



Radiation exposure in a region with natural high background radiation originated from rare earth element deposits at Bat Xat district, Vietnam

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Abstract

Surface deposits of rare earth element (REE) were identified in the Bat Xat district, Northern Vietnam. As the area is inhabited, an in-depth investigation was carried out to assess the radioactivity levels and to evaluate the radiation exposure and radiological risks for the population. Samples of REE ore, soils, water and several foods locally produced were collected and analyzed by gamma spectrometry for determination of ^{232}Th , ^{226}Ra , and ^{40}K . Radon (^{222}Rn and ^{220}Rn) measurements were carried out in dwellings at the villages located near by the REE deposit area, and in Hanoi for comparison. Based on the measured radionuclide concentrations, several radiological parameters were calculated to evaluate the radiological risks for the population and to compare with worldwide values. Results indicate that the representative person of the most exposed group living near the REE deposits may receive a total annual effective dose as high as 37.9 ± 10.6 mSv, i.e., approximately 16 times the world average dose of 2.4 mSv y^{-1} . Consequently, this area should be considered a high background radiation area. Exposure to external radiation and internal radiation from ingestion and inhalation of radionuclides were assessed separately. Radon inhalation contributed to approximately 70% of the total annual effective dose, with both radon isotopes contributing significantly, while the ingestion of local foods and water gave minor contributions to the total effective dose. Several measures were considered to improve the radiation safety and recommendations are given to relocate the most exposed members of the population.

Keywords Naturally occurring radioactive material (NORM) · Radon · Thoron · Radiation exposure · Natural high background radiation area

Introduction

Rare-earth elements (REEs) represent a set of seventeen metallic elements which include fifteen lanthanide elements (from lanthanum $_{57}\text{La}$ to lutetium $_{71}\text{Lu}$), plus scandium ($_{21}\text{Sc}$) and yttrium ($_{39}\text{Y}$). The REEs possess similar chemical properties and occur in a wide array of mineral types

from carbonates to silicates, phosphates, and oxides, being known about 250 individual REE-bearing minerals (McLeod and Shaulis 2018; Al-Ani et al. 2018; Balaram 2019). The REEs are relatively abundant in the earth-crust and occur in several types of deposits, including alkaline igneous rocks, heavy mineral placers, coal deposits, and marine sediments. However, they are rarely concentrated in minable ore deposits and, consequently, the number of worldwide active mines is relatively small (Balaram 2019; Van Gosen 2017). The world reserves of REE oxides are estimated at 132 million tons, and the annual production of REEs in 2020 was about 248.5 kilo tons in total REE oxides (TRE_2O_3), with China being the leading producer since the 1990s (Garside 2021).

Because of their physical and chemical properties, REEs become increasingly used and, currently, they are essential in a wide variety of high technology applications from computer memories, DVDs, catalysts, LED lighting,

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glass additives, wind turbines, aeronautics, and many more (Haque et al. 2014; Van Gosen et al. 2017; Zhou et al. 2017). Every year, the growing demand for REEs has motivated the exploration for additional REE deposits and the intensification of mining, with increasing environmental impacts and so far with little REE recycling (Haque et al. 2014; Van Gosen et al., 2017; Zhou et al. 2017; Carvalho 2017).

Rare earth elements are often associated with primordial radionuclides, such as uranium and thorium (IAEA 2011; Ault et al. 2015). Activity concentrations of those primordial radioactive elements in REE ores have been reported for ^{238}U , ranging from 10,000 to 50,000 Bq kg $^{-1}$, and for ^{232}Th , ranging from 5000 to 350,000 Bq kg $^{-1}$ (UNSCEAR 2008). Because REE minerals contain such primordial radionuclides and their radioactive progenies, these minerals are considered naturally-occurring radioactive materials (NORM) and, therefore, their exploitation may have radiological impacts (Ali 2014). When NORM are highly radioactive, preventive actions may be needed to protect miners from occupational radiation exposure, and to protect the public and the environment (IAEA 2011; ICRP 2019).

In Vietnam, REE deposits were identified in the northwest region of the country at Dong Pao (Lai Chau province) and at Bat Xat district (Lao Cai province). Exploratory work was performed to assess the REE reserves, but the development

of the deposits has not started yet. The REE oxide reserves of the Dong Pao deposit are estimated at 7 million tons, while reserves of the Bat Xat deposit are estimated at 0.4 million tons (Dzien 1990; Moody 2013). The area of the Bat Xat REE deposits (hereafter named “BX REE deposit area”) includes Muong Hum, Nam Pung and Sang Ma Sao mountain villages of Bat Xat district, Lao Cai province, in the northwest of Vietnam, and extends over an area of 142 km ^{2} at an altitude of 1200 m above mean sea level (Figs. 1, 2). The climate in the area is subtropical and characterized by high humidity, ranging from 70 to 90% all over the year, with an annual rainfall ranging from 1800 to 2000 mm. The daily average minimum air temperature is 14 °C occurring in January, while the daily average maximum is 33 °C occurring in June–July (Nguyen and Nguyen 2004).

The BX REE deposits include seven large ore bodies in placers of sand, gravel, and clay with the age of Neogene–Quaternary, surrounded by granite and syenites (Fig. 2). These ore deposits occur at the surface and have thicknesses ranging from 5.5 to 16.7 m. The deposits display an upper layer of weathered rock and a deeper layer of limestone. The composition of the BX REE deposits includes minerals such as monazite, thorium, oxinite, bastnaesite, checcchite, smacskite, quartz, magnetite, ilmenite, inmenorutin, zircon, octit, sphe, and barite. In these deposits, the

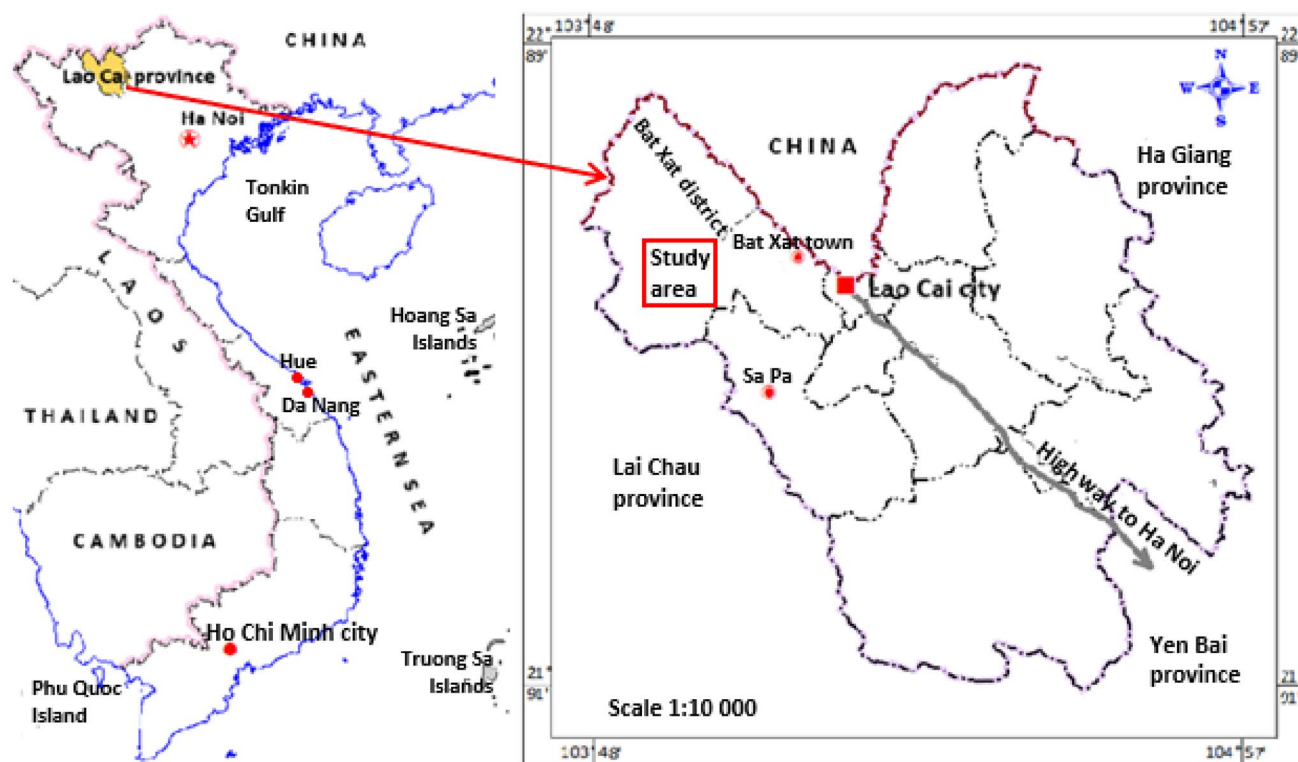


Fig. 1 Geographical position of the study area in Bat Xat district, Lao Cai province, Vietnam, where seven rare earth elements ore deposits were discovered

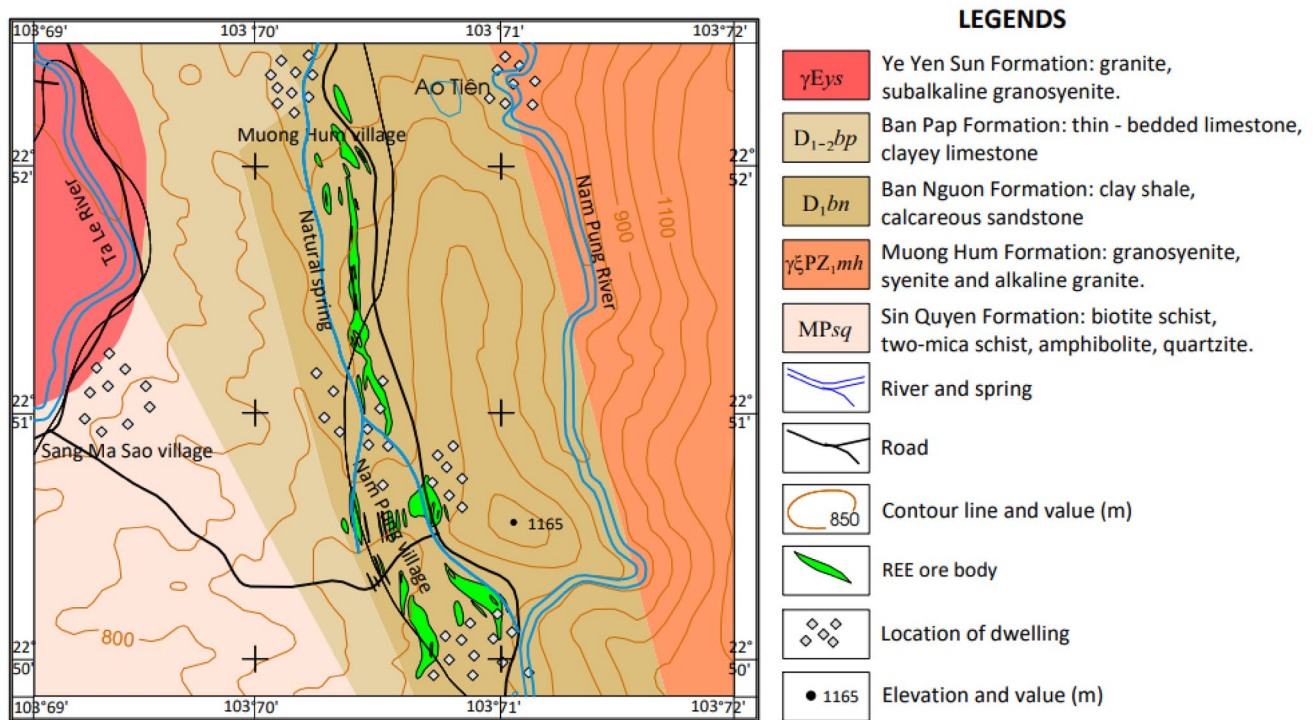


Fig. 2 Geology of the study area: Muong Hum, Nam Pung and Sang Ma Sao mountain villages

ore grade in rare earth oxides (TRE_2O_3) averages 1.5% in weight (wt) (range 0.8–3.0%), while the thorium content averages 0.16% wt (0.11–0.19%) and the uranium content averages 0.016% wt (0.012–0.028%). Uranium and thorium elemental concentrations in the upper layer of the deposits were reported to be 1.2–1.5 times higher than in the deep layer (LDDCXH 2020).

In the BX REE deposit area, the resident population belongs to the ethnic minority Black Ha Nhi. The population lives in near isolation, maintaining a very conservative way of life largely based on their own agriculture, economy, and traditions. The population of the three villages accounted for approximately 7000 inhabitants in the investigation period. The population structure indicates a young population, and nearly 40% is less than 14 years old. There are no dwellings built on top of the ore body, and the houses are located at variable distances from the ore body. The village houses are traditional constructions with one ground story only, built with wood poles and earth (clay), and the house floor is compacted soil. Windows, doors and ceilings are not tight and the natural ventilation is elevated.

The main economic activity of the Black Ha Nhi is agroforestry and the main production is timber. A large fraction of this region has a forest cover, which also covers the soils at the REE deposit. The foods are mostly locally grown and locally consumed, and include rice, corn, cassava and vegetables. Rice is grown in a lowland field with nearly 7

km² surface area, while corn and cassava are planted on the mountain slopes around the three villages, and vegetables are planted in inhabitants' kitchen gardens in the villages. The income of local farmers is low, averaging around 60 USD per month and per capita (Lao Cai People's Committee 2016).

At the Bat Xat REE deposit area, an assessment of the annual external dose received by members of the public living in the proximity of deposits was performed recently, based on the determination of ^{232}Th , ^{226}Ra , and ^{40}K activity concentrations in the ore and in soils (Nguyen et al. 2020). The results showed high dose values from external radiation over the body of the REE ore and in the vicinity. This has caused high concern among the managers of the local government as the area is inhabited, because there is scientific evidence of harmful effects from ionizing radiation on human health, in particular when it involves prolonged exposure to high radon concentrations (WHO 2009). Although no mine has been developed yet and the natural radiation environment remains unchanged in the BX REE deposit area, the assessment of environmental radioactivity levels, radiation exposure pathways, and relative contribution of radionuclides to the annual effective dose received by members of the public needed further investigation and is considered an important radiation protection goal.

This study aimed at confirming the high levels of natural radioactivity and at assessing the total annual effective dose

originated by NORM in the BX REE deposit area (Fig. 2). Beyond the assessment of external radiation exposure originated by radionuclides contained in the ore and soils, and by cosmic radiation, this study included the first measurement and evaluation of exposure to radon (^{222}Rn and ^{220}Rn) through inhalation, and the first assessment of radionuclide intake with local foods and local drinking water. This allowed evaluation of the contribution of several exposure pathways to the radiological health risks for the human population living in the area. This investigation aimed also at assisting environmental managers of the provincial government with scientifically sound recommendations to protect the local population from radiological hazards caused by the NORM.

Materials and methods

Sampling procedure and sample preparation in the laboratory

Samples from the study area were collected in December 2019 and July 2020 for the determination of radionuclide concentrations in environmental materials of the BX REE deposit area. These samples included nineteen surface soil samples (0–20 cm depth), six rice samples, five corn seed samples, five cassava root samples, and five water samples from Ta Le River (w3), Nam Pung River (w1, w4) and from a natural spring (w2, w5) that recharges the river (Fig. 3a). The Ta Le and Nam Pung River as well as the spring are the only water supplies available and, without any special treatment, they meet the daily consumption needs of the population. The soil samples were chosen to represent three areas: samples S1, S2 and S3 (Fig. 3a) represent the top soil at the REE ore body, and these were undisturbed soil samples; samples S4–S10, and S19 (Fig. 3a), were taken from kitchen gardens located at distances ranging from 300 to 500 m from the ore body. These samples were taken from the area most densely populated in Muong Hum and Nam Pung villages. The soil samples S11–S17 (Fig. 3a) were collected from a rice field located at a distance slightly larger than 2 km from the ore body. These samples represent agriculture soils. Finally, sample S18 (Fig. 3a) was taken from kitchen gardens in Muong Hum located at a distance of around 2 km from the ore body. In the region, the soil is of a Ferralic Acrisols type according to the classification proposed by the Food and Agriculture Organization (FAO). This soil is characterized by a low organic matter content as well as by low nutrient concentrations. It is noted that the local farmers make little application of fertilizers.

To ensure the representativeness of soil samples, the sampling was conducted following the procedure described by Huy et al. (2012). Briefly, at each soil sampling site, an

equilateral triangle with edges of 20 m length was drawn on the ground. Within the triangle, soil sub-samples were collected using a corer made from a stainless steel tube, with 6 cm internal diameter and 30 cm length. These sub-samples were taken at ten points: six points along three edges and other four points distributed evenly inside the triangle. All the sub-samples (each with about 1 kg) were pooled and packed together in high-density polyethylene (HDPE) bags for transport to Hanoi and later laboratory analysis.

Rice, cassava, corn, and vegetables, are the main components of the local population diet. Consequently, these foodstuffs of the local food production were selected to assess the radionuclide ingestion of the local population. Rice samples, each of about 2 kg, were bought from farmers in the three villages and harvested from the agricultural land nearby Sang Ma Sao village, 2 km from the REE deposits. Corn seeds were also bought from farmers in the three villages, each sample with about 2 kg. Corn samples M1, M2, M3 were harvested from fields located at 0.3 km from the REE deposit, while M4 and M5 were from the fields nearby Sang Ma Sao village. Fresh cassava roots were harvested directly from the fields with the permission from owners. Samples C1, C2 and C3 were harvested from fields located on the foot of REE deposit, while C4 and C5 were harvested from fields located at about 2 km distance from the ore body. These cassava samples, of about 3 kg each, were composites of sub-samples taken from the four corners and one from the middle of the investigated cassava fields. Vegetable (colza, *Brassica juncea*) samples, each of about 2 kg, were bought from farmers and collected from their kitchen gardens at 500 m and 2 km from the REE deposit. The dead leaves of colza were removed while the edible leaves were packed into a HDPE bag and, similar to the bags for the other food samples, the bags were duly identified and transported to the laboratory for further treatment and analysis. Figure 3a shows the sampling locations for soil, water and food. The corresponding geographic coordinates along with the activity concentrations of radionuclides determined for all samples are shown in the electronic supplement.

In the laboratory, the soil samples were air-dried and sieved through a 1 mm mesh silk sieve to remove gravel, plant roots, leaves etc. The samples were dried further at 105 °C until constant weight and then homogenized using a drum mixer. The collected rice and corn samples were washed to remove the bran and dust adhered on the seeds and then cooked in boiling water following the cooking tradition of the local population. The cassava roots were peeled and the edible parts washed and steamed. The vegetable samples were washed off and boiled. The cooked food was dried at 105 °C, the moisture content determined, and then foods were subjected to grinding (Tran et al. 2018). The water samples were filtered through polycarbonate

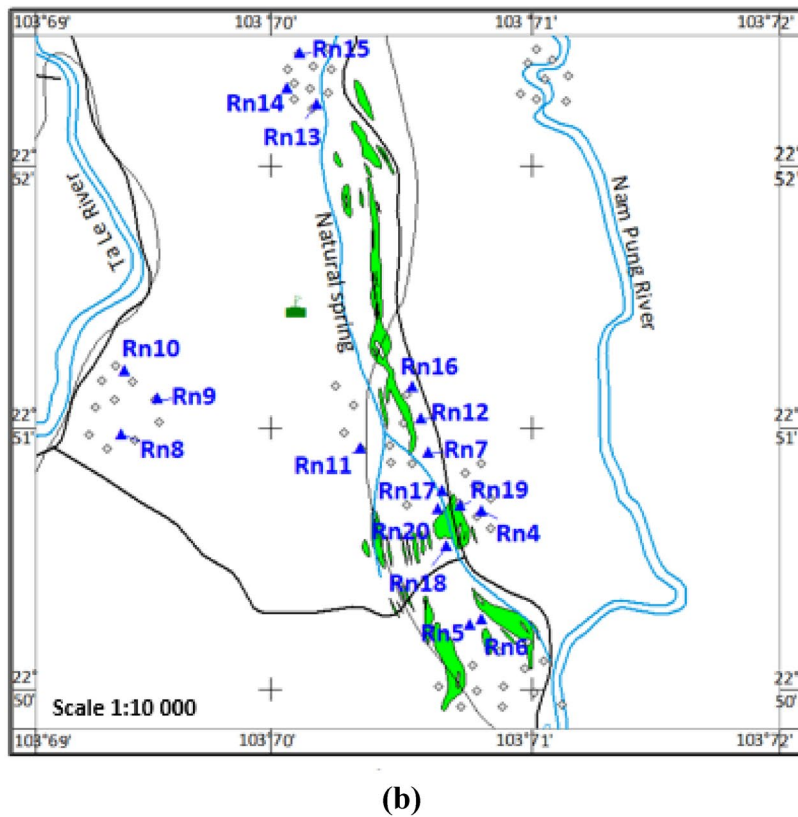
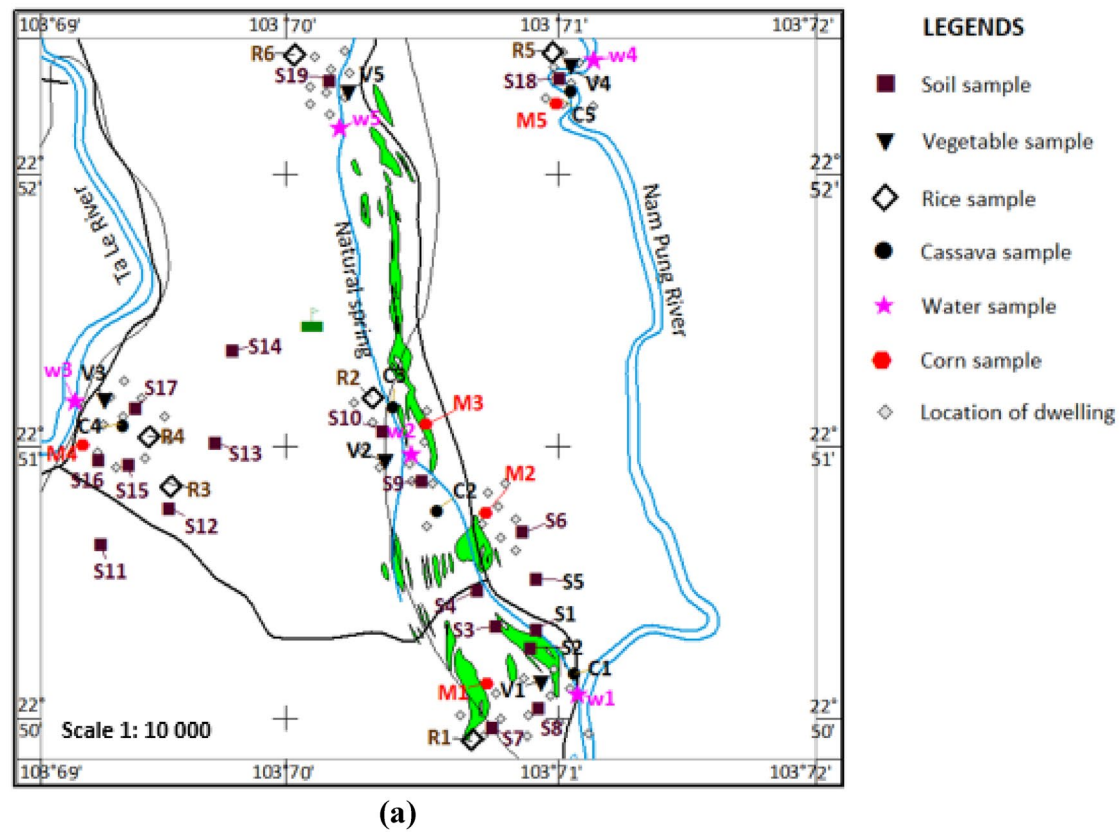


Fig. 3 Sampling sites of soil, water and foods (a), and radon measurements (b) for the assessment of annual committed effective doses to the members of the public in the study area (See the Electronic supplement for geographic coordinates of the sampling sites)

membrane filters of 0.45 µm pore size in order to remove any suspended matter.

The dried soil, food powder, and filtered water samples were weighed and hermetically packed in HDPE cylindrical boxes of 115 mm diameter and 110 mm height, and closed with plastic lids. To prevent radon escape from the sample container, the lid was sealed with epoxy resin. The samples in containers were stored for 30 days to allow formation of secular radioactive equilibrium between the progenitors (^{232}Th , ^{226}Ra) and their progenies (^{228}Ac from ^{132}Th ; ^{214}Bi and ^{214}Pb from ^{226}Ra) before radiometric determinations.

Determination of radionuclides' activity concentrations by gamma spectrometry

The activity of gamma emitting radionuclides in soil, food, and water samples was determined using a low background gamma-spectrometer (OrtecTM) equipped with a high resolution HPGe detector, and the spectra analysis was performed using the Gamma Vision (OrtecTM) software. The energy resolution of the detector was 1.9 keV at the 1.33 MeV of ^{60}Co gamma-ray peak and the relative counting efficiency was 18%. The samples were counted for 24 h to minimize the statistical counting error and to achieve a counting relative uncertainty lower than $\pm 10\%$. As a quality control procedure to ensure the accuracy and precision of radionuclide determinations, several Certified Reference Materials (CRM) produced by the International Atomic Energy Agency (IAEA) were systematically used, in particular the CRM IAEA-375 (top soil from agriculture field).

Potassium-40, ^{228}Ac , ^{214}Bi , and ^{214}Pb , in samples were identified and their activities were quantified based on the gamma peak intensities at 1.46 MeV, 911 keV, 609 keV, and 352 keV, respectively (Tran et al 2018; Dang et al 2014). Assuming secular radioactive equilibrium, the activity of ^{228}Ac was assigned to the activity of ^{232}Th , and the average of ^{214}Bi and ^{214}Pb activities was assigned to the activity of ^{226}Ra .

Correction for self-gamma absorption and the difference in density between the investigated samples and the CRM used as a calibration standard was made following the method described by Jodlowski (2006). The activity concentrations of ^{226}Ra (^{214}Bi and ^{214}Pb), ^{232}Th (^{228}Ac), and ^{40}K were calculated based on Eq. 1 (Jodlowski and Kalita 2010):

$$A_s = \frac{N_s \times M_{\text{std}} \times A_{\text{std}} \times F_i}{N_{\text{std}} \times M_s} \quad (1)$$

where A_s and A_{std} are the activity concentrations of an investigated and a standard sample, respectively; N_s , N_{std} , M_s , and M_{std} are the net measured gamma ray intensity (N) and mass (M) of an investigated sample and a standard sample (CRM), respectively, and F_i is the correction factor for the

differences between the densities of the samples and the standard sample.

The Minimum Detectable Activity (MDA, Bq kg^{-1}) of the gamma-activity measurement system was evaluated based on Eq. 2 (L'Annunziata 1998):

$$\text{MDA} = \frac{2.71 + 4.65 \sqrt{N_B}}{t \times \varepsilon \times I_\gamma \times M_s} \quad (2)$$

where N_B is the background count, t is the measurement time (s), ε is the absolute efficiency of the detector (%), I_γ is the gamma emission probability, and M_s is the dry mass of the sample (kg).

The MDA of ^{228}Ac (911 keV), ^{214}Pb (352 keV) and ^{214}Bi (609 keV) was estimated (Eq. 2) to be as low as 0.13, 0.16 and 0.14 Bq kg^{-1} dry mass (d.m.), respectively, while that for ^{40}K was 10 times higher.

The activity concentrations of the radionuclides in cooked and ready-to-eat foods were calculated based on the gamma intensities measured on dry samples and the activity concentrations converted to a wet mass basis using the corresponding dry-to-wet mass ratios. Activity concentrations of radionuclides are expressed with the propagated uncertainty at $\pm 1\sigma$ in Bq kg^{-1} d.m. for soil and food, and in Bq L^{-1} for water.

In a previous study it was identified that ^{210}Pb and ^{210}Po , progenies of the ^{238}U decay series, accumulate in plants and especially in aquatic organisms, but they were largely eliminated by volatilization in the preparation of cooked food (Tran et al. 2018). Therefore, in this study, activity concentrations of ^{210}Pb and ^{210}Po were not included in the estimation of the annual committed effective dose.

Measurement of cosmic rays

The radiation background from cosmic rays was measured using an Aloka survey meter (model TGS-121) equipped with a Geiger Müller (GM) detector. This device was calibrated in the Secondary Standard Dosimetry Laboratory (SSDL) of the Institute for Nuclear Sciences and Technology (INST) in Hanoi. For shielding from the terrestrial gamma radiation, the measurements were conducted on a floating bamboo raft at the middle of Ao Tien water reservoir at an altitude of 1100 m above mean sea level. The reservoir has a surface area of around 1 km^2 and a depth of 2 m, and is located at 7 km south of the BX REE ore deposit area near Muong Hum village (Fig. 2).

^{222}Rn (radon) and ^{220}Rn (thoron) concentrations in the air

Twenty-five double-chamber RADUET solid state nuclear track detectors (SSNTD) were tied on wood beams, 2 m

above the ground, inside dwellings of Muong Hum, Nam Pung and Sang Ma Sao inhabitants (Fig. 3b). The detectors were exposed for six months, from December 2019 to May 2020. Three pairs of similar radon detectors were installed in living rooms of three apartments in Hanoi city for comparison with the radon dose received by the inhabitants of BX REE deposit area.

The RADUET detector consists of two chambers with CR-39 film detectors installed inside the chambers allowing determination of radon (^{222}Rn) and thoron (^{220}Rn) activities (Radosy 2008). The main chamber is selective primarily for ^{222}Rn . Due to the detector design, ^{220}Rn cannot reach the main chamber because of its relatively short half-life of 55 s, but the secondary chamber is sensitive for both ^{222}Rn and ^{220}Rn (Radosy 2008). A simple linear calculation allows reporting separately the activity concentrations of radon and thoron (Tokonami 2005).

After collecting the RADUET detectors, the CR-39 films were removed from the chambers and subjected to etching at 80 °C in 6 M NaOH solution for four hours. The etching process developed the tracks produced in the films by the alpha particles emitted from ^{222} and ^{220}Rn during radioactive decay. The number of tracks was counted under an OLYMPUS CX21 microscope (Bui et al. 2014). The number of tracks formed in the detector surface is proportional to the number of alpha particles from ^{222} and ^{220}Rn hitting the detector. To convert the track numbers into ^{222}Rn and ^{220}Rn activities in Becquerel per unit air volume (Bq m^{-3}), the RADUET detectors were calibrated at the Institute of Radiochemistry and Radioecology of the University of Pannonia, Hungary, using a radon and thoron calibration chamber (Kavasi et al. 2006). The sensitivity and background of the detectors were determined as 2.4 tracks $\text{cm}^{-2} \text{ kBq}^{-1} \text{ h}^{-1} \text{ m}^3$ and 0.3 tracks cm^{-2} , respectively (Radosy 2008).

Absorbed gamma dose rate

The calculated dose rate at 1 m above the ground was used to assess the human body exposure to the gamma radiation emitted by naturally occurring radionuclides contained in the soil. The dose rate, D , was calculated using the soil activity concentration to dose conversion factors, i.e., the dose in nGy h^{-1} was calculated from the activity concentrations of radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in the soil using Eq. 3 (UNSCEAR 2008):

$$D = 0.46A_{\text{Ra}} + 0.62A_{\text{Th}} + 0.042A_{\text{K}} \quad (3)$$

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

The absorbed gamma dose rates calculated through Eq. 3 for the 19 soil sampling points were compared with direct dose rate measurements made 1 m above ground at the same

19 sampling points using the portable Aloka survey meter (see electronic supplement). The correlation between calculated and measured data sets displayed a coefficient of correlation $R^2 = 0.9896$ ($p < 0.001$) confirming the robustness of the method for calculating the absorbed gamma dose rate D .

Annual effective dose from external gamma radiation exposure

The annual gamma external effective dose (AGED in Sv y^{-1}) was calculated by summing the outdoor and indoor annual effective doses which were calculated on the basis of the gamma absorbed dose rate as follows (Eq. 4):

$$\text{AGED} = \text{OAED} + \text{IAED} \quad (4)$$

where OAED and IAED (Sv y^{-1}) are the outdoor and indoor gamma annual external effective dose, respectively.

The OAED was calculated based on Eq. 5:

$$\text{OAED} = D \times \text{DCF} \times \text{OF} \times T \quad (5)$$

where D is the gamma dose rate (Eq. 3), DCF is the dose conversion factor assumed to be 0.7 (Sv Gy^{-1}) (UNSCEAR 2008); OF is an outdoor occupancy factor (assumed to be 0.2) (UNSCEAR 2000; UNSCEAR 2008); T is the hours in the year ($T = 8760 \text{ h y}^{-1}$).

The IAED was calculated based on Eq. 6:

$$\text{IAED} = D \times \text{DCF} \times \text{OF} \times T \quad (6)$$

where D and DCF have the same meaning as above. For the IAED the occupancy factor (OF) 0.8 was assumed (UNSCEAR 2008). Actually, as houses are built with earth and the floor is just the natural soil, OAED and IAED were calculated using the same external gamma dose rate D (Eq. 3) and they differ only in the Occupancy Factor.

Annual committed effective dose from ingestion

The average annual committed effective dose due to the intake of radionuclides contained in the food and water (E_{in} , Sv y^{-1}) was evaluated using Eq. 7 (UNSCEAR 2008; Silva et al. 2019; Ho et al. 2019; Madruga et al. 2020; Pintilie-Nicolov et al. 2020):

$$E_{\text{in}} = \sum_i (Q_i \times C_{i,r}) \times f_r \times g_r \quad (7)$$

where i denotes a food group (cereal, vegetable, etc.) and water; Q_i and $C_{i,r}$ denote the consumption rate of food (kg y^{-1}) or water (L y^{-1}) and the activity concentration of radionuclide r in the i th food group (Bq kg^{-1}) or in drinking water (Bq L^{-1}), respectively; f_r is the fractional absorption of radionuclide r in the gastrointestinal tract, and g_r is the

Table 1 Committed effective dose per unit intake (g_r , Sv Bq⁻¹) and fractional absorption in the gastrointestinal tract (f_r) of several major naturally occurring radionuclides for adults (IAEA 2014)

Nuclide	Half-life, $T_{1/2}$	g_r , Sv Bq ⁻¹	f_r
⁴⁰ K	1.28·10 ⁹ y	6.20·10 ⁻⁹	1
²²⁶ Ra	1.60·10 ³ y	2.80·10 ⁻⁷	0.2
²¹⁴ Bi	0.332 h	1.1·10 ⁻¹⁰	0.05
²¹⁴ Pb	0.447 h	1.4·10 ⁻¹⁰	0.2
²³² Th	1.40·10 ¹⁰ y	2.30·10 ⁻⁷	5.0·10 ⁻⁴
²²⁸ Ac	6.13 h	4.30·10 ⁻¹⁰	5.0·10 ⁻⁴
²³⁴ Th	24.1 d	3.40·10 ⁻⁹	5.0·10 ⁻⁴
²³⁴ U	2.44·10 ⁵ y	4.90·10 ⁻⁸	0.02
²³⁸ U	4.47·10 ⁹ y	4.50·10 ⁻⁸	0.02

y year, d day, h hour

Table 2 Major diet components and food consumption rate (mean and range) of adults in the mountainous region of northern Vietnam (NIN 2010)

Food	Consumption rate in g per capita per day mean (min–max)
Rice	404.8 (390.71–418.88)
Corn	14.7 (11.83–17.51)
Tubers (cassava)	3.5 (2.14–4.85)
Vegetables (leaves)	208.6 (198.16–219.05)
Meat	91.9 (82.66–101.18)
Eggs	13 (11.4–14.5)
Fish	29.8 (25.67–33.89)

activity to dose conversion factor for the ingested radionuclide r (Sv Bq⁻¹).

The activity to dose conversion factors g_r and the radionuclide fractional absorption factors in the gastrointestinal tract f_r , recommended by the International Atomic Energy Agency (IAEA) were used in the calculations and are summarized in Table 1 (IAEA 2014). The RadToolbox computer code supplied by the National Regulatory Commission of the United States was used to compute the annual committed effective dose for adults in the study area based on their diet and on the activity concentrations of radionuclides in cooked foods and drinking water. The representative diet composition and food consumption rates by the population of the mountainous region of northern Vietnam are based on data from the National Institute of Nutrition (Vietnam) and shown in Table 2 (NIN 2010).

The consumption rate of meat, eggs and fish by the local population was almost five times less than the consumption rate of cereals and vegetables (NIN 2010) and, consequently, in this study the activity concentrations of radionuclides in such minor food items were not determined. Hence, the

dose contributions from radionuclides in the diet calculated through Eq. 7 are based on the major diet components that altogether represent 82% of the total diet.

The annual consumption of drinking water (Q_i in Eq. 7) for adults was assumed to be 730 L y⁻¹ as recommended by the World Health Organization (WHO 2017). This value was used to calculate the annual committed effective dose from the intake of radionuclides in drinking water.

Annual effective dose from inhalation of radon and thoron

The annual effective dose due to inhalation of radon (²²²Rn) and its progenies was calculated on the basis of Eq. 8 (UNSCEAR 2000):

$$E_{Rn} = C_{Rn} \times F_{Rn} \times K_{Rn} \times T \quad (8)$$

where E_{Rn} is the annual effective dose from radon (nSv y⁻¹); C_{Rn} is the radon concentration measured in the air in dwellings of the study area (Bq m⁻³); F_{Rn} is an average equilibrium factor for radon and its progenies assumed to be 0.46 (Ramachandran et al (2003)) which is close to the value of 0.4 recommended by UNSCEAR (2000); K_{Rn} is the international recommended activity to dose conversion factor of radon, 9 nSv Bq⁻¹ h⁻¹ m³ (UNSCEAR 2000); and T is the time of one year in hours (8760 h y⁻¹).

Similarly, the annual effective dose due to inhalation of thoron (²²⁰Rn) and its progenies was calculated as follows (UNSCEAR 2000):

$$E_{Tn} = C_{Tn} \times F_{Tn} \times K_{Tn} \times T \quad (9)$$

where E_{Tn} is the annual effective dose from thoron (nSv y⁻¹), C_{Tn} is the measured thoron concentration in the air of dwellings (Bq m⁻³), F_{Tn} is an average equilibrium factor for thoron and its progenies assumed to be 0.09 (Ramola et al. (2003)) for Indian dwellings, K_{Tn} is the UNSCEAR (2000) recommended activity to dose conversion factor of thoron, 40 nSv Bq⁻¹ h⁻¹ m³ (UNSCEAR 2000); and T is the time of one year in hours (8760 h.y⁻¹).

The total annual effective dose from inhalation ($AEDI_{Rn+Tn}$) of radon isotopes (²²²Rn and ²²⁰Rn) was calculated by summing up the E_{Rn} and E_{Tn} values.

It is noted that in the study area local houses are simple constructions made from earth (clay) and wood poles. By the time of this study, the walls of such houses displayed cracks, and doors and windows were not airtight. In this study, the C_{Rn} and C_{Tn} were measured for the indoor air and assumed to be the same during day and night periods, because of the natural ventilation and, anyway, averaged by the integrative radon determination method. It was assumed that radon and thoron concentrations in air determined over a six months period, from December to May that cover both dry and rainy

seasons, are representative of the corresponding annual average concentrations.

Excess lifetime cancer risk

The excess lifetime cancer risk (ELCR) estimates the probability of cancer incidence in a human population for a specific lifetime from the exposure to naturally occurring radionuclides. The ELCR was estimated using Eq. 10 (ICRP 2007):

$$\text{ELCR} = \text{AGED} \times \text{DL} \times \text{RF} \quad (10)$$

where AGED is the annual gamma effective dose; DL is the duration of life (70 y), and RF is the risk factor, this is a detriment-adjusted nominal risk coefficient for cancer caused by radiation per Sievert received. The International Commission on Radiological Protection (ICRP) recommends an RF value of 0.055 Sv^{-1} for the whole population under exposure (ICRP 2007).

Statistical treatment of activity concentrations of radionuclides in soils and in the air

As the number of samples in this study is limited, the distribution of data is expected to be non-normal and the *t*-test for two-samples, assuming unequal variances, was applied to compare the average activity concentrations of ^{232}Th , ^{226}Ra , ^{40}K in the soil collected at the REE ore body with those in other soil samples, and to compare the concentrations of radon and thoron in indoor air in dwellings near the ore body with those in dwellings further away from the ore body and in apartments in Hanoi. The averages of the sample groups would be significantly different from each other, with a 95% confidence level ($\alpha=0.05$), if $|t\text{-Stat}|$ value $> t$ critical value.

Results

A statistical summary of the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in food, water, and soil samples from the BX REE deposit area is presented in Table 3 (see Fig. 3a for the locations and the electronic supplement for the geographic coordinates of the sampling locations).

The *t*-test showed that the average activity concentrations of all radionuclides determined (Table 3) in soil collected from the ore body (S1, S2, S3) were different from those in soils collected from the kitchen gardens and from the agricultural field outside the ore body (S4 through S19). The absolute *t*-Stat value for the average ^{226}Ra activity concentration in soil from the ore body vs. average ^{226}Ra concentration in soils from kitchen gardens (0.3–0.5 km from the ore body) was 6.14, higher than the *t*-critical value of 2.78 at $\alpha=0.05$. The absolute *t*-Stat value for the average ^{226}Ra activity concentration in soils from the kitchen gardens vs. average ^{226}Ra activity concentration in soils from the agricultural field was 2.53, higher than the *t*-critical value of 2.36 at $\alpha=0.05$. The absolute *t*-Stat value for the average ^{232}Th activity concentration in soils from the ore body vs. average ^{232}Th concentration in soils from the kitchen gardens was 6.77, higher than the *t*-critical value of 2.36 at $\alpha=0.05$ and, finally, the absolute *t*-Stat value for the average ^{232}Th activity concentration in soils from the kitchen gardens vs. the average ^{232}Th concentration in soils from the agricultural field was 6.55, higher than the *t*-critical value of 2.36 at $\alpha=0.05$. These results indicate that the radiation doses from exposure to gamma radiation were significantly different for the population residing in different areas. Details of this statistical treatment are presented in the electronic supplement.

Based on the activity concentrations of radionuclides (^{232}Th , ^{226}Ra , and ^{40}K) determined in soils (Table 3 and

Table 3 Summary of activity concentrations (average $\pm$ standard error and range in parenthesis) of naturally occurring radionuclides in food, water, and soil (S) samples from BX REE ore deposits area (see

Fig. 3a for sampling sites); all concentrations are given in terms of Bq kg^{-1} d.m. or in Bq L^{-1} ; *n* number of samples

Sample	^{40}K	^{226}Ra (^{238}U)	^{232}Th
Food			
Rice (boiled), <i>n</i> =6	29 ± 7 (25–35)	<MDA (0.15)	<MDA (0.13)
Corn (boiled), <i>n</i> =5	28 ± 6 (23–37)	<MDA (0.15)	<MDA (0.13)
Cassava (steamed), <i>n</i> =5	48 ± 13 (38–59)	<MDA (0.15)	<MDA (0.13)
Colza (boiled), <i>n</i> =5	34 ± 9 (22–45)	0.45 ± 0.12 (0.25–0.56)	0.70 ± 0.18 (0.46–0.86)
Water <i>n</i> =5	38 ± 8 (27–46)	2.1 ± 0.7 (1.4–2.7)	2.2 ± 1.0 (1.3–3.7)
Soil			
S1–S3 (at the ore deposit)	2401 ± 87 (2306–2476)	3633 ± 65 (3061–4447)	$15,160 \pm 80$ (13,076–18,742)
S4–S10, S19 (0.3–0.5 km from the ore deposit)	459 ± 236 (251–1007)	800 ± 716 (333–2538)	2293 ± 832 (1001–3898)
S11–S18 (1–2 km from the ore deposit)	1137 ± 241 (781–1358)	207 ± 54 (144–292)	360 ± 57 (257–421)

MDA minimum detectable activity expressed in Bq kg^{-1} d.m. Absolute MDA values are in parenthesis

electronic supplement), the gamma absorbed dose rate, the outdoor as well as indoor annual external gamma effective dose (OAED and IAED), total annual gamma effective dose (AGED), and the excess lifetime cancer risk (ELCR) were calculated for each site following Eqs. 3, 4, 5, 6, and 10. The statistical results of these calculations are presented in Table 4 (details of the calculations are presented in the electronic supplement). As there were no houses and no residents over the ore deposits, the OAED, IAED and AGED were not calculated for sites S1, S2 and S3. As shown in Table 3, the ^{232}Th , ^{238}U , and ^{226}Ra concentrations in cooked rice were below the MDA. In boiled colza, ^{226}Ra and ^{232}Th were determined at concentrations of $0.45 \pm 0.12 \text{ Bq kg}^{-1} \text{ d.m.}$ and $0.70 \pm 0.18 \text{ Bq kg}^{-1} \text{ d.m.}$, respectively. In steamed cassava only ^{40}K was detected, and ^{226}Ra and ^{232}Th were lower than the MDA. Thorium-232 was detected in boiled corn from the sample site M1 only (see electronic supplement).

Table 5 presents an estimate of the average annual committed effective dose to adults through the ingestion pathway, which was calculated using Eq. 7. The dose from ingestion was calculated at 0.25 mSv y^{-1} and the drinking water was the major contributor with 93% of the total dose from ingestion, followed by vegetable (*Brassica juncea*) which accounted for 6% of the dose (Table 5).

Table 6 shows the results of radon and thoron concentrations measured in indoor air in dwellings near by the

BX REE ore deposits and in Hanoi, 300 km south of the study area. Although a total of 25 pairs of radon-thoron RADUET detectors have been installed, only 20 pairs were harvested, and five pairs were lost. The annual effective doses due to radon and thoron inhalation were calculated using Eqs. (8) and (9) and the activity concentrations of the two radon isotopes measured in indoor air. The results of these calculations are also presented in Table 6.

The *t*-test showed that the average radon and thoron activity concentrations in the three areas (i.e., Hanoi, proximity of the ore body (0.3–0.5 km), and at a distance of 1–2 km from the ore body) were significantly different from each other. The absolute *t*-Stat value of the average radon concentration in indoor air in Hanoi vs. that in dwellings at 1–2 km from the ore body was 2.89, higher than *t*-critical value of 2.58 at $\alpha=0.05$. The absolute *t*-Stat value of the average radon concentration in indoor air in dwellings at 1–2 km far from the ore body vs. that in dwellings in the proximity (0.3–0.5 km) to the ore body was 4.00, higher than *t*-critical value of 3.18 at $\alpha=0.05$. Similarly, for thoron the absolute *t*-Stat value for the average ^{220}Rn concentration in indoor air in dwellings at 1–2 km far from the ore body vs. that in dwellings at 0.3–0.5 km from the ore body was 3.83, higher than the *t*-critical value of 3.18 at $\alpha=0.05$. Details of this statistical treatment are presented in the electronic supplement.

Table 4 Calculated (average $\pm$ standard error and range in parenthesis) gamma absorbed dose rate (*D*); outdoor annual effective dose (OAED); indoor annual effective dose (IAED); the total annual

gamma external effective dose (AGED), and the excess lifetime cancer risk (ELCR) factor for members of the local population living at different distances from the BX REE ore deposit area

Sample	<i>D</i> $\mu\text{Gy h}^{-1}$	OAED mSv y^{-1}	IAED mSv y^{-1}	AGED mSv y^{-1}	ELCR
S1–S3 (at the ore deposits)	11.0 ± 2.0 (9.5–13.2)			67 ± 12 (58–81)	
S4–S10, S19 (0.3–0.5 km from the ore deposits)	1.8 ± 0.9 (1.0–2.8)	2.2 ± 1.1 (1.2–3.4)	8.9 ± 4.2 (4.9–13.5)	11.1 ± 5.3 (6.2–16.9)	$4.0 \cdot 10^{-02}$
S11–S18 (2 km from the ore deposits)	0.37 ± 0.07 (0.28–0.43)	0.45 ± 0.09 (0.34–0.53)	1.8 ± 0.4 (1.4–2.1)	2.3 ± 0.4 (1.7–2.7)	$7.9 \cdot 10^{-03}$
Worldwide average (UNSCEAR 2000, 2010)	0.059	0.07	0.41	0.48	$2.9 \cdot 10^{-04}$

Table 5 Estimated annual committed effective doses to the adults living in the BX REE ore deposits area, due to ingestion of radionuclides with the foods

Food	Dry:wet mass ratio in cooked food	Food consumption rate ($\text{kg capita}^{-1} \text{ y}^{-1}$)	Annual committed effective dose from ingestion ($\text{mSv person}^{-1} \text{ y}^{-1}$)	Contribution of food type to the total committed effective dose (%)
Rice	0.76	147.75	$1.67 \cdot 10^{-04}$	0.2
Corn	0.82	5.37	$1.06 \cdot 10^{-03}$	0.5
Cassava	0.76	1.28	$4.12 \cdot 10^{-04}$	0.3
Vegetable	0.72	76.14	$1.04 \cdot 10^{-02}$	6.0
Water		730	$2.04 \cdot 10^{-01}$	93.0
Total			$2.50 \cdot 10^{-01}$	100

Table 6 Activity concentrations of radon (^{222}Rn) and thoron (^{220}Rn) in indoor air (average $\pm$ standard error); annual effective doses (E_{Rn} and E_{Tn}) and total annual effective dose ($\text{AEDI}_{\text{Rn+Tn}}$) caused by radon and thoron inhalation to members of the population in Hanoi and BX REE ore deposit area (see Fig. 3b for the measurement sites in the study area)

No.	Site	^{222}Rn (Rn) Bq m $^{-3}$	^{220}Rn (Tn) Bq m $^{-3}$	E_{Rn} , mSv y $^{-1}$	E_{Tn} , mSv y $^{-1}$	$\text{AEDI}_{\text{Rn+Tn}}$, mSv y $^{-1}$
In Hanoi dwellings, 300 km south from BX REE ore deposits area:						
1	Rn1	41 $\pm$ 7	60 $\pm$ 5	1.4	0.7	2.1
2	Rn2	74 $\pm$ 7	22 $\pm$ 8	2.4	0.3	2.7
3	Rn3	58 $\pm$ 6	< 1	1.9	0	1.9
$\text{AEDI}_{\text{Rn+Tn}}$						2.2 $\pm$ 0.4
In Bat Xat dwellings, built at 1–2 km distance from BX REE ore deposits:						
4	Rn4	130 $\pm$ 11	17 $\pm$ 7	4.3	0.2	4.5
5	Rn5	97 $\pm$ 10	104 $\pm$ 9	3.2	1.2	4.4
6	Rn6	85 $\pm$ 11	32 $\pm$ 6	2.8	0.4	3.2
7	Rn7	91 $\pm$ 12	63 $\pm$ 6	3.0	0.7	3.7
8	Rn11	64 $\pm$ 7	48 $\pm$ 5	2.1	0.6	2.7
9	Rn12	111 $\pm$ 10	111 $\pm$ 8	3.7	1.3	5.0
10	Rn16	123 $\pm$ 12	186 $\pm$ 8	4.1	2.1	6.2
11	Rn8	68 $\pm$ 8	31 $\pm$ 5	2.2	0.4	2.6
12	Rn9	66 $\pm$ 6	13 $\pm$ 5	2.2	0.2	2.4
13	Rn10	72 $\pm$ 8	67 $\pm$ 6	2.4	0.8	3.2
14	Rn13	73 $\pm$ 9	50 $\pm$ 6	2.4	0.6	3.0
15	Rn14	87 $\pm$ 11	15 $\pm$ 3	2.9	0.2	3.1
16	Rn15	77 $\pm$ 9	439 $\pm$ 10	2.5	5.1	7.6
$\text{AEDI}_{\text{Rn+Tn}}$						4.0 $\pm$ 1.6
In Bat Xat dwellings, built at 0.3–0.5 km distance from BX REE ore deposits:						
17	Rn17	507 $\pm$ 26	350 $\pm$ 15	16.8	4.0	20.8
18	Rn18	896 $\pm$ 22	73 $\pm$ 13	29.6	0.9	30.5
19	Rn19	304 $\pm$ 14	2,352 $\pm$ 22	10.1	27.1	37.1
20	Rn20	310 $\pm$ 15	586 $\pm$ 14	10.2	6.7	17.0
$\text{AEDI}_{\text{Rn+Tn}}$						26.3 $\pm$ 9.2

Discussion

Absorbed gamma dose rate, outdoor and indoor annual gamma effective dose originated from radionuclides (^{40}K , ^{238}U series, ^{232}Th series) in soil

In the present study it was observed that the average ^{40}K activity concentration in soils from the REE ore body (samples S1, S2 and S3) was 3.6 times higher than that in samples taken apart the ore body (samples S4 through S10 and S19 from kitchen gardens; samples S11–S18 from agricultural fields, Table 3). This might be due to the fact that the soil at the REE ore deposits has a forest cover and was not tilled for cultivation and, thus, it has been less affected by erosion, which usually leads to the loss of potassium. In contrast, kitchen garden soils and agricultural field soils, with a frequent tillage and under the heavy regional rainfall easily lose potassium with the surface run off.

As seen from Table 4, the absorbed gamma dose rate (D) caused by the radionuclides in the BX REE deposits (S1, S2, S3) at 1 m above the ground surface was $11.0 \pm 2.0 \mu\text{Gy h}^{-1}$ (9.5 – $13.2 \mu\text{Gy h}^{-1}$). For the areas at distances of

0.3–0.5 km and 2 km from the ore body, the D values were $1.8 \pm 0.9 \mu\text{Gy h}^{-1}$ (1.0 – $2.8 \mu\text{Gy h}^{-1}$) and $0.37 \pm 0.07 \mu\text{Gy h}^{-1}$ (0.28 – $0.43 \mu\text{Gy h}^{-1}$), respectively. These findings were somewhat higher than the mean absorbed gamma dose rates reported before for the same area of $0.46 \mu\text{Gy h}^{-1}$ (0.05 – $4.14 \mu\text{Gy h}^{-1}$) (Nguyen et al. 2020). This difference could be due to the fact that different sampling sites had been chosen in the two studies. It is well-known that the chemical composition of the studied ore deposits is inhomogeneous. Notwithstanding, the high outdoor absorbed gamma dose rate was a common finding from both studies.

A comprehensive survey of the background (ambient) gamma radiation in 63 provinces across the country was carried out in the year 2000. This survey reported a national average absorbed dose rate of $0.07 \pm 0.03 \mu\text{Gy h}^{-1}$ (Huy et al. 2012). This national average is 24 and 5 times lower than the average gamma absorbed dose rate of $1.8 \pm 0.9 \mu\text{Gy h}^{-1}$ and $0.37 \pm 0.07 \mu\text{Gy h}^{-1}$ received, respectively, by the groups of the most and the less exposed population in the BX REE deposit area. With such high gamma exposure dose rates, the BX REE deposit area can be considered a high background radiation area (HBRA) (UNSCEAR 2000). There are several

well-known HBRA worldwide as, for example, Ramsar (northern Iran), Kerala (a coastal area in the west of India) and Guarapari (a coastal area in the eastern Brazil). The absorbed dose rates in Ramsar, Kerala, and Guarapari were reported as high as $29.7 \mu\text{Gy h}^{-1}$, $9.8 \mu\text{Gy h}^{-1}$, and $4.0 \mu\text{Gy h}^{-1}$, respectively (UNSCEAR 2000; Mortazavi and Mozdarani 2012; Derin et al. 2012). Table 7 presents a comparison of the activity concentrations of primordial radionuclides in soils along with absorbed gamma dose rate reported for typical HBRA worldwide as compared to those found in the present study.

As can be seen from Table 7, the gamma dose rates in outdoor air of HBRA contrast with the world average value of absorbed dose rate calculated from activity concentrations of radionuclides in soils from ordinary areas (59 nGy h^{-1} for outdoor gamma absorbed dose rate). For corresponding indoor gamma absorbed dose rate and total annual gamma effective dose values of 84 nGy h^{-1} and 0.48 mSv y^{-1} are reported, respectively (UNSCEAR 2000). The high radiation background in Ramsar region was attributed to the high content of ^{226}Ra in the mud brought to surface by the water of hot springs (Mortazavi and Mozdarani 2012). The high gamma radiation in Kerala and Guarapari areas was reported to be due to monazite minerals containing high ^{232}Th levels (Derin et al. 2012; Veiga et al. 1999; Pfeiffer et al. 1981). The high radiation background in Selama district, Perak State of Malaysia was explained by the high content of ^{238}U and ^{232}Th in the metamorphic granite composition of soil (Ramli et al 2009).

From the present study it is clear that REE ore deposits may also generate high gamma dose rates to the public, due to the primordial radionuclides associated with the REE ore. The total annual gamma external effective dose (AGED) to the public living around (0.3–0.5 km) the BX REE deposit area investigated here resulted in an average annual effective dose of $11.1 \pm 5.3 \text{ mSv y}^{-1}$ (range from 6.2 to 16.9 mSv y^{-1}) (Table 4), i.e., 22 times higher than the world average annual effective dose of 0.48 mSv y^{-1} originated by radionuclides in soils (UNSCEAR 2000).

Radiation from cosmic rays

The dose rate from cosmic radiation over the study area was measured at $30 \pm 7 \text{ nGy h}^{-1}$, corresponding to $0.26 \pm 0.07 \text{ mSv y}^{-1}$. Typically, at sea level the total world average effective dose from charged particles, photons and neutrons of cosmic rays is 0.29 mSv y^{-1} (UNSCEAR 2000). Considering that the dose rate from cosmic rays depends on altitude (Bouville and Lowder (1988)) the average effective dose caused by the cosmic radiation at the altitude of 1200 m above sea level at the BX REE deposit area can be estimated to be as high as 0.48 mSv y^{-1} . Compared to that, the dose rate measured at the investigated area was almost two times lower. This difference is probably due to the uncertainties in the experimental dose-rate measurements. In any case, the results show that the dose from cosmic radiation at Bat Xat is much lower than the doses from radionuclides in REE ore and soils.

Table 7 Comparison of absorbed gamma dose rate in outdoor air of some well-known high background radiation areas (HBRA) worldwide

Country	Area	Characteristics of area	Activity concentrations, Bq $\text{kg}^{-1}\text{d.m.}$			Absorbed gamma dose rate in air, $\mu\text{Gy h}^{-1}$	References
			^{232}Th	^{226}Ra	^{40}K		
Brazil	Guarapari	On streets	nd	nd	nd	0.09–1.7 ^a	Pfeiffer et al (1981)
		A beach, non-inhabited area	nd	nd	nd	0.09–90.0 ^a	Veiga et al (1999)
Iran	Ramsar	Spring water	nd	nd	nd	0.80–17.0 ^a	Mortazavi and Mozdarani (2012)
China	Yangjiang, Quandong province	A coastal area, non-inhabited area	na	na	na	0.37 ^a (average)	Wei et al (1990)
India	Kerala	A coastal area, non-inhabited area	2071–41,280	213–6023	222–2531	0.2–4.0 ^b 1.8 (average)	Derin et al (2012)
Malaysia	Selama district, Perak state	Agricultural land and inhabited area	628 ± 169	196 ± 43	475 ± 89	0.08–1.04 ^b	Ramli et al (2009)
Vietnam	Muong Hum, Lao Cai	Agricultural land and inhabited area at 0.3–0.5 km apart the ore body	2293 ± 832	800 ± 716	459 ± 236	1.0–2.8 ^b (1.8 ± 0.9)	This study
Worldwide average and range			30 11–64	35 17–60	400 140–850	0.059	UNSCEAR (2000)

nd: not determined; a: directly measured in the field; b: calculated based on the activity concentrations of primordial radionuclides in soil

Annual committed effective dose from ingestion

Results in Table 3 show that for all the cooked food the activity concentrations of ^{232}Th and ^{226}Ra (assumed in equilibrium with ^{238}U) were lower than the MDA of 0.13 and 0.15 Bq kg^{-1} d.m., respectively. The low activity concentration of ^{238}U and ^{232}Th in food is expected because of the generally low transfer factors (TFs) of these nuclides from soil to plants (IAEA 2010; Li and Shang 2008). In particular, Luu (2014) has studied the TFs of uranium and thorium from soil to colza (*Brassica juncea*) grown on Ferralic Acrisols, which is the typical soil type in the Bat Xat district, and reported the TFs for ^{232}Th and ^{238}U to be 0.029 ± 0.001 and 0.017 ± 0.003 , respectively.

The radionuclide ^{40}K was determined in all the foods and drinking water with activity concentrations ranging from 28 ± 6 to 48 ± 13 Bq kg^{-1} d.m. (Table 3). The radionuclide ^{40}K was the major contributor of the annual committed effective dose (ACED) to the population of the study area. The ACED for adults in the BX REE deposit area was estimated to be as high as 0.25 mSv y^{-1} (Table 5), out of which ^{40}K contributed up to 96%, and the remaining 4% was from ^{232}Th (see electronic supplement for the calculation). This estimated annual dose was in the range of doses obtained for other countries in Asia, as well as worldwide. For example, Iyengar et al. (2004) reported that the ACED from ingestion to the populations of Bangladesh, China, India, Japan, Pakistan, Philippines, Republic of Korea, and Vietnam, ranged from 0.20 to 0.34 mSv y^{-1} . Tran et al. (2018) reported that the ACED for adults in the Red River delta region of Vietnam ranged from 0.24 to 0.42 mSv y^{-1} with an average of 0.32 mSv y^{-1} . Reeba et al. (2017) reported that Indian population living in a high-background radiation area in Kerala would be exposed to an ACED of 0.41 mSv y^{-1} due to the ingestion of primordial radionuclides, mainly ^{40}K . For the population worldwide, the ACED was assessed to be 0.29 mSv y^{-1} (UNSCEAR 2000).

Annual effective dose from radon and thoron inhalation

Results for radon and thoron are shown in Table 6 can be arranged into three groups. The first group, Nos. 1–3, represents radon concentrations in the indoor air at Hanoi city, with an average ^{222}Rn concentration of 58 Bq m^{-3} ($41\text{--}74 \text{ Bq m}^{-3}$). This concentration is two times higher than the ^{222}Rn concentration recorded during the 1990s of $28 \pm 11 \text{ Bq m}^{-3}$ ($n=208$) (Nguyen et al. 1996). It should be noted that the number of measurements made in the present study was much smaller than that in the 1990s. Despite of this weakness, the present result may indicate that this difference in indoor radon concentrations could be due to the renovation of buildings during the last decades. In the Hanoi

urban area there has been intensive construction of modern type dwellings built with different construction materials and reduced ventilation and better insulation in comparison with older houses. Nonetheless, the area remains a region with low indoor radon concentrations, well below the recommended maximum value of 100 Bq m^{-3} (WHO 2009).

The second group, Nos. 4–16, represents the concentrations of ^{222}Rn and ^{220}Rn in indoor air of dwellings located at a 1 to 2 km distance from the REE ore body. Here, the average indoor concentration was 88 Bq m^{-3} ($64\text{--}130 \text{ Bq m}^{-3}$). The third group, Nos. 17–20, represents ^{222}Rn and ^{220}Rn concentrations in the indoor air of dwellings located at 300–500 m distance from the REE ore body. For this group, the average indoor concentration was 504 Bq m^{-3} ($304\text{--}896 \text{ Bq m}^{-3}$). These results clearly show that the closer to the ore body the higher are ^{222}Rn and ^{220}Rn concentrations in indoor air. It is assumed that radon and thoron concentrations determined in the investigated dwellings are representative for the annual average concentrations, but seasonal fluctuation may exist. It was noticed that radon and thoron concentrations in indoor air do not display a single proportion, which might be due to the variable concentration of parent radionuclides in the clay used to build the house walls. Thus, in the clay used to build, especially, the houses Rn15, Rn19, and Rn20, the ^{232}Th content was probably higher than the ^{238}U content allowing for ^{220}Rn concentrations in indoor air to be markedly higher than those of ^{222}Rn (Table 6).

It is widely recognized that prolonged exposure to airborne radon and thoron and its short-lived daughters can be detrimental to human health (WHO 2009). For protection of the members of the public against the harmful effects of ionizing radiation and reduction of the lung cancer risk caused by high radon concentration in indoor air, the US EPA introduced a so-called Action Level (AL) for ^{222}Rn in American dwellings of 148 Bq m^{-3} (4 pCi L^{-1}) (US EPA 1999). In the European Union, as a transition towards the radon reference level recommended by WHO, a maximum tolerable value for ^{222}Rn concentration in indoor air was proposed at 300 Bq m^{-3} (ICRP 2009) applicable to dwellings and workplaces. Furthermore, when the annual average radon concentration exceeds 1000 Bq m^{-3} a remedial action is mandatory to abate radon concentrations (EU 2013). These radon concentration levels aim at keeping the effective radiation dose to members of the public below the radiation dose limit recommended by current international radiation safety standards (IAEA 2014).

In brief, the estimated annual effective dose due to ^{222}Rn and ^{220}Rn inhalation for the population living in apartments in Hanoi city was $2.2 \pm 0.4 \text{ mSv}$. For the members of the population living at 1–2 km distance from the BX REE deposits the annual effective dose was $4.0 \pm 1.6 \text{ mSv}$, while for those living closer at a distance of 300–500 m

from the ore deposits the dose was of $26.3 \pm 9.2 \text{ mSv y}^{-1}$ (Table 6). For the most exposed group of Bat Xat population, and taking into account the dose from radon inhalation ($26.3 \pm 9.2 \text{ mSv y}^{-1}$), the external dose from radiation emitted by radionuclides in the soil ($11.1 \pm 5.3 \text{ mSv y}^{-1}$), the cosmic radiation ($0.26 \pm 0.07 \text{ mSv y}^{-1}$), and the dose from ingestion of radionuclides with the diet (0.25 mSv y^{-1}), the individual total annual effective dose averages 38 mSv.

The excess lifetime cancer risk

The excess lifetime cancer risk (ELCR) is a parameter helping to evaluate radiation-induced health risks. The world average value of ELCR is $2.9 \cdot 10^{-4}$ (UNSCEAR 2010). Values for the study area are consistently one to two orders of magnitude higher than the world average and indicate a non-negligible higher risk of cancer incidence especially among the residents living close to the REE ore body (Table 4). Currently, there are 23 families with a total of 132 individuals living in close proximity (within 300–500 m distance) to the REE ore body. These inhabitants are exposed to doses from radon inhalation which is comparable to the exposure of the most exposed individuals in the Ramsar (Iran) population. Furthermore, in several houses near the REE ore body, radon (^{222}Rn plus ^{220}Rn) concentrations in indoor air largely exceeded 1000 Bq m^{-3} (Table 6) and intervention is recommended.

Potential measures to reduce the exposure of this population to ionising radiation from the REE ore were considered. One possible measure could be to cover the ore bodies with a thick layer of compacted soil, which would shield from external gamma radiation and could reduce radon emanation from the ore. However, radon in soils diffuses also laterally and, consequently, indoor radon concentrations would probably not be sufficiently reduced. Implementation of radon countermeasures such as sealing the floor is not practicable in an area of low income. The most effective and cheapest way to abate the radiation exposure would be to relocate those families living close to the ore bodies into another area farther away from any REE deposit.

Conclusions

Analysis of ^{232}Th , ^{226}Ra and ^{40}K activity concentrations in soils, foods and drinking water, plus radon (^{222}Rn) and thoron (^{220}Rn) measurements in indoor air, were carried out in an area rich in REE deposits in Bat Xat district, Lao Cai province, Vietnam, for assessing the radiation exposure of the population. The activity concentrations of ^{232}Th and ^{226}Ra in soil from the ore body were 5.7 and 3.0 times higher, respectively, than in soil from kitchen gardens located at a distance of 300–500 m from the ore body,

and 22 and 9 times higher than in soil from a lowland rice field located at about 2 km from the ore body. The BX REE deposits are responsible for a high gamma radiation giving rise to an external effective dose of up to $11.1 \pm 5.3 \text{ mSv y}^{-1}$, which is 23 times higher than the world average of 0.48 mSv y^{-1} . This result confirms that the investigated area must be considered as a high background radiation area (HBRA).

The activity concentrations of radionuclides in local foods and drinking water were comparable with those reported from regions without REE deposits, and the annual committed effective dose due to radionuclide ingestion was 0.25 mSv y^{-1} , which is comparable to the corresponding values in the Asian region and worldwide, estimated at 0.29 mSv y^{-1} . The effective dose due to cosmic radiation at the study area was 0.26 mSv y^{-1} , a value which is close to the global average of 0.29 mSv y^{-1} .

The annual average effective dose from inhalation of radon and thoron by the population living at 1–2 km distance from the ore body was $4.0 \pm 1.6 \text{ mSv}$, while that for the 123 inhabitants currently living within 300–500 m distance from the ore body was much higher, i.e., $26.4 \pm 9.2 \text{ mSv}$. Consequently, the total annual effective dose to the population living close to the REE ore body was as high as 38 mSv, i.e., 16 times higher than the world average of 2.4 mSv y^{-1} .

These results highlight that the major contribution to total effective dose is from radon inhalation, up to 70%, followed by the contribution from external gamma radiation (29%), while ingestion of radionuclides with food and water accounted only for a minor contribution (1%) to the total effective dose. Moreover, the results of the measurements of thoron (^{220}Rn) suggest that this short-lived radon isotope, often neglected in radon surveys, may result in a higher contribution to the effective dose from inhalation than the longer-lived radon isotope ^{222}Rn , e.g., in dwellings Rn15, Rn19, and Rn20 (Table 6).

Although there is no mine developed on the Bat Xat REE deposits, the natural radiation background in the area is considered very high. Based on the results from this work, there is no need for the radiation risk management to ban consumption of local food and use of local water sources. In contrast, based on the results obtained in the present study it is recommended to the local government of Lao Cai province to offer alternative lands farther away from the ore body, at 2 km or more, to those 23 families (132 persons) who currently are living close to the ore body, and to support the construction of new houses. This resettlement would drastically reduce radiation exposure of those families and protect them from an increased risk for stochastic effects of ionization radiation such as cancer, thus contributing to an improved public health. Furthermore, the use of clay from the area with REE deposits in house construction shall be avoided. Instead, use of clay containing trace uranium and thorium from other areas is recommended,

in order to decrease the levels of indoor radon and thoron concentrations.

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Author contributions NVD: formal analysis, software; DDT: investigation; DDN: supervision, methodology, writing original draft; CFP: methodology, validation, writing-review and editing; DVT: formal analysis; NHQ: validation. All authors read and approved the final manuscript.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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