)	$C(z)=3{,}454-3{,}754e^{-z/97}$	97	2.08	10.1
6		$C(z)=2{,}546-3{,}376e^{-z/51}$	51	2.11	12.5
7	Quaternary formation (Q)	$C(z)=4{,}554-5{,}065e^{-z/98}$	105	1.49	11.7
8		$C(z)=3{,}897-4{,}364e^{-z/77}$	77	1.54	10.1

As seen in Table 2, the diffusion length of radon in the soil at the survey locations ranged from 44 cm to 105 cm and averaged 72.4 cm. This result was in good agreement with those found for soils in Japan and Iran [7, 8].

The radon diffusion length of a maximum of 98 cm in soil of the study area implies that the amount of harmful radon emission into the air near the local population could mainly originate from depths less than 100 cm. Radon concentrations at the depth of 100 cm ranged from 591 $Bq.m^{-3}$ to 3,219 $Bq.m^{-3}$. The International Atomic Energy Agency (IAEA) has recommended that the radon concentration in soil could affect human health with a high risk if it exceeds 30,000 $Bq.m^{-3}$ [17]. The results of this study show that in the survey locations, radon emissions are within the permissible limit.

The concentration distribution by depth fits an exponential law as the correlation value R2 ranged from 0.87 to 0.97 (Figs. 3 and 4). The discrepancy between experimental data and theoretical calculation is due to the fact that almost all surveyed sites were influenced by human activities and the soil was no longer primitive. Soil samples from the Ban Nguon and quaternary formations had a higher concentration of radon compared to those in soil samples from the Suoi Chieng and Sin Quyen formations. The reason for this

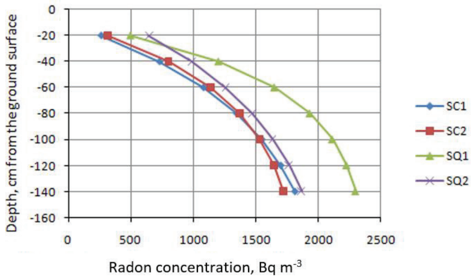

Fig. 3. The profile of radon concentration in Suoi Chieng and Sin Quyen formations.

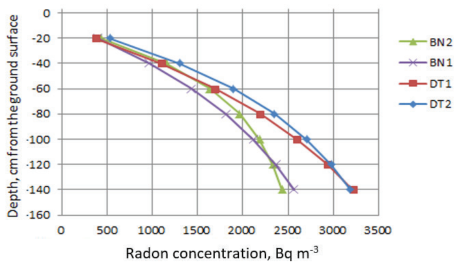

Fig. 4. The profile of radon concentration in Ban Nguon and quaternary formations.

is that the soil from the Ban Nguon and quaternary formations was weathered from rocks containing a high content of uranium. Results of this study also show that radon in the soil from the study area has a small diffusion length. This means that the diffusion capacity of radon into the air is low such that the concentration of radon in the soil is high. In addition, the distribution of radon concentration in the soil is highly dependent on the distribution of uranium content in the soil particles [10].

The dependence of the diffusion length on soil density and soil moisture are shown in Table 2. Results in Table 2 show that the diffusion length of radon in soil is inversely proportional to the density and moisture content of the soil ($R^2<0.5$) due to high soil density and moisture hinders the diffusion of radon (Figs. 5A and 5B). These preliminary findings could be explained by the fact that the high density and moisture of the soil hinder the diffusion of radon molecules. However, the diffusion of radon in soil also depends on several other factors such as soil composition, temperature, weather, etc., which need to be further investigated in the future.

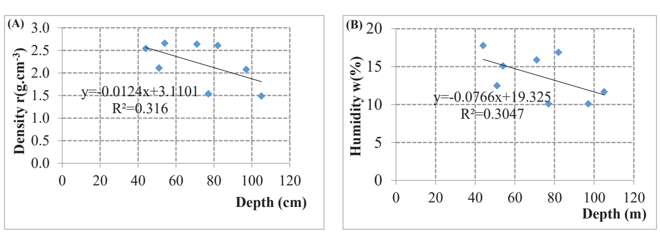

Fig. 5. The dependence of radon diffusion lengths on the (A) density and (B) moisture in the soil.

Conclusions

The distribution of radon concentration by depth in the surveyed soil samples obeyed the exponential law with values of correlation coefficient R2 ranging from 0.87 to 0.97. This shows that the survey sites have no risk of geological variation. The diffusion length of radon in clay is quite small compared to sandy soils and soils containing a lot of gravel. This is because clay samples have a relatively large soil density and water content.

The study results also show that the density and moisture content of the soil is inversely proportional to the diffusion distance of radon in the soil.

The radon concentration in the surveyed soil samples is within safe limits set by IAEA. The diffusion length of radon in the soil samples from the study area are smaller than that from other parts of the world. The radon diffusion length is greater for soil samples with low density and moisture content. Different soil samples have radon concentrations and diffusion lengths that depend on the origin of the formation, its composition, soil structure, temperature, humidity, and weather, etc.

COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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