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PAPER

Radiological risk assessment of outdoor ^{222}Rn and ^{220}Rn around rare earth element and uranium mines from northern Vietnam

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Abstract

The outdoor ^{222}Rn and ^{220}Rn concentrations at 320 sampling points at 1 m above the ground in different sites surrounding rare earth element (REE) and uranium mines from northern Vietnam were measured using the RAD7. Results showed that ^{222}Rn concentrations were always higher than ^{220}Rn concentrations with large variation ranges from 25.7 to 573 Bq m^{-3} and from 18.5 to 385 Bq m^{-3}, respectively. The high correlation between ^{220}Rn and ^{228}Ra concentrations in surface soil of the studied sites were observed. The highest ^{220}Rn and ^{222}Rn concentrations are found at the sampling points of the REE NX-Lai Chau site. The ^{220}Rn and ^{222}Rn activities surrounding the REE mines were found to be higher than those surrounding the uranium mines. The average annual committed effective doses originated from the inhalation of ^{220}Rn and ^{222}Rn outdoor concentrations is about five times higher than the worldwide average value.

1. Introduction

Natural radionuclides from different sources in the environment, even in our bodies can originate from weathering of the earth's crust (rocks, soils, ores), food consumption, mining activity, and fertilizer materials (Azeez *et al* 2019, Querfeld *et al* 2019, Takagi *et al* 2019, Van *et al* 2020a, 2020b, 2020c, Loat *et al* 2021, Van *et al* 2021a, 2021b, 2021c, 2021d). In the air, radionuclides such as ^{222}Rn and ^{220}Rn originate from the alpha decay of ^{226}Ra in the ^{238}U series and ^{228}Ra (^{224}Ra) in the ^{232}Th decay series, respectively (Omori *et al* 2016). The half-lives of ^{222}Rn and ^{220}Rn are 3.8 d and 55.6 s, respectively. The lung cancer risk from exposure to ^{222}Rn radioactive and its decay products through inhalation are well known (Tomášek and Plaček 1999, Al-Zoughool and Krewski 2009, Clement *et al* 2010). One of the main causes of lung cancer in the population is the inhalation of ^{222}Rn and ^{220}Rn (UNSCEAR 2000). Therefore, an investigation on ^{222}Rn and ^{220}Rn dispersion in the environment can be used to assess the population exposure to radiation and to estimate the radiological hazard.

The concentrations and radiation doses due to inhalation of ^{222}Rn and ^{220}Rn have been also extensively investigated worldwide (Iida *et al* 1996, Chung and Tokonami 1998, Wang 2002, Bochicchio *et al* 2003, Oikawa *et al* 2003, Phon *et al* 2015, Omori *et al* 2016, Ayres da Silva *et al* 2018, Kojo *et al* 2021). In general, the ^{222}Rn and ^{220}Rn concentrations in the soil and air depend on the types of rocks, their migration from rock to soil, the release of ^{222}Rn and ^{220}Rn from the soil and rock to the atmosphere, and the weathering characteristics, it therefore varies from one site to another.

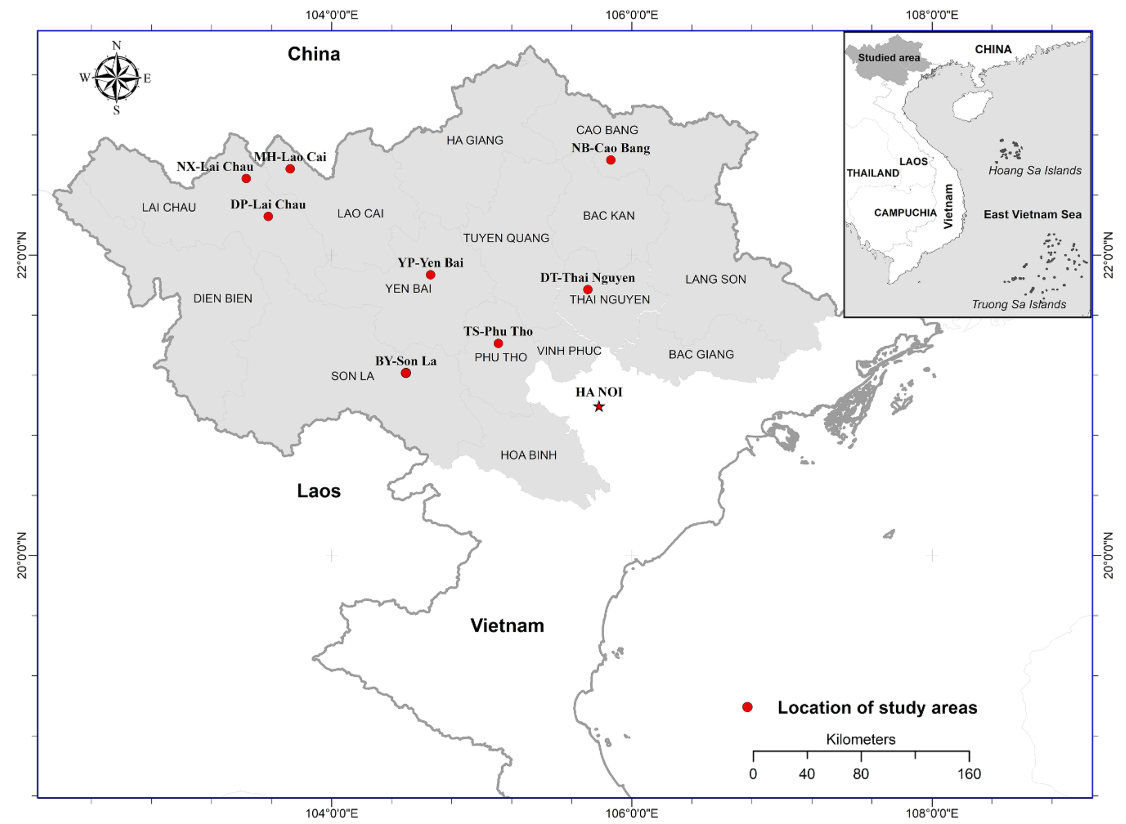

Figure 1. Eight sites where measurements of ^{222}Rn and ^{220}Rn concentrations were performed.

There are many mines in northern Vietnam, which contain a high content of natural radionuclides such as rare earth mines in NX, DP (Lai Chau), MH (Lao Cai), YP (Yen Bai); polymetallic mines (contain high uranium concentrations) in NP (Thai Nguyen) and uranium ore in BY (Son La), TS (Phu Tho), NB (Cao Bang) (figure 1). These mines were recently reported to have a high radioactive background by the Vietnam Geological division for Radioactive and Rare elements. The ^{226}Ra and ^{228}Ra high activity concentrations in surface soil samples of those areas were earlier reported in Van *et al* (2021c) (table 1). Therefore, the surrounding environments and local communities of those areas can be exposed to high ^{222}Rn and ^{220}Rn concentrations. In this study, the baselines of natural radiological hazard assessment of ^{222}Rn and ^{220}Rn concentrations in those areas surrounding rare earth element (REE) and uranium mines are presented. Results will provide the baseline data to evaluate the radioactive pollution during the exploitation of these mines and supply information for local stakeholders to manage the impacts of the radioactivity risks.

2. Measurement and methods

2.1. The sampling points

Forty sampling points near a residential area were chosen to measure the ^{222}Rn and ^{220}Rn concentrations in each of eight studied sites to give a total of 320 sampling points (figure 1). Each measurement was continuously conducted during four days in weather conditions of no rain, humidity of 70%–85%, a wind speed of 0.3–1.5 m s^{-1}, and temperature of 22 °C–30 °C. The distance from the measurement points to the centre of the deposits in eight studied sites depends on the area of the mines, but all the sampling points are around the studied mines. Each measurement was performed at 1 m above the soil ground surface, which related to the calculation of dose rate and air inhalation for humans living or working the studied areas.

2.2. Measurement of ^{222}Rn and ^{220}Rn concentrations by a RAD7

For measurement *in situ*, the ^{222}Rn and ^{220}Rn concentrations were measured using a RAD7 (A radon Detector from DURRIDGE Company Inc). The RAD7's air pump system pumped air through a chamber with a flow rate of ∼0.5 (dm^3 min^{-1}). Two hours of air pumping for each of the sampling points was undertaken counts. The most significant background effect is that it is not possible to distinguish the current contribution to the count rate from ^{222}Rn progeny and traces of ^{222}Rn and ^{220}Rn remaining from previous

Table 1. Outdoor ^{222}Rn and ^{220}Rn concentration in northern Vietnam.

Type of mines	Locations	Value	Activity (Bq m^{-3}) in this study		Activity (Bq kg^{-1}) (Van et al 2021c)	
			^{222}Rn	^{220}Rn	^{226}Ra	^{228}Ra
REE mines	NX-Lai Chau	Range	25.7–573	18.5–385	540–790	750–990
		Average	116	62.3	660	890
		Skewness	2.72	3.08		
		Kurtosis	10.8	9.85		
	DP-Lai Chau	Range	17.0–172	8.8–87.5	150–210	180–270
		Average	91.3	35.7	180	220
		Skewness	0.01	1.30		
		Kurtosis	−1.08	3.03		
	MH-Lao Cai	Range	15.6–144	12.5–109	360–490	550–1350
		Average	56.5	45.9	430	850
		Skewness	0.88	1.01		
		Kurtosis	1.67	1.04		
	YP-Yen Bai	Range	11.4–38.6	14.1–108	25–150	53–230
		Average	23.1	35.7	76	140
		Skewness	0.55	2.36		
		Kurtosis	−0.47	5.94		
Uranium mines	BY-Son La	Range	6.9–21.7	11.9–32.5	13–160	21–250
		Average	14.4	22.4	60	11
		Skewness	0.18	0.07		
		Kurtosis	0.08	−0.70		
	TS-Phu Tho	Range	11.3–54.7	17.7–105	130–190	330–480
		Average	28.0	42	150	390
		Skewness	0.72	1.09		
		Kurtosis	0.69	1.13		
	DT-Thai Nguyen	Range	6.8–79.7	13.6–31.9	54–130	53–100
		Average	27.2	22.2	100	71
		Skewness	2.10	0.47		
		Kurtosis	6.45	−0.46		
	NB-Cao Bang	Range	6.1–156	15.3–46.0	400–740	85–130
		Average	45.9	24.1	590	100
		Skewness	1.46	1.00		
		Kurtosis	1.61	1.75		
Overall		**Minimum**	**14.4**	**22.2**		
		Maximum	**116**	**62.3**		
		Overall average	**28.1**	**10.3**		

measurements (DURRIDGE Company Inc. 2017). To avoid the decreased detection efficiency of the RAD7 due to the relative humidity, a desiccant was used all the time to dry the air stream prior to entering the RAD7. The instrument was calibrated annually using inter-comparing ^{222}Rn chambers.

2.3. Evaluation of radiological hazard indices

2.3.1. Annual effective dose (AED)

The AED originated from the inhalation of ^{222}Rn and ^{220}Rn outdoor dwellings is calculated using:

$$\text{AED}\ \left(\text{mSv.y}^{-1}\right) = C \times F \times t \times K \qquad (1)$$

where, C is the average ^{222}Rn or ^{220}Rn concentration outdoors (Bq m^{-3}), F is the outdoor equilibrium factor for ^{222}Rn and its progeny or for ^{220}Rn and its progenies ($F = 0.6$ and $F = 0.003$ for ^{222}Rn and ^{220}Rn, respectively); t is annual time spent outdoor ($t = 1760$ h); K = dose conversion factors ($K = 9$ nSv Bq^{-1}.h.m^3 and $K = 40$ nSv Bq^{-1}.h.m^3 for ^{222}Rn and ^{220}Rn, respectively) (UNSCEAR 2000).

3. Results and discussion

3.1. ^{222}Rn and ^{220}Rn activity concentrations

The measured activities of ^{222}Rn and ^{220}Rn in eight sites in northern Vietnam are presented in table 1. The distribution of ^{222}Rn and ^{220}Rn activities mostly showed a slight tail relative to a normal distribution (kurtosis <3), with an exception of Rn in NX-Lai Chau (kurtosis = 10.8), DT-Thai Nguyen (kurtosis = 6.45) and for Tn in NX-Lai Chau (kurtosis = 9.85). The highest variations of ^{222}Rn and ^{220}Rn concentrations were observed in the REE mine of NX-Lai Chau, ranging from 25.7 to 573 Bq m^{-3} and from 18.5 to 385 Bq m^{-3}, respectively. The average ^{222}Rn and ^{220}Rn concentrations were several times higher than the worldwide outdoor average of 10 Bq m^{-3} (UNSCEAR 2000). The result was similar to the previous report in the rare earth mine in Lai Chau province (Phon *et al* 2015). The average ^{222}Rn and ^{220}Rn concentrations in NX-Lai Chau reached the highest value among the eight studied sites. This could relate to the exploitation activities of the REE in this area that led to release the ^{222}Rn and ^{220}Rn into the surrounding environment and this may have given rise to the large variation of the radionuclide activities. The lowest ^{222}Rn and ^{220}Rn concentrations ranged from 6.9 to 21.7 Bq m^{-3}, with an overall mean of 14.4 Bq m^{-3} for the uranium mine in BY-Son La. The lowest ^{222}Rn and ^{220}Rn concentrations in this uranium mine were close to the worldwide average value. It related to the underground location of the uranium mines that prevented the ^{222}Rn and ^{220}Rn from reaching the upper soil layer. Regarding the origin of ^{222}Rn and ^{220}Rn present at the studied areas, the ^{226}Ra and ^{228}Ra activities in surface soil of study locations were considered. A significant correlation between the average activity of ^{222}Rn, ^{220}Rn and its parent activities ^{226}Ra, ^{228}Ra in surface soil of the studied sites was observed ($r = 0.66$ and 0.91, respectively). ^{220}Rn has a short half-life of 55 s, which makes it less transportable. The measured ^{220}Rn value will therefore indicate the nature of the ^{228}Ra decay source in the surface soil layer, resulting of a high correlation coefficient ($r = 0.91$). By contrast, ^{222}Rn (a decay product of the ^{226}Ra) has a relatively long half-life of 3.8 d which supports the mobility of the ^{222}Rn. Consequently, the ^{222}Rn activity measurement at 1 m from ground surface does not only relate to the situ source but also reflects inputs from surrounding areas.

Overall, the average ^{222}Rn concentration varied from 14.4 to 116 Bq m^{-3}, while that of ^{220}Rn ranged from 22.2 to 62.3 Bq m$^{-3.}$ In general, the concentrations of ^{222}Rn and ^{220}Rn around of the REE mines (NX-Lai Chau, DP-Lai Chau, MH-Lao Cai) were higher than those of the uranium mines (BY-Son La, TS-Phu Tho, DT-Thai Nguyen, NB-Cao Bang) (table 1). The higher ^{220}Rn concentration surrounding the REE mine could be attributed to the high concentration of ^{228}Ra (^{232}Th) in the REE mines (Omori *et al* 2016).

As mentioned, the uranium mines are located underground, while the REE mines were formed as weathering deposits and could be exposed on the surface. Thus, radon could be easily released into the atmosphere. The ^{222}Rn and ^{220}Rn concentrations at 1 m above the ground could be affected by the meteorological conditions (Moses *et al* 1963, Kulali *et al* 2017, Tchorz-Trzeciakiewicz and Kłos 2017). The ^{222}Rn concentration was higher than the ^{220}Rn concentration in most studied sites. This could be related to a short half-life of 55 s of ^{220}Rn. The average concentrations of ^{222}Rn and ^{220}Rn in studied sites were higher than the average worldwide values. Particularly, the ^{222}Rn and ^{220}Rn highest concentrations were observed in the NX-Lai Chau area which was 58 times higher than the worldwide average.

The outdoor ^{222}Rn and ^{220}Rn concentrations in several countries are shown in table 2. The ^{222}Rn concentration in northern Vietnam was higher than in the almost listed countries, with an exception for USA and Poland (Jagielak *et al* 1998, Harley *et al* 2005, Shweikani and Hushari 2005, Malczewski and Żaba 2007, Vaupotič *et al* 2010, Almayahi *et al* 2012, Wu *et al* 2016, Habib *et al* 2018, Wasikiewicz *et al* 2019). Specifically, the outdoor ^{222}Rn concentration in Poland was significantly higher, up to 2160 Bq m^{-3} due to the measurement points at the uranium mine. Additionally, it should be noted that the ^{222}Rn concentration in Poland was measured at the surface (Malczewski and Żaba 2007), which will be less affected by meteorological conditions, while the ^{222}Rn concentration was only about 8.9 Bq m^{-3} at 1 m above the ground (Jagielak *et al* 1998). It should be noted here that the consequence of higher outdoor ^{222}Rn and ^{220}Rn activities is because the study locations are the high-level radiation background areas (with high concentrations of soil/rock) and taking into account meteorological, sampling point, and soil/rock characteristic conditions. For better understanding of the ^{222}Rn and ^{220}Rn concentrations present in the study areas, long-term monitoring of concentrations and meteorological conditions should be undertaken.

3.2. Radiological hazard indices

The AED values due to outdoor ^{222}Rn and ^{220}Rn inhalation in eight sites are shown in table 3. The AED for ^{222}Rn and ^{220}Rn varied from 0.14 to 1.10 mSv.y^{-1} and from 0.005 to 0.01 mSv.y^{-1}, respectively. The total AED of ^{222}Rn and ^{220}Rn ranged from 0.15 to 1.11 mSv.y^{-1} with an average value of 0.49 mSv.y^{-1}, in which the contribution of ^{220}Rn to the AED was insignificant. The highest total AED was found in NX-Lai Chau, while the lowest one was determined in BY-Son La. The average value of the total AED due to inhalation of

Table 2. Outdoor ^{222}Rn and ^{220}Rn measurements in several countries.

Countries	^{222}Rn (Bq m^{-3})	^{220}Rn (Bq m^{-3})	Measure methods	References
China	3–30	—	CR39	Wu *et al* (2016)
Lebanon (spring/summer season)	3.2–47.6	—	E-PERM	Habib *et al* (2018)
Malaysia	6–79	—	SNC	Almayahi *et al* (2012)
Syria	5–66	—	PC	Shweikani and Hushari (2005)
Poland (surface air)	4–2160	4–228	RAD7	Malczewski and Żaba (2007)
Poland (1 m above the ground)	8.9	—	CR39 (Track detector)	Jagielak *et al* (1998)
Slovenia (2005–2006)	12.4	—	CR39	Vaupotič *et al* (2010)
UK (2015–2017)	6	—	PADC	Wasikiewicz *et al* (2019)
USA	11–146	14–43	CR-39	Harley *et al* (2005)
Canada	2–19	9–10.4		
Thailand	7–10	8–19		
Finland	10–12	N.D-12		
North Vietnam	14.4–116	22.3–62.3	RAD7	This study

Table 3. Annual effective doses due to inhalation of ^{222}Rn and ^{220}Rn.

Locations	Annual effective dose (AED) (mSv.y^{-1})		Total (mSv.y^{-1})
	^{222}Rn	^{220}Rn	
NX-Lai Chau	1.10	0.01	1.11
DP-Lai Chau	0.87	0.01	0.88
MH-Lao Cai	0.54	0.01	0.55
BY-Son La	0.14	0.01	0.15
TS-Phu Tho	0.27	0.005	0.27
YP-Yen Bai	0.22	0.01	0.23
DT-Thai Nguyen	0.26	0.005	0.26
NB-Cao Bang	0.44	0.01	0.45
Minimum	0.14	0.005	0.15
Maximum	1.10	0.01	1.11
Average	0.48	0.01	0.49
UNSCEAR (2000)	0.10	0.002	0.10

outdoor ^{222}Rn and ^{220}Rn in this study was nearly five times higher than that worldwide average 0.10 mSv.y^{-1} (UNSCEAR 2000) and slightly higher than the AED due to inhalation of outdoor ^{222}Rn in Jordan with a value of 0.37 mSv.y^{-1} (Alali *et al* 2019).

4. Conclusions

The outdoor ^{222}Rn and ^{220}Rn activities at 1 m above the ground at different sites of REE and uranium mines from northern, Vietnam were determined by the RAD7. The ^{222}Rn and ^{220}Rn concentrations significantly varied and depended on the natural characteristics of the mines. The ^{222}Rn concentration was always higher than that of ^{220}Rn, and the ^{222}Rn and ^{220}Rn concentrations at the sampling points surrounding the REE mines were significantly higher than those surrounding the uranium mines, relating the circumstance conditions and types of those mines. There was a significant correlation between the ^{222}Rn and ^{220}Rn activities and the ^{226}Ra and ^{228}Ra parent activities in surface soil of the studied sites. The ^{222}Rn and ^{220}Rn concentrations in NX-Lai Chau varied over the largest range and were significantly higher than those of other sites. The pattern could be related to the exploitation activities in the REE ore of this site. In general, the average ^{222}Rn and ^{220}Rn concentrations in all studied sites were higher than the worldwide average values. The AED due to inhalation of ^{222}Rn and ^{220}Rn in this study was nearly five times higher than the average worldwide value. Results suggest that the studied sites should be invested in the future in a large-scale monitoring project, including assessing the radiological hazards of indoor ^{222}Rn and ^{220}Rn for the local communities.

Data availability statement

The data cannot be made publicly available upon publication because no suitable repository exists for hosting data in this field of study. The data that support the findings of this study are available upon reasonable request from the authors.

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Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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