Radon concentrations and forecasting exposure risks to residents and workers in rare earth and copper mines containing radioactivity in northwest Vietnam

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Abstract:

Radon and its isotopes are inert gases as they do not interact with any chemical compounds. Compared with thoron (²²⁰Rn) and radon-219 (²¹⁹Rn), the risk of radioactive exposure of radon-222 (²²²Rn) is very high due to its long half-life of 3.8 d, while the half-life of ²²⁰Rn is 55 sec and of ²¹⁹Rn is 4 sec. As a gas, radon can escape from the surfaces of ore, minerals, and rocks, then dissolve into groundwater and move very far from the formation site. While all these radioisotopes emit alpha radiation, Rn-222 is the most important as it is the main factor behind dangerous doses to the respiratory tract that are harmful to human health. Survey results of radon concentration in the air and retrospective data (from 2017 to 2019) on the health of residents and workers near and in the rare earth mines Dong Pao and Muong Hum, as well as the Sin Quyen copper mine, illustrated the health characteristics of the people involved in the northwestern mineral mines (Lao Cai - Lai Chau) that are exposed to radon. At the Dong Pao and Muong Hum rare earth mines, as well as the Sin Quyen copper mine, residents and workers were exposed to high concentrations of radon gas and thus developed some related illnesses such as respiratory, urological, digestive, genetic, and neurological diseases. Assessing the risk of pulmonary tuberculosis and estimating the average death rate from lung cancer with radon exposure shows that, in the surveyed area, the risk value is high (0.046) compared to other regions of Vietnam. However, it is within the limits allowed by the United States Environmental Protection Agency (EPA).

Keywords: lung cancer, radioactive, radon, radon exposure, risk level.

Classification numbers: 4.2, 5.3

Introduction

Radon gas is present in almost every part of the earth's crust as it escapes from the soil, finding its way through cracks, holes, faults, and groundwater, then enters the air by diffusion and convection. Radon exists in high concentrations in radioactive mineral mines, in houses, especially in closed rooms such as a bedroom or office; and in construction materials. This type of gas is classified by international organisations such as the Centers for Disease Control, the American Lung Association, and the United States Environmental Protection Agency (EPA), which all have radon gas listed as a carcinogen along with an information on its effects on human health.

The main danger of radon to health is from the exposure of alpha radiation during breathing and eating. Radon has also been associated with many deaths from lung cancer and is suspected to be associated with several other types of cancer such as leukemia, melanoma, kidney cancer, and some cancers of children. Epidemiological investigations have shown that radon can enter the body and mix with fat and blood cells in the same way that oxygen enters the blood [1-5]. One result is the accumulation of fat cells in bone marrow. In essence, radon can enter the human body and leave behind unpredictable consequences [6, 7]. Among the types of cancer caused by radon, lung cancer is considered to be the most dangerous because it holds the highest number of deaths [6].

Among these lethal risks, radon is estimated to cause about 21,000 (13.4%) deaths from lung cancer each year across the United States, more than all other risks posed to humans. This shows that the danger from radon in the home is very large [6].

Radon exposure does not cause acute illness, nor irritation, nor any early warning signs compared to other common environmental risks. Nonetheless, concentrated radon exposure increases the risk of lung cancer, especially

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in smokers. This risk increases with a person's radon level, length of exposure, and the amount of cigarettes smoked [6, 7].

Some studies show that radon is a related cause of leukemia, skin cancer, melanoma, kidney cancer in children, and some other cancers. Studies are based on statistical analysis of indoor radon and the extent of cancer effects [7]. The main damages caused by chronic exposure to radon are lung cancers (usually arising from bronchi), lung fibrosis, chronic obstructive pulmonary disease, pneumoconiosis, and overall respiratory damage.

Therefore, the determination of radioactive aerosol content caused by radon is very important for the purpose of monitoring and warning of the risk of lung cancer to communities near mineral mines, in mining areas, in houses and especially in bedrooms and offices. According to the United States Environmental Law, the permissible level of radon in homes is <4 pCi.l⁻¹.year⁻¹, which is equivalent to 148 Bq.m⁻³.year⁻¹. According to the radiation safety standards of the International Atomic Energy Agency (IAEA), the annual radiation dose limit for workers is recommended not to exceed 20 mSv.year⁻¹ [8].

The content of this paper presents the results of radon gas concentration surveys and risk assessments of radon exposure from residents and workers near and in the Dong Pao and Muong Hum rare earth mines and the Sin Quyen copper mines.

Research subjects

Mineral geological characteristics of the study area

The northwest region of Vietnam is concentrated with many radioactive minerals with large reserves such as the Dong Pao, Muong Hum, and Nam Xe rare earth mines, and the Sin Quyen copper mine, etc. Currently, the abovementioned radioactive mines have been explored and exploited and the resulting minerals are processed there as well, thus, radioactive elements including radon (Rn-222) are perpetually being released into the environment [9].

The Dong Pao rare earth mine is bounded by the coordinates $103^{\circ}33$ 'E, $22^{\circ}18$ 'N, in the Tam Duong commune of the Tam Duong district, Lai Chau province. The Dong Pao rare earth mine houses a group of fluorcarbonites with the main mineral components being bastnaesite, parisite, baryt (or barite), and fluorite. The proportion of minerals can be divided into the following types of ore: rare earth $TR_2O_3 > 5\%$; rare earth - baryt TR_2O_3 0.5-30%; rare earth fluorite TR_2O_3 1-5%. Trusted resources of TR_2O_3 make up 645,000 tons, estimated resources are 7 million

tons; CaF_2 is 9.4 million tons; and $BaSO_4$ is 288 million tons (Fig. 1). The content of the radioactive mineral ThO_2 ranges from 0.002-0.054% with an average of 0.013% and the content of radioactive U_3O_8 ranges from 0.002-0.014% with an average of 0.007% [9-11].

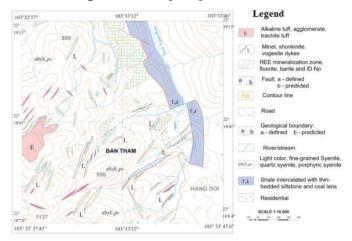


Fig. 1. Geological characteristics of Dong Pao rare earth mine [11].

The Muong Hum rare earth mine belongs to the Nam Pung and Muong Hum communes of the Bat Xat district, Lao Cai province, and is bounded by the coordinates of 22°51'-22°55'N and 103°68'-103°74'E. This is an area of complex terrain; the center along the northwest-tosoutheast direction is a low mountainous terrain that is surrounded by two sharply separated high mountain ranges (Fig. 2). According to the survey results of the Geological Division for Radioactive and Rare Elements [9, 11], this is a rare earth mine with large reserves with 175,000 tons of rare earth resources of TR₂O₃ on the spot and 37,500 tons of heavy rare earth resources. In addition to rare earth minerals, these areas also contain the radioactive substances U and Th with high content. For example, the uranium content U₂O₆ ranges from 0.007-0.0264% with an average of 0.0175% and the thorium ThO₂ content ranges from 0.0039-0.279% with an average reaching 0.176%.

The Sin Quyen copper mine is located in the Bat Xat district, Lao Cai province, with geographical coordinates 22°37′20′N, 103°45′50′E. The ore area is located on the northeastern flank of the Hoang Lien Son mountain range in the Lao Cai province, on the right bank of the Red river, right next to the Vietnam - China border, about 1-3 km from the Red river and 25 km from southeast of Lao Cai. The mountainous terrain extends from the northwest to the southeast. The geological characteristics of the area include the Suoi Chieng formation (PP_{sc}), the Sin Quyen formation (PP-MP_{sq}), the Ban Nguan formation (D₁bn), the Cha Pa formation (NPcp), and the Ban Pap formation (D₁bp) (Fig. 2). The copper content of this ore form

varies from 0.1-4.7% and its rare earth content is <1%. In primary copper ores, radioactive elements such as uranium, thorium, etc. have been detected. The content of $\rm U_3O_8$ ranges between 0.005-0.265% and the $\rm ThO_2$ content ranges between 0.006-0.03% [12].

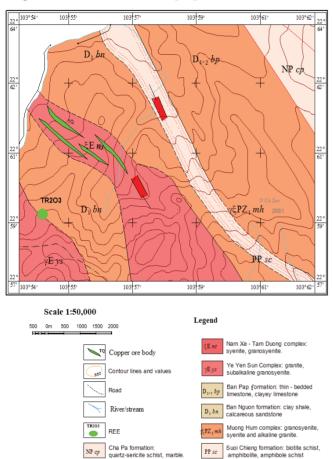


Fig. 2. Regional characteristics of Bat Xat district, Lao Cai province [12].

Research subjects

The surveyed subjects are residents and workers living and working near and in the rare earth mines of Dong Pao and Muong Hum and the Sin Quyen copper mines.

Research methods

- Collecting health data of people in mines and surrounding areas (outside the mines) over the last 3 years through retrospective methods at local health facilities.
- Reviewing periodic medical records of workers in the medical facilities of the mine with focus mainly on diseases related to the effects of radon. Diseases are statistically surveyed for research include respiratory diseases, digestive diseases, circulatory diseases, genetic diseases, and cancers.

- Surveying radon gas concentration in houses, workers' houses, workplaces, schools, sorting workshops, etc., to determine the internal radiation dose level caused by radon according to the following formula [13-15]:

$$H_{\bullet}(mSv.year^{1})=0.047\times C_{p,n}(Bq.m^{-3})$$
 (1)

where H_t is the internal radiation dose caused by radon in the air and $C_{Rn}(Bq.m^{-3})$ is the concentration of radon in the air.

- Assessing the level of radon exposure from the work process, especially at places with high radon content could be experienced by residents and workers living and working in rare earth mines (Dong Pao, Muong Hum) and the copper mine Sin Quyen, as well as other humans affected by radon radioactivity [6, 7, 16, 17].

Calculation of cumulative exposure: accumulated exposures are defined as all activity levels (WL) multiplied by the exposure time. In the exposure assessments, this cumulative exposure is calculated by exposure for 1 month (or 170 working hours). Cumulative exposure is calculated using the following formula [6]:

$$WLM = \sum_{i=1}^{N} (WL)_i \left(\frac{t_i}{170}\right)$$
 (2)

where WLM is cumulative exposure; $(WL)_i$ is the average concentration of radon and offspring during exposure; and t_i is the total exposure time with 1 WL= $C_{pn}(Bq.m^{-3})\times0.00027$.

The above formula is used for calculating cumulative exposures over time intervals with concentrations corresponding to those time periods. However, due to the limitations of research time and statistical data systems over a short time period, this article only intends to show the general method and calculation of the assessments for radon exposure in workers in different positions [6].

Average exposure rate value is estimated for one year:

$$EX = C \times \left[F \times 0.01 \text{WL} \left(\frac{\text{pCi}}{\text{l}} \right)^{-1} \right] \times 12 \text{ WLM} (\text{WLy})^{-1}$$
(3)

where EX is the average exposure rate estimated in a year (WLy)⁻¹; C is the average radon concentration (pCi.l⁻¹ or Bq.m⁻³); F is the coefficient of balance between radon and its products, e.g., F=0.6 in the mine.

At a concentration of 1 pCi.l⁻¹, the exposure to offspring is calculated by:

 $EX=1pCil^{-1}[0.6\times0.01WL (pCil^{-1})^{-1}]\times[12 WLM(WLy)^{-1}]=0.072 WLM/y (4)$

At a concentration of 1 Bq.m⁻³, the exposure of descendants of radon is calculated by the formula:

 $EX=1Bq.m^{-3}[0.6\times0.00027WL(Bq.m^{-3})^{-1}]\times[12WLM(WLy)^{-1}]=0.00194WML/y$ (5)

Results and discussion

Concentration of radon radioactive gas in surveyed areas

The results of radon emission survey at the Dong Pao and Muong Hum rare earth mines and the Sin Quyen copper mine over 3 years from 2017 to 2019 are as follows:

In the Dong Pao rare earth mine area, the radon concentration ranges from 15 Bq.m⁻³ to 648 Bq.m⁻³ with an average of 67.8 Bq.m⁻³, which is 1.8 times higher than the world average for radon concentration in a house (40 Bq.m⁻³) as measured by UNSCEAR 2000 [15]. The typical value of internal radiation dose at Muong Hum mine is 3.18 mSv.year⁻¹.

In the Muong Hum rare earth mine area, the radon concentration ranges from 35 to 856 Bq.m⁻³ with an average of 88.4 Bq.m⁻³, which is 2.4 times higher than the world average for radon concentration in a house (40 Bq.m⁻³) as measured by UNSCEAR 2000 [15]. The typical value of internal radiation dose at Muong Hum mine is 4.15 mSv.year¹.

At the Sin Quyen copper mine area, the radon concentrations in mining areas, sorting workshops, dump sites, and workplaces of officials and employees were valued from 15.7 to 476 Bq.m⁻³ with an average of 48.1 Bq.m⁻³, which is 1.3 times higher than the world average for indoor radon concentration (40 Bq.m⁻³) as measured by UNSCEAR 2000 [15]. The typical dose measured in the Sin Quyen copper mine is 2.26 mSv.year⁻¹.

Health characteristics of residents and workers in the survey areas

Statistics of disease over three years from 2017 to 2019 in residents and workers near and in the rare earth mines Dong Pao and Muong Hum and the Sin Quyen copper mines (Fig. 3), where radon gas concentrations are high, demonstrate mainly respiratory, circulatory, and digestive system diseases.

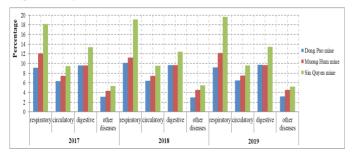


Fig. 3. Disease classification diagram of the Dong Pao and Muong Hum rare earth mines and the Sin Quyen copper mine.

The chart in Fig. 3 of disease classifications over the 3-year period of investigation shows diseases of the respiratory system, circulatory system, digestive system, and other diseases of residents living near the rare earth mines of Dong Pao and Muong Hum are not high. Residents living in this area mainly suffer from respiratory, circulatory, and digestive diseases. At the Dong Pao mine, the incidences of respiratory, circulatory, and digestive diseases by year are: 2017 (9.09; 3.37; 6.57%); 2018 (10.13; 3.45; 6.67%); and 2019 (9.13; 3.48; 6.75%) and at Muong Hum mine, they are: 2017 (12.12; 4.43; 6.61); 2018 (11.21; 4.46; 6.67%); and 2019 (12.17; 4.51; 6.63%). People classify circulatory weakness with major cardiovascular diseases such as limited hypertension and cardiac arrhythmia.

For the Sin Quyen copper mine, the proportion of workers suffering from respiratory, circulatory, and digestive diseases is higher than that of Dong Pao and Muong Hum mines over the 3-year investigation, which are, respectively, 2017 (17.18; 6.46; 10.37%); 2018 (18.08; 6.51; 11.42%); 2019 (18.67; 6.58; 11.41%), with circulatory diseases mainly presented as hypertension and arrhythmia.

Results of a 3-year disease situation survey at locations with high concentrations of radon gas such as field sites, transports, and screening plants are shown in Fig. 4.

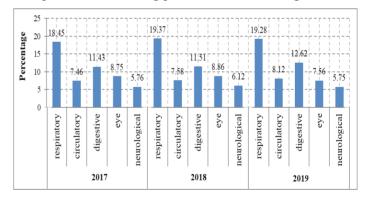


Fig. 4. Disease situation over 3 years (2017-2019) at training sites and sorting workshops.

In these locations, there have been many radon-related diseases such as respiratory, digestive, circulatory, eye, and neurological diseases. These diseases are most common in mining sites, ore transports, and sorting workshops. This may be due to the influence of many factors but the effect of exposure to relatively high radon content in the air is unavoidable.

Notably, in the results of the health examination of officials and workers of Sin Quyen Copper Company over the three years of survey, there were several workers with quite serious respiratory diseases: there were 12 cases of pneumonia, 28

cases of lung damage, 7 cases of tuberculosis, and 2 cases of collapsed lung(s). In 2017, the Company had 5 cases classified as poor respiratory health and 7 cases classified as poor health, which account for 2.15 and 1.8%, respectively. According to each working position in Table 1, it can be seen that the number of infected people is quite high at the mining, transporting, and recruitment sites. Among those working in the office, some people fell ill because they have had time to work in direct production positions. In 2018, the number of people with lung disease was the highest with 19 of the 42 people in the entire Company suffering from lung disease.

Table 1. Statistics of lung diseases by working positions at Sin Quyen copper mine.

No.	Partial	Total number of people infected	Total number of cases infected by year		
			2017	2018	2019
1	Office	10	3	4	3
2	Technical Department	3		2	1
3	Mining area	12	3	6	3
4	Shipping workshop	7	3	3	1
5	Electric workshop	2		2	
6	Recruitment Workshop	8	4	2	2
Total		42	13	19	10

Assess the exposure of radon to Sin Quyen copper mine workers

According to the results of the radon concentration survey in the Sin Quyen copper mine area, there no value exists that exceeds the permitted level by Vietnamese and world standards for workers. However, radon is a carcinogen with no harmful threshold, and, with long exposure time and other impact factors such as smoking, health care condition, etc., it can adversely affect worker health.

The radon concentration data at different working sites in the Sin Quyen copper mine will be used to calculate the exposure for workers with the theory that the workers of each location are exposed at this concentration during the working process until retirement. Calculation results are presented in Table 2.

Table 2. Assessment of exposure at work site.

Position	Exposure			
rosition	Bq.m ⁻³		WL	WLM.y1
Mining pits	431	11.64	0.110	0.828
The bottom of the pan	376	10.16	0.102	0.711
Load the transport vehicle	328	8.86	0.089	0.651
Waste dump	87.4	2.36	0.024	0.181
Recruitment area	136	3.67	0.037	0.254
Mechanical house	123	3.32	0.033	0.217
Office	35.3	0.95	0.009	0.062

The results of the average annual exposure assessment of workers at the mine site indicate that exposure is within 0.062-0.828 WLM.y¹. This level of exposure is relatively safe for worker health, but considering long-term exposure and smoking status, there may be some effects that need to be considered and assessed.

Assessing health risks due to radon exposure for officials and workers working at Sin Quyen copper mine

To assess the risk of radon exposure for officials and workers at the Sin Quyen copper mine, the authors used the risk calculation model under the guidance of EPA (2009) to use a single model instead of 2 models like BEIR VI (NAS) [6] because the two previously proposed models almost all depend on the age and time of exposure. EPA uses a concentration model for risk calculations because a concentration model can assess the health effects of exposure at levels that change over time.

In BEIR VI, the risk/WLM is 6.52×10^{-4} for the concentration model and is 4.43×10^{-4} for the time period model. The EPA has calculated the concentration model so that the risk/WLM will be equal to the geometric significance of these two values, i.e., 5.38×10^{-4} . The risk factor according to the concentration model is $\beta=0.0768\times(4.43/6.52)^{0.5}=0.0634$, and the risk/WLM is $5.38\times10^{-4}\approx(6.52\times10^{-4})\times(4.43/6.52)^{0.5}$ [6].

The concentration model indicates that the relative risk of excess exposure depends on the time the exposure was initiated, the age reached, and the rate of exposure (concentration) as follows [6]:

$$ERR = \beta \times (w_{5-14} + \theta_{15-24} \times w_{15-24} + \theta_{25+} \times w_{25+}) \times \phi_{ave} \times y_{z}$$
 (6)

where ERR is an assessment of the level of risk; β is the risk factor; $w_{_{5-14}}, w_{_{15-24}}, w_{_{25+}}$ are exposure at ages 5-14; 15-24, and 25 years or more, respectively; $\theta_{_{5-14}}; \theta_{_{15-24}}; \theta_{_{25+}}$ are the risk that is relatively dependent on the time the exposure is initiated; ϕ_{age} describes the dependence on the age achieved; for mine workers and for retired people at 55 years old, ϕ_{age} =1.0 and with retirement age of 60, ϕ_{age} =0.57; y_z is the classification from 1 for exposure <0.5WL to 0.11 for exposure >15WL, which describes the exposure speed dependence. For this calculation, since all WL values are <0.5, y_z =1.

Setting $\beta^* = \beta \phi_{agc}$ and using the parameters shown in Table 3, the authors estimate the parameters for the risk model [6] with the following equation for calculating the excess relative risk:

ERR=
$$\beta^* \times (w_{5.14} + 0.778 w_{15.24} + 0.51 w_{25+}) \times \phi_{ave}$$
 (7)

where β^* =0.0768 for age x<55y; β^* =0.0438 for age 55 y≤x<65y; β^* =0.0223 for age 65 y≤x<75y; and β^* =0.0069 for age x≥75y.

Table 3. Estimated parameters for concentration model [6].

Concentration model (β×100=7.68)			
Time of owners	$\theta_{15-24} = 0.78$		
Time of exposure	$\theta_{25+} = 0.51$		
	0.0768 for x<55y		
XVI O*	0.0438 for 55 y≤x<65y		
Where β*	0.0223 for 65 y≤x<75y		
	0.0069 for x≥75y		

In order to estimate the relative health risks for workers at different positions in the Sin Quyen copper mine, the authors make some assumptions:

- In direct labor positions, employees retired at the age of 55. For indirect working positions, employees retired at the age of 60.
- Workers maintain one position until retirement without changing.
- The average annual exposure to radon is applied for each position during the time the employee works until retirement.
 - Age to being work and exposure to radon is 25.

The results of risk estimation for workers at the Sin Quyen copper mine are presented in Table 4.

Table 4. Estimation of risks at job locations at Sin Quyen copper mine.

Position	Relatively risk		
Position	EER (%)		
Field workers	147	0.828	
Classification worker	119	0.711	
Workers loading and transporting vehicles	117	0.651	
Workers in the disposal area	35.2	0.181	
Workshop workers	42.6	0.254	
Mechanic	38.5	0.217	
Office staff	0.68	0.062	

Comment: the above calculation model is applied for calculating the risks over a long time and the radon exposure phase of different periods are carefully observed as well as the concentration in each period. However, in this paper, due to the short time of execution and the short time measurement of exposure calculations, the authors only determine health risks for a select number of working positions under exposure conditions at a measured average concentration.

Estimated average risk of death from lung cancer due to radon exposure of workers at Sin Quyen copper mine

The estimated average risk of death from lung cancer with life-long exposure to average radon exposure using the EPA formula is calculated as follows [6]:

$$ER=w\times t\times risk$$
 estimate (8)

where ER is the estimate of the average risk of death from radon exposure; w is the estimated average exposure value in a year (WLMy⁻¹); and t is the average life expectancy of a country.

Risk estimate = 5.38×10^{-4} /WLM for the entire population

 $=9.68\times10^{-4}$ /WLM who used to smoke

=1.67×10⁻⁴/WLM for non-smokers

Table 5 shows the estimation of the risk/WLM by gender and smoking status.

Table 5. Estimating risk/WLM by gender and smoking status [6].

Gender	Smoking category	Risk per/ WLM (10 ⁻⁴)	Average life expectancy (year) EPA	
	Smoking	10.6	71.5	69.4
Male	No smoking	1.74	72.8	70.7
	All men	6.40	72.1	70
	Smoking	8.51	78.0	74.2
Female	No smoking	1.61	79.4	75.6
	All women	4.39	78.8	75
	Smoking	9.68	74.2	70.8
General	No smoking	1.67	76.4	73
population	The whole population	5.38	75.4	72

To be able to estimate risks more accurately, it is assumed that, after retirement, workers will continue to live in this area for the rest of their lives. The average estimation of death risk is then written as [6]:

$$ER = (w_1 \times t_1 + w_2 \times t_2) \times risk \text{ estimate}$$
 (9)

where w_1 is the estimated average exposure rate for a year corresponding to the concentration measured at the workplace; w_2 is the estimated average exposure rate for a year corresponding to background concentration in the area of the Bat Xat district; t_1 is the time the employee worked at a position in the mine (male worker directly 35 years; indirectly 30 years; female worker less than 5 years in both positions); t_2 is the average life expectancy minus t_1 .

A representation of the average risk of death from lung cancer related to radon of 4 gender groups and smoking status at Sin Quyen copper mines is shown in Fig. 5. The results show that the risk of death in male smokers is 6 times higher than non-smokers, which is similar to a female smoker's risk that is 5 times higher than non-smokers.

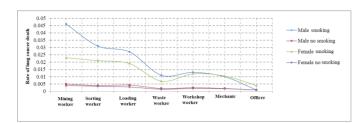


Fig. 5. Estimated average risk of death from lung cancer of several jobs at Sin Quyen copper mines.

Fig. 5 also shows that, in terms of exposure, mining workers, ore loaders, and sorting workers face the highest risk of death from lung cancer, especially smokers. Therefore, in the long term, the solution of transferring workers to different working positions can be helpful to reduce radon exposure for this group of workers.

Conclusions

Some health characteristics and diseases of residents and workers at the radioactive mineral mines in northwest Vietnam related to radon exposure are not really definitive.

At the Sin Quyen copper mine, where officials and workers are exposed to relatively high levels of radon (where the radon concentration in the air is up to 431 Bq.m⁻³), there have been some related diseases such as respiratory, circulatory, digestive, eye, and neurological diseases. In particular, there were patients with lung damage.

The risk estimates for tuberculosis and the average estimate of lung cancer deaths with radon exposure in the survey area are quite high for smokers (0.046) compared to other regions of Vietnam.

COMPETING INTERESTS

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

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