



Radioactive Thoron ^{220}Rn Exhalation From Unfired Mud Building Material Into Room Air of Earthen Dwellings

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Thoron (^{220}Rn), an isotope of radon with a strong α -decay energy, and its short-lived metallic progeny can pose an elevated lung cancer hazard in room air when unfired-soil derived building materials are used in earthen dwellings. Changes in moisture content and density influencing the thoron exhalation rate from earthen materials into room air were studied in the laboratory with terra rossa from a village on the Đồng Văn Karst Plateau Geopark, Việt Nam, where ethnic minorities construct traditional dwellings with unfired terra rossa walls and floors. Our results show that the thoron exhalation rate from mud surfaces depends on (i) the content of radioactive parental nuclides in mineral components; (ii) the moisture content of mud where ~5–10 weight % water maximizes the ^{220}Rn exhalation rate; and (iii) the density of dry mud as primarily controlled by internal macroscopic voids, fractures, and porosity. Additional time-series of ^{220}Rn exhalation data from an interior mud wall of a terra rossa-built house under different seasonal and weather conditions show that the temperature is influencing thoron exhalation *via* the water vapor pressure deficit (VPD) in air and the associated amount of atmospheric moisture adsorbed onto indoor mud surfaces. Our data suggest that occupants of “mud house” earthen dwellings in northern Việt Nam are exposed to an increased thoron geohazard during cooler weather, low VPD, and high relative humidity in air. Detailed studies are needed to evaluate the thoron geohazard for inhabitants of mud-built dwellings in other climates and geological terrains.

Keywords: earthen dwelling, inhalation hazard, radioactivity, mud house, radon isotope, room air, soil brick, thoron

INTRODUCTION

Most radioactivity in room air of buildings is typically caused by radon isotopes and their radioactive metallic progeny after exhalation of radon isotopes from building materials, soil, sediment, and rock containing parental radionuclides. Over half of the natural environmental radiation dosage in room air originates from radon-emitting building materials (Meisenberg and Tschiersch, 2010; Cevik et al., 2011; Trevisi et al., 2013). An abundance of studies is available about the radiation geohazard from the radon isotope ^{222}Rn with a comparatively long half-life

of 3.84 days (Samuelsson and Petterson, 1984; Sahoo et al., 2007). In fact, the name of the element radon is often used uncritically and synonymously for the isotope ^{222}Rn (Faheem and Matiullah, 2008; Chen et al., 2010; Sakoda et al., 2010; Hassan et al., 2011; Bavarnegin et al., 2013; Yang et al., 2019), whereas few studies are concerned with the exhalation of the radon isotope ^{220}Rn called thoron with a half-life of only 55.6 s (Tuccimei et al., 2006; Hosoda et al., 2007; Ujčić et al., 2008, 2010; Kanse et al., 2013). The relatively short half-life of ^{220}Rn and its limited transport distance *via* diffusion and convection from solid sources into room air often results in the common misconception that thoron exhalation and its radiation geohazard from building materials are negligible. As a consequence, thoron and its progeny have not been considered in official safety thresholds of indoor radiation exposure (Nazaroff and Nero, 1988; Urosević et al., 2008; Meisenberg and Tschiersch, 2010; Ujčić et al., 2010). Moreover, thoron is rarely measured individually because the detection of short-lived thoron requires specialized equipment that can discriminate between different α -decay energies, thus often side-lining thoron quantification in radon studies (e.g., Steinhäusler, 1996). In light of the World Health Organization (WHO) (2009) efforts to reduce radiation from radon, proper recognition of the thoron radiation geohazard is important, especially for inhabitants of unfired soil-derived dwellings with significant concentrations of thoron in room air (Meisenberg and Tschiersch, 2011; Meisenberg et al., 2017).

Exhalation rates of radon isotopes from minerals and their subsequent transport into room air depend on the concentration and distribution of parental radionuclides in soil/rock-derived building materials, on the size and shape of grains, porosity, moisture content, temperature, and meteorological conditions (Balek and Beckman, 2005; Hassan et al., 2009; Sakoda et al., 2010, 2011; International Atomic Energy Agency (IAEA), 2013). This study addresses the thoron geohazard from terra rossa that is typically utilized by economically disadvantaged ethnic minorities for the construction of traditional earthen dwellings (i.e., mud houses) in mountainous regions of northern Việt Nam. Such mud houses are constructed by compacting moist, local terra rossa to gradually build up walls (Figures 1A–C) with a thickness of ~ 60 cm. A typical family of six to eight people, encompassing three generations, lives in a mud house with bare earthen walls and floor. Surveys of indoor air proved that the radiation geohazard of ^{220}Rn substantially exceeds the average environmental background thoron concentration in outdoor air, especially at close distances to earthen walls and floor (Nguyễn-Thùy et al., 2019), while thoron and its progeny account for > 80 % of the combined average effective dose and represent the predominant radioactive inhalation hazard for residents of mud houses (Nguyễn-Vân et al., 2020).

Gypsum-based plaster covering on mud walls and ceramic tiles on floors can provide effective diffusion barriers for radon, yet such modern home improvements are not affordable for most inhabitants of mud houses. Bare earthen walls and floors are able to exchange moisture with ambient room air that is seasonally providing high levels of humidity. The objective of this study is to reproducibly determine and

compare the ^{220}Rn exhalation rates from unfired terra rossa building material (i.e., representing typical mud walls in traditional earthen dwellings) under various weather conditions. The results of this study are guiding us toward devising economically affordable and culturally acceptable surface treatment options in mud houses to remediate the thoron geohazard in room air.

MATERIALS AND METHODS

Origin of Mud, and Preparation of Experimental Mud Bricks

Clay-rich terra rossa was collected in December 2016 in the Đồng Văn Karst Plateau Geopark, Hà Giang Province ($23^{\circ}11'$ N, $105^{\circ}03'$ E) at a location where villagers traditionally collect material for the construction of their earthen dwellings (i.e., mud houses) (Figures 1A,D,F). After removing surface litter, ~ 100 kg of moist terra rossa was transferred from a depth interval of ~ 20 to ~ 100 cm into polyethylene bags and transported to Việt Nam National University (VNU) in Hà Nội where the terra rossa was air-dried for 1 month, powdered, and sieved to a grain size of ~ 2 mm to eliminate pebbles, leaves, and roots.

Kilogram aliquots of powdered terra rossa were moistened by addition of 0.2 L of water, kneaded to achieve homogeneity, and manually pressed into a square wooden mold with an interior dimension of $11 \times 11 \times 7.5$ cm to produce 22 standard-sized mud bricks (Figure 1E) varying in wet weight from 1.2 to 2.0 kg. The variance was mainly caused by the inclusion of air bubbles when limited pressure was applied by hand during compaction. After their release from the mold, the mud bricks were air-dried for 1 week and re-weighed. Further moisture and volume losses were monitored from first stage measurements on January 9, 2017, until final measurements on December 6, 2017, to document the bricks' surface area, weight, and bulk density. We started with 22 original mud bricks in the first stage, but that number decreased for later stages because some of the original bricks received various surface coverings to evaluate the effect on exhalation of radon isotopes. The water content of mud bricks during the first stage of measurements was determined for ~ 2 -g brick fragments (i.e., < 0.1 wt. % of a total brick) in wt. % *via* their weight loss after 24 h of heating to 105°C , whereas the moisture contents of bricks during the second and third stages of measurements were determined gravimetrically for whole bricks after prolonged drying at room temperature between measurement stages.

Grain Size

Three randomly chosen samples of collected terra rossa were used for analyses of their grain size distribution. Prior to laser diffraction of wet samples with a Horiba LA 950 Grain Size Analyzer (HORIBA, Ltd., Kyoto, Japan) at VNU, organic matter was removed according to Mikutta et al. (2005). Three milliliters of 6 wt. % aqueous hydrogen peroxide (H_2O_2) was used to treat organic matter in approximately 10 g of terra rossa over 1 h at room temperature, followed by fivefold rinsing with deionized water, centrifugation, and disposal of the supernatant water.



FIGURE 1 | (A) Local mining of terra rossa for the construction of (B) Mud houses in the Đồng Văn Karst Plateau Geopark, Hà Giang Province, northern Việt Nam. (C) Closeup view of the exterior wall of a mud house. The scale on the left is in cm. (D) Collecting terra rossa in Yên Minh District for manufacturing of (E) Mud bricks in the laboratory to measure ²²⁰Rn exhalation rates. (F) The black rectangle on the map of Việt Nam indicates the location of the Yên Minh District where terra rossa was mined for mud houses and mud bricks. Pictures of quarry (A) and mud house (B) courtesy of Jian Liu, Indiana University.

Metallic Radionuclides in Terra Rossa

The metallic radionuclide occurrence in terra rossa was evaluated by randomly selecting two samples of dried terra rossa from the total collected material. Additional sieving yielded ~15-g aliquots (equivalent to a volume of ~10 cm³) of the 1-mm grain size fractions. After oven-drying at 105 °C for 12 h, the two samples were analyzed for activities of metallic radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K at the Analytical Center for Multi-Elemental and Isotope Research, Siberian Branch of the Russian Academy of Sciences, Novosibirsk, Russia (SB, RAS) *via* high-resolution semiconductor γ -spectrometry (Melgunov et al., 2003; Gavshin et al., 2005). Low concentrations of radionuclides required an extended counting time from 12 to 48 h. The accuracy and reproducibility of the analyses were controlled by parallel measurements of Russian National Geological Reference Materials, including samples of SG-1A, SG-3, SG-2, DVG, DVT, ZUK-1, BIL-1, and ST-1A (Govindaraju, 1994). The overall statistical error in the determination due to the random process of decay was < 5 % for all analytical γ lines. Detection limits for ²²⁶Ra and ²³²Th in the measured samples were ~0.04 Bq, while the limit for ⁴⁰K was 0.3 Bq.

The combined activity concentrations of radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in terra rossa samples are expressed as the radium equivalent activity (Ra_{eq}) that is calculated *via* the algebraic expression in Eq. 1 (OECD Nuclear Energy Agency (NEA-OECD), 1979; United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1982, 2000; Tufail, 2012):

$$Ra_{eq} = C_{226-Ra} + 1.43C_{232-Th} + 0.077C_{40-K} \quad (1)$$

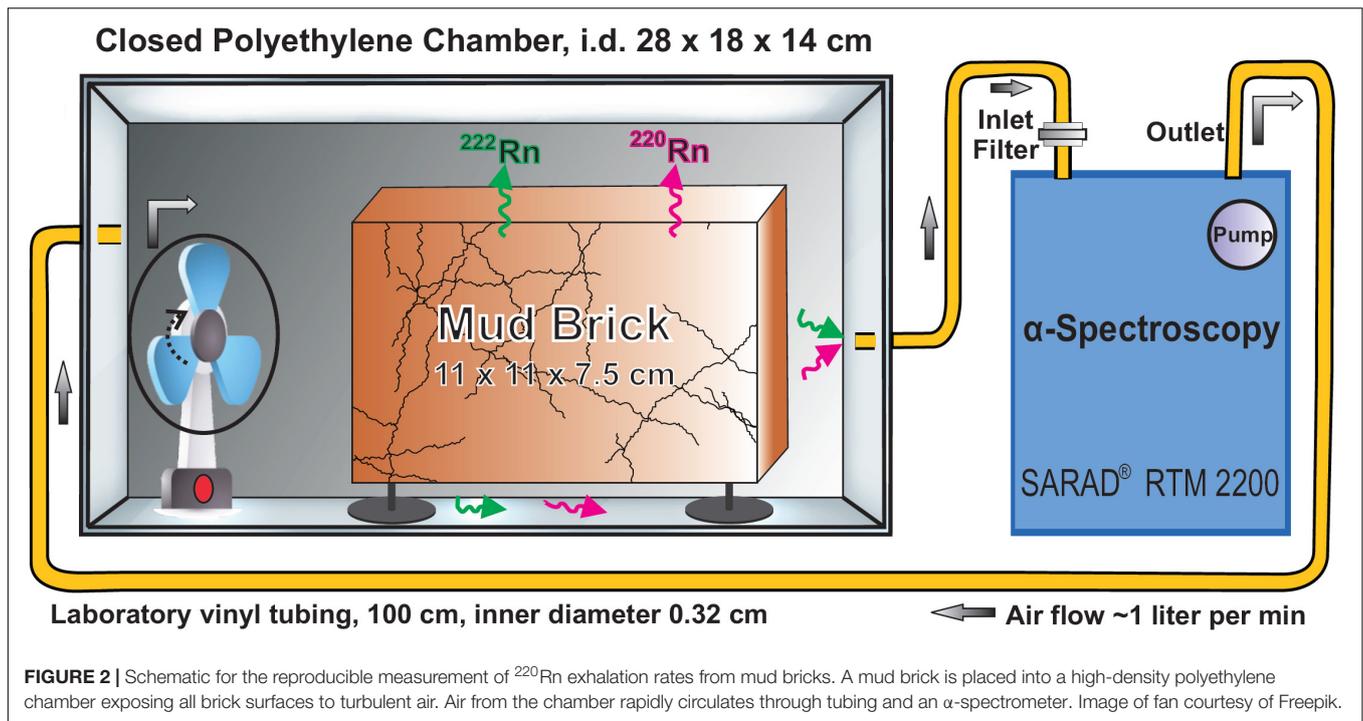
where Ra_{eq} is the radium equivalent γ -activity (Bq kg⁻¹), and C_{226-Ra} , C_{232-Th} , and C_{40-K} are the activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in Bq kg⁻¹, respectively.

Exhalation Rate of ²²⁰Rn From Mud Bricks

The thoron exhalation rate from mud bricks was repeatedly measured in 2017 under standard conditions in turbulent air from a chamber (Figure 2) in three stages on January 9, January 18, and December 6 with increasing dryness of the mud bricks. A single mud brick was placed into an airtight high-density polyethylene chamber where all brick surfaces were exposed to turbulent air. Forced convection was achieved by a small fan (Figure 2). The interior dimension of the chamber measured 28 × 18 × 14 cm with a wall thickness of 2 mm. Air from the chamber rapidly streamed at a flow rate of 1 L min⁻¹ in closed-circuit mode through laboratory vinyl tubing, a 1.0- μ m air filter, and a SARAD® RTM 2200 α -spectrometer (SARAD GmbH, Dresden, Germany) with an internal membrane pump. The concentration of ²²⁰Rn was measured in 10-min intervals for at least 0.5 h. The ²²⁰Rn exhalation rate is based on Eq. 2:

$$E_{Tn} = \frac{C_{Tn} V \lambda_{Tn}}{A} \quad (2)$$

where E_{Tn} is the ²²⁰Rn exhalation rate (Bq m⁻² h⁻¹; Tuccimei et al., 2006; Kanse et al., 2013); C_{Tn} is the equilibrium concentration of ²²⁰Rn in the air of the chamber (Bq m⁻³); V is the volume of air surrounding the brick in the chamber (m³); A is the geometric surface area of the mud brick (m²); and λ_{Tn} is the ²²⁰Rn decay constant (0.0126 s⁻¹).



²²⁰Rn Exhalation in a Mud House

A full-sized mud house was constructed near Hà Nội (Figure 3A) with original moist terra rossa building material from Hà Giang Province, northern Việt Nam, to monitor the long-term ²²⁰Rn exhalation from mud walls throughout drying and under changing weather conditions. After completion of the construction in November 2017, a portable SARAD® Thoron Scout (SARAD GmbH, Dresden, Germany) was placed at a distance of 20 cm from an interior mud wall and 50 cm above the mud floor (Figures 3C–E). A computer fan drew air from the space between the mud wall and a wooden board through a central hole in the board into a perforated polyethylene enclosure that contained the SARAD® Thoron Scout. Although the Thoron Scout is a passive thoron detection device without an internal pump, the active air flow from an external fan makes its thoron data compatible with those from the SARAD® RTM 2200 with an internal pump (Schimmelmann et al., 2018). Hourly average ²²⁰Rn isotope concentrations in room air were automatically logged by the Thoron Scout from November 8, 2017 until January 14, 2018, with a small hiatus in early January. Measurements resumed from June 2 until August 18, 2018.

Meteorological time-series of daily mean air temperature and relative humidity were recorded at the Hà Nội–Nội Bài International Airport ~40 km north of the mud house location (Weather Underground¹). A regional precipitation time-series covering the entire 2018 monsoon season in Hà Nội was obtained from Meteoblue². The water vapor pressure deficit (VPD) corresponds to the difference between the actual amount of

water vapor in the air and the theoretical amount of water vapor at 100 % relative humidity. VPD is primarily dependent on temperature, and to a lesser extent on barometric pressure. A higher VPD indicates more intense drying of moist surfaces. To determine the VPD for outdoor air, we first calculated the actual vapor pressure at dew point temperature and the saturation vapor pressure at air temperature using Tetens’ formula (Monteith and Unsworth, 2013). Dew point temperature was calculated with coefficients A = 237.3 and B = 17.269 (Weiss, 1977). In addition to ²²⁰Rn concentrations, the SARAD® Thoron Scout instrument recorded temperature, barometric pressure, and relative humidity of the air inside the mud house as input for VPD calculation. Outdoor temperature and humidity data next to the mud house near Hà Nội were recorded in 30-min intervals by an Elitech GSP-6 temperature and relative humidity logger (Elitech, London, United Kingdom).

Adsorption of Atmospheric Moisture on Terra Rossa Mud

The initial moisture contents of five terra rossa fragments (~1.5 g each) of a brick and seven fragments (10–70 g each) from the interior wall of a mud house (Figure 3A) were gravimetrically determined *via* weight loss in weight % (wt. %) after heating at 105 °C for 24 h. Three dried fragments from brick and seven dried fragments from the mud wall were placed next to water-soaked sponges in a hermetically closed chamber 40 × 60 × 6 cm, without any direct contact between liquid water and terra rossa samples. The chamber with close to 100 % relative humidity in air was kept at 25 °C in the laboratory. The fragments were re-weighed daily for 14 days to monitor any weight changes owing to adsorption of moisture.

¹<https://www.wunderground.com/>

²<https://www.meteoblue.com>

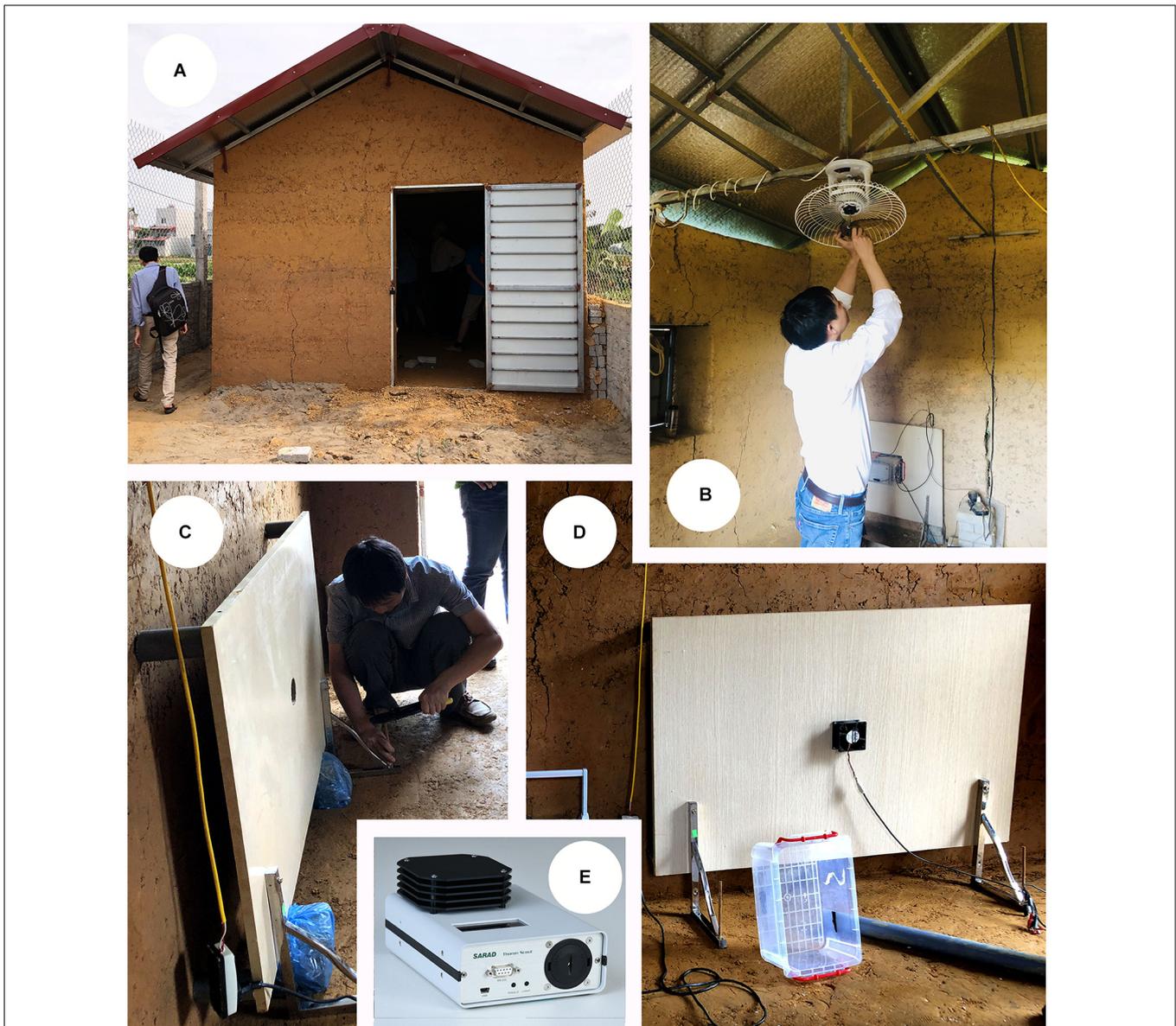


FIGURE 3 | (A) Mud house near Hà Nội that was constructed with terra rossa from Hà Giang Province and was completed in November 2017. **(B)** An oscillatory ceiling fan provides optional turbulence for room air. The background shows the SARAD® Thoron Scout assembly in position ~50 cm above the floor. **(C)** White board with central hole to draw air from close to the mud wall. **(D)** White board with fan attached. The perforated translucent polyethylene container will house the **(E)** Thoron Scout when attached against the white board.

RESULTS

Physical Properties of Terra Rossa Samples

Terra rossa represents a silty clay loam according to the classification of the U.S. Department of Agriculture (Garcia-Gaines and Frankenstein, 2015). The sand, silt, and clay size fractions within terra rossa’s grain size range from 0.1 to 150 μm are 12.8, 36.9, and 50.2 wt. % on a dry basis, respectively.

The surface area and density of mud bricks were calculated according to measured changes in brick size and weight along

slow drying (Table 1). The water content of bricks decreased through the three stages of measurements, whereas the bulk density and surface area decreased slightly (Table 1).

Adsorption of Atmospheric Moisture on Terra Rossa

Fragments of terra rossa from the interior side of the ~60-cm thick wall of a mud house near Hà Nội 3 years after its construction (consisting of similar terra rossa as used for the manufacture of mud bricks) averaged 4.2 (±1.1) wt. % moisture. In the 5 days prior to sampling of fragments during October

TABLE 1 | Surface area (cm²), bulk density (g cm⁻³), and water content (wt. %) of cuboid-shaped mud bricks after air-drying through three experimental stages, and ²²⁰Rn (thoron) concentrations (Conc.) and surface exhalation rates (Surface exh.) of mud bricks in the polyethylene chamber.

Stages of measurements	Parameters of mud bricks			²²⁰ Rn (thoron)		
	Surface area (cm ²)	Density (g cm ⁻³)	Water (wt. %)	Conc. (Bq m ⁻³)	Surface exh. (kBq m ⁻² h ⁻¹)	
First stage (January 9, 2017) <i>n</i> = 19	Min	519	1.4	20.1	344	1.75
	Max	611	1.9	23.0	1832	9.31
	Aver.	559	1.6	21.5	1200	6.01
	SD	25.8	0.1	1.0	389	1.88
Second stage (January 18, 2017) <i>n</i> = 12	Min	472	1.3	6.3	1288	7.63
	Max	606	1.8	14.3	2419	12.27
	Aver.	541	1.5	9.8	1964	10.01
	SD	32.8	0.1	2.5	290	1.16
Third stage (December 6, 2017) <i>n</i> = 18	Min	472	1.3	0.6	713	4.09
	Max	571	1.7	5.5	1288	6.31
	Aver.	522	1.5	2.6	970	5.30
	SD	30.7	0.1	1.3	123.3	0.63

The surface area is the sum of the areas of all faces of a brick. Aver., average; SD, standard deviation; *n*, the number of measured mud bricks.

2020, the outdoor temperature and outdoor relative humidity in the mud house averaged ~24 °C and ~75 %, respectively. After exposure of the dried fragments for about 2 weeks to laboratory air at 25 °C and about 100 % relative humidity, the terra rossa samples had gained on average more than 5 wt. % of moisture (Figure 4).

Concentrations of ²²⁶Ra, ²³²Th, ⁴⁰K, and Their Radium Equivalent in Terra Rossa

The concentrations of metal radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in two terra rossa samples DYM and SYM were measured by semiconductor γ-spectrometry (Table 2). The two terra rossa samples show similar respective concentrations with averages of 52.0 Bq kg⁻¹ for ²²⁶Ra, 68.5 (±2.1) Bq kg⁻¹ for ²³²Th, and 259.0 (±1.0) Bq kg⁻¹ for ⁴⁰K.

²²⁰Rn Exhalation Rate From Experimental Mud Bricks

Measured concentrations of ²²⁰Rn in chamber air surrounding mud bricks and calculated ²²⁰Rn exhalation rates (Table 1) encompassed large ranges during the first stage of measurements when bricks still contained ≥ 20 wt. % moisture. Values became more stable in the second and third stages (Figure 5). Maximum ²²⁰Rn exhalation rates were observed during the second stage with a mean value of 10.01 (±1.16) kBq m⁻² h⁻¹ in comparison to the first stage at 6.01 (±1.88) kBq m⁻² h⁻¹ and the third stage at 5.30 (±0.63) kBq m⁻² h⁻¹.

²²⁰Rn Exhalation Rate From Interior Mud Wall in Mud House

Time-series of the ²²⁰Rn concentration in air close to the inside wall of a mud house near Hà Nội are compared to meteorological time-series of outdoor Hà Nội air in Figure 6. The ²²⁰Rn concentration started low in November of 2017 when the ~60-cm thick mud walls were still glistening wet

from recent construction. Over the following months, the mud walls gradually dried and the ²²⁰Rn exhalation increased until June of 2018. Drying was most intense by the end of June when high summer temperatures increased the outdoor air VPD. Continued drying during falling temperatures in the later part of the monsoon season witnessed decreasing ²²⁰Rn concentrations.

A 9-day time-series of ²²⁰Rn concentration in indoor air of the mud house near Hà Nội with ventilation controlled by an optional oscillating ceiling fan (Figure 3B) in October 2020 is represented in Figure 7 together with temperature and VPD of indoor and outdoor air. The large variability of hourly mean ²²⁰Rn concentrations suggests dramatic changes in air flow and air residence time between the white board and the mud wall in the single-room mud house (Figures 3B–D). No discernible correlation with temperature or VPD is apparent over the 9 days of measurements when the moisture content of the thick mud wall was essentially constant. Air turbulence introduced in the first part of the time-series by the oscillatory fan limited the thoron concentration in air that was admitted to the Thoron Scout by the computer fan blowing air from behind the white wall into the perforated polyethylene enclosure. Turning the ceiling fan off during the fifth day of measurements almost doubled the average thoron concentration in the polyethylene enclosure from 202 to 382 Bq m⁻³ (Figure 7).

DISCUSSION

Sources of Radioactivity in Terra Rossa

Radioactive nuclides like ²³⁸U of uranium, ²²⁶Ra of radium, ²³²Th of thorium, and ⁴⁰K of potassium occur naturally in bedrock and soils, as well as in mineral-containing construction materials. While ²²²Rn is a product from the decay of ²³⁸U via ²²⁶Ra, ²²⁰Rn (thoron) is produced along the ²³²Th decay chain. The measured concentrations for ²²⁶Ra (mean value of 52 Bq kg⁻¹) and ²³²Th (mean value of 68.5 Bq kg⁻¹) in terra

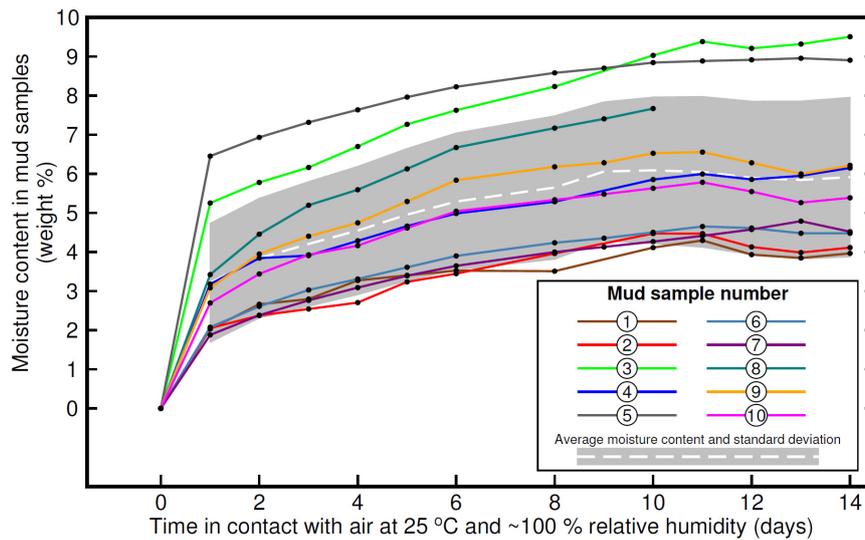


FIGURE 4 | Time-series of adsorption of water to pre-dried (at 105 °C) mud fragments from the inner wall of a mud house while exposed to air with ~100 % relative humidity at 25 °C. The running average and its standard deviation are shown by a white dashed line and a gray band, respectively.

rossa samples are above their corresponding worldwide average concentrations in natural soils (i.e., 35 Bq kg⁻¹ for ²²⁶Ra and 30 Bq kg⁻¹ for ²³²Th), whereas the concentrations of ⁴⁰K in terra rossa samples fall below the worldwide average concentration of 400 Bq kg⁻¹ for ⁴⁰K in soils (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1982, 2000). Elevated activities of ²²⁶Ra and ²³²Th in terra rossa foster the exhalation of radon isotopes from exposed mud surfaces into indoor air.

The radiological hazard from ²²⁶Ra, ²³²Th, and ⁴⁰K activities in terra rossa was calculated *via* the radium equivalent activity, Ra_{eq} (OECD Nuclear Energy Agency (NEA-OECD), 1979; United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1982, 2000). The calculated average radium equivalent activity of the two terra rossa samples DYM and SYM of 169.9 (±3.1) Bq kg⁻¹ is lower than the criterion limit of 370 Bq kg⁻¹ (OECD Nuclear Energy Agency (NEA-OECD), 1979; Ravisankar et al., 2014), which makes an annual dose of 1.5 mGy equal to the maximum permissible annual effective dose of 1 mSv for the general public as recommended by the International Commission on Radiological Protection (ICRP) (1991).

With regard to Ra_{eq} alone, terra rossa would be a safe building material for mud houses in the studied area. However, it would

be misleading to only take γ-radiation into account and ignore α-radiation. Thoron is an intermittent nuclide along the thorium decay chain, which originates from primordial ²³²Th. Thoron’s transport distance in indoor air is limited owing to the short half-life of 55.6 s. As a consequence, thoron’s spatial distribution in room air is inhomogeneous (Meisenberg and Tschiersch, 2011). Building materials are recognized as one of the main sources of radon isotopes in indoor air. The concentration of thoron in indoor air declines with increasing distance from mud walls and floors toward the center of the room. High concentrations of α-emitting thoron and progenies in indoor air of mud houses in the center of rooms (> 100 Bq m⁻³) and especially near bare mud walls (> 350 Bq m⁻³) are responsible for > 80 % of the average effective dosage affecting inhabitants (Nguyễn-Thùy et al., 2019; Nguyễn-Văn et al., 2020). Severely elevated concentrations of thoron up to 1500 Bq m⁻³ close to bare mud walls of traditional dwellings were also found in China (Shang et al., 2008) and in an experimental room constructed with unfired mud bricks in a laboratory at the Helmholtz Zentrum in Munich, Germany (Meisenberg and Tschiersch, 2011), although in both cases ²³²Th in the mud building material only produced γ-radiation with an average activity concentration of 40–50 Bq kg⁻¹. Thoron concentrations in room air adjacent to surfaces of unfired mud are directly linked to the content of ²³²Th in the building material. Thus, the combined α- and γ-radiation geohazards of the studied terra rossa call into question the safety of unfired terra rossa as a building material.

TABLE 2 | Activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K (Bq kg⁻¹) together with their radium equivalent activity (Ra_{eq}, Bq kg⁻¹) in two terra rossa samples.

Terra rossa sample	Activity concentration (Bq kg ⁻¹)			Ra _{eq} (Bq kg ⁻¹)
	²²⁶ Ra	²³² Th	⁴⁰ K	
DYM	52	70	260	172.1
SYM	52	67	258	167.7

Influence of Terra Rossa Bulk Density

The bulk density of experimental mud bricks was primarily influenced by the inclusion of air bubbles when pressing wet terra rossa into a mold to shape bricks, and later decreased *via* drying as water in pore spaces was replaced by air (Table 1). During the first stage of measurements, when the

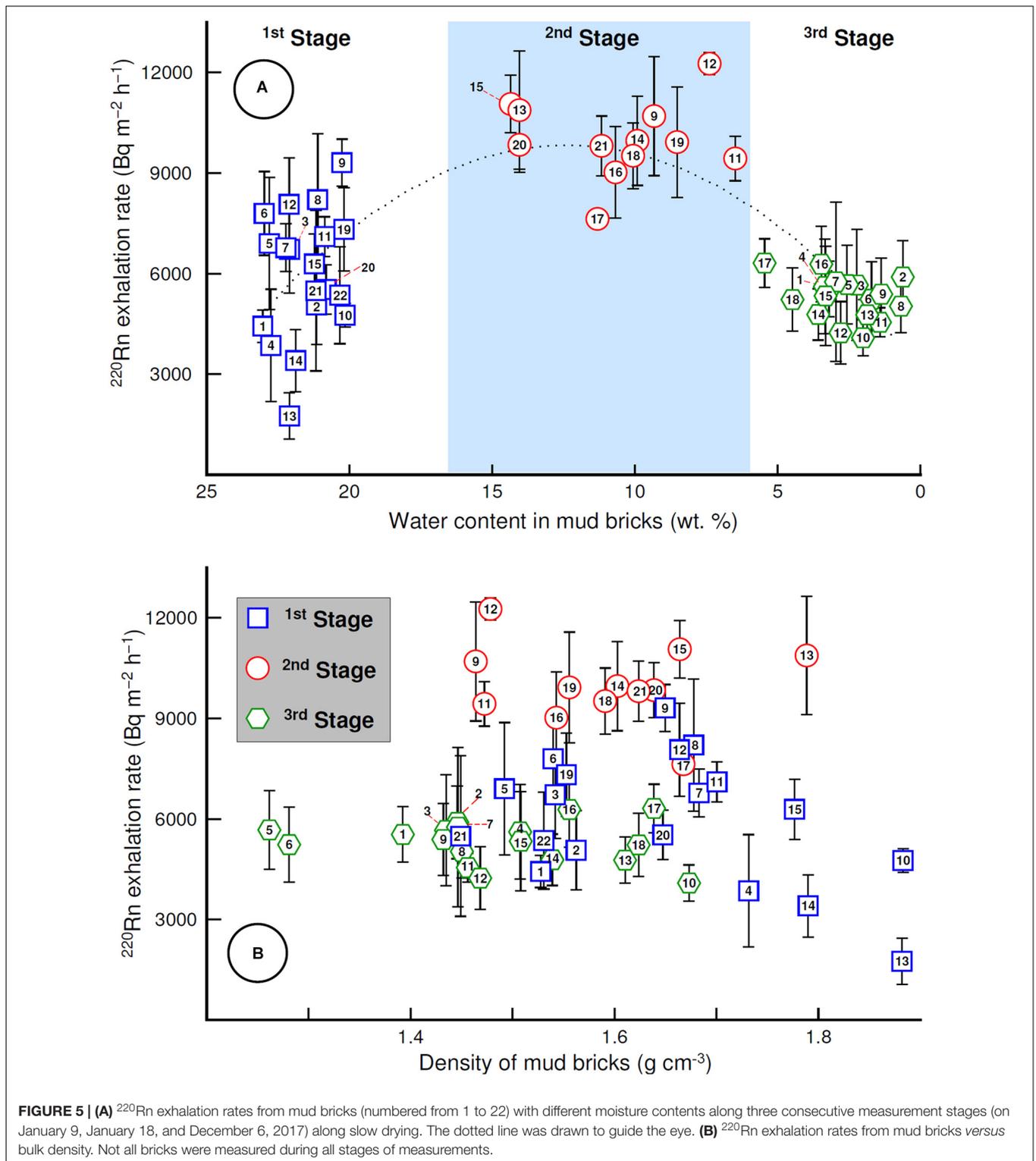


FIGURE 5 | (A) ²²⁰Rn exhalation rates from mud bricks (numbered from 1 to 22) with different moisture contents along three consecutive measurement stages (on January 9, January 18, and December 6, 2017) along slow drying. The dotted line was drawn to guide the eye. **(B)** ²²⁰Rn exhalation rates from mud bricks versus bulk density. Not all bricks were measured during all stages of measurements.

mud bricks were still relatively wet with ≥ 20 wt. % water, the high bulk density from 1.5 to 1.9 g cm⁻³ primarily meant the exhalation rate of ²²⁰Rn because radon atoms recoiling out of the mineral grains were predominantly retained in water-filled pore space (Rogers and Nielson, 1991; Faheem and Matiullah, 2008) (Figure 5B).

bulk density in the presence of liquid water decreased the exhalation rate of ²²⁰Rn because radon atoms recoiling out of the mineral grains were predominantly retained in water-filled pore space (Rogers and Nielson, 1991; Faheem and Matiullah, 2008) (Figure 5B).

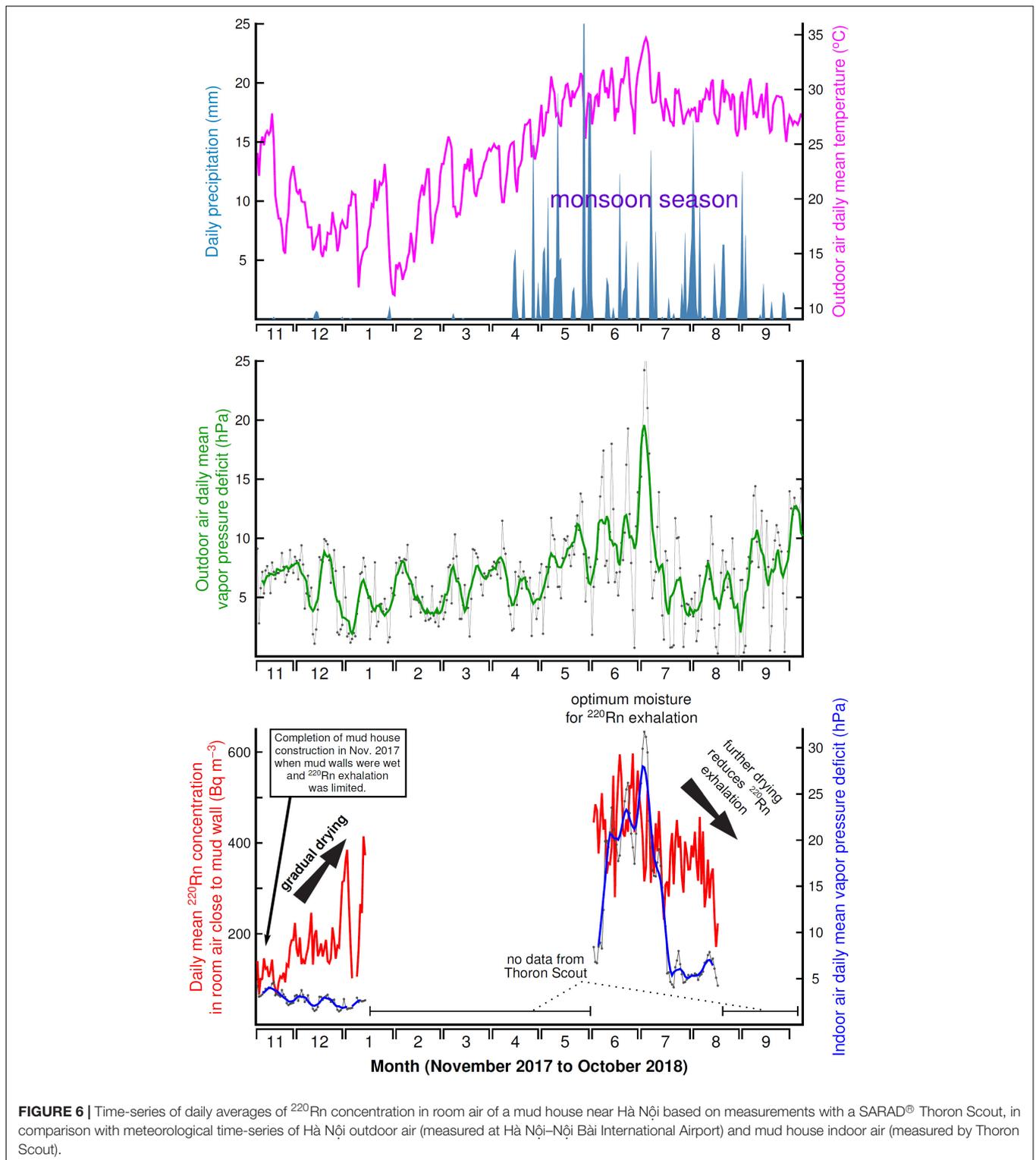
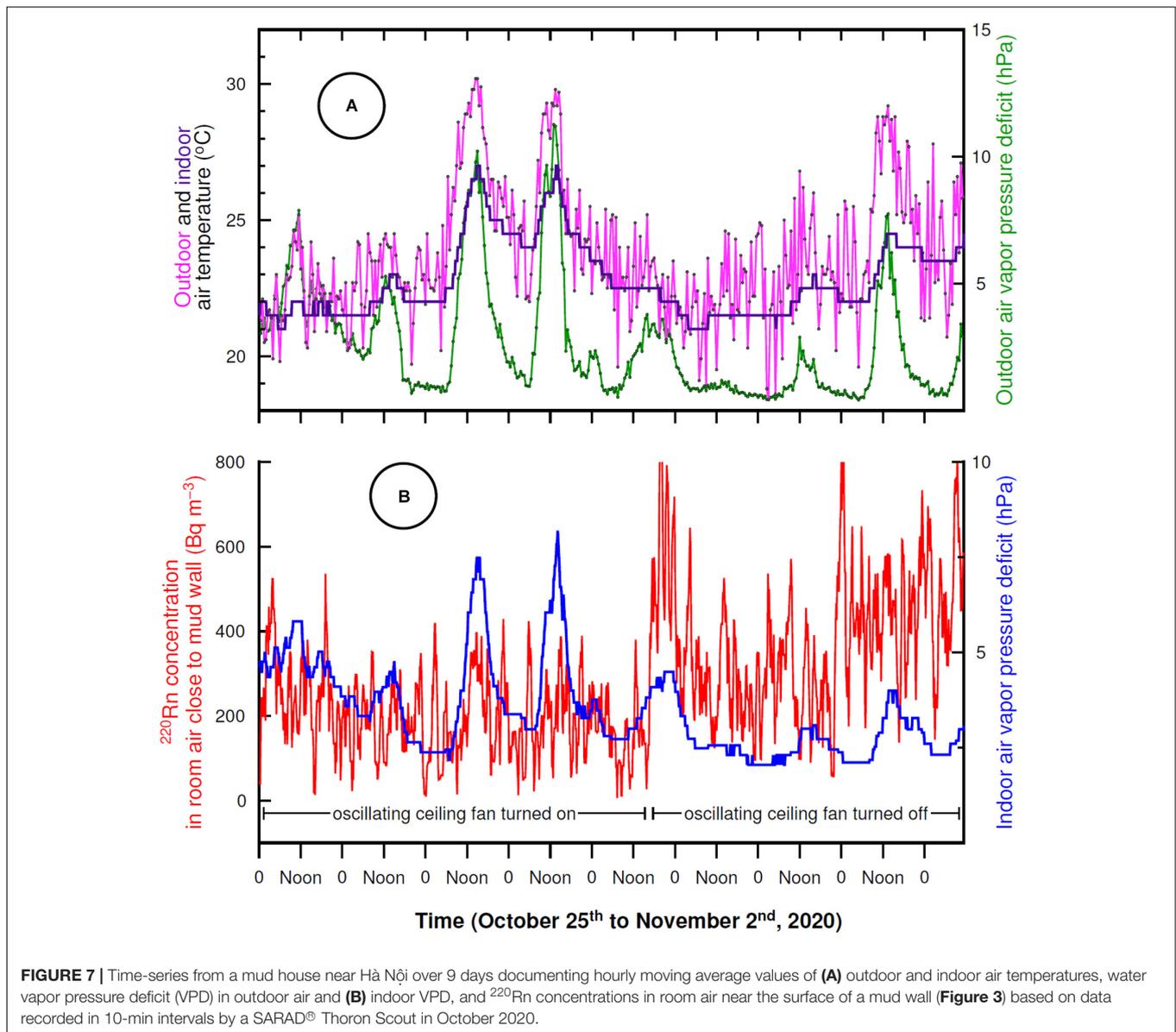


FIGURE 6 | Time-series of daily averages of ^{220}Rn concentration in room air of a mud house near Hà Nội based on measurements with a SARAD® Thoron Scout, in comparison with meteorological time-series of Hà Nội outdoor air (measured at Hà Nội–Nội Bài International Airport) and mud house indoor air (measured by Thoron Scout).

Shrinkage of drying mud bricks caused the formation of open cracks that likely connected internal voids in the bricks to outside air. During the second stage of measurements with reduced moisture content, the strongly turbulent air in the chamber also forced convection and air flow into

macroscopic cracks, and thus assisted in the exhalation of ^{220}Rn (Figure 5B). During the third stage of measurements with low moisture contents of < 5 wt. %, the bulk density had strongly decreased in comparison with the first and second stages (Table 1). The decreased moisture content in



mud bricks filled pore spaces and cracks almost completely with air and provided diffusion pathways for radon (Faheem and Matiullah, 2008), but fewer recoiling ²²⁰Rn atoms slowed down enough to remain in open pore space and become available for exhalation.

The Role of Residual Moisture for Thoron Exhalation From Terra Rossa

Our results from progressive drying of mud bricks across three stages of measurements emphasize the importance of moisture content (Figure 5A and Table 1) and indicate highest ²²⁰Rn exhalation rates of up to 12.27 kBq m⁻² h⁻¹ with a mean of 10.01 (±1.16) kBq m⁻² h⁻¹ during the second stage at an intermediate moisture content of 9.8 (±2.5) wt. %. The observed residual moisture content in mud bricks during the second stage falls into

the range of typical values of 8–13 wt. % moisture in soil that was reported to maximize radon exhalation (Hosoda et al., 2007; Faheem and Matiullah, 2008; Yang et al., 2019).

The principle of maximized exhalation of ²²⁰Rn from mud surfaces with an intermediate moisture content of between 5 and 10 wt. % was confirmed by time-series of measurements in the room of a research mud house near Hà Nội (Figure 6) where the mean moisture content on the inside of the ~60-cm thick wall of the mud house was still 4.2 (±1.1) wt. % almost 3 years after construction, at the tail end of the summer monsoon season in mid-October of 2020. Laboratory experiments with fragments of terra rossa samples from the interior wall of the mud house indicated a weight gain commensurate with a moisture content between ~4 and ~9 wt. % upon prolonged exposure to air with high humidity (Figure 4). A dry interior wall or mud floor of a mud house will adsorb moisture from the air

until it reaches a dynamic equilibrium between adsorption and drying. The ~60 cm thick walls of traditional mud houses can store a substantial amount of moisture that acts as a reservoir and not only prevents rapid drying over hours to days, but also buffers against fast accumulation of adsorbed moisture close to interior mud surfaces on days when the relative humidity in air is high and the water VPD is low. In effect, diurnal and short-term weather changes cannot rapidly affect the thoron exhalation from interior mud surfaces (Figure 7). However, longer-term to seasonal changes in VPD significantly influence the residual moisture in terra rossa and the thoron exhalation.

Practical Considerations for Mud House Inhabitants

The moisture loading in northern Việt Nam's (sub)tropical ambient air is highly variable throughout the year, which is mostly due to the large temperature difference between summer and winter (Figure 6). The temperature range across seasons at higher altitude in Hà Giang Province is even larger than in Hà Nội. Low temperatures in mountainous Hà Giang Province combined with low clouds, fog, and high relative humidity in winter decrease VPD in air, increase adsorption of moisture into exposed mud to possibly reach ~10 wt. %, and temporarily increase the exhalation of radon isotopes from mud into indoor air of mud houses. Occupied mud houses traditionally use open wood fires for cooking and have no chimney. Over time, ash from cooking fires, fingerprints, animal droppings, and aerosols may deposit hygroscopic salts on interior mud surfaces increasing the potential for adsorption of moisture and enhanced ^{220}Rn exhalation.

Paradoxically, although the moisture loading in air is higher during the warm summer monsoon months, the higher summer temperatures greatly increase VPD, which dries mud surfaces inside of mud houses and limits the exhalation of ^{220}Rn from mud into room air (Figure 6). The observed high concentrations of ^{220}Rn in the mud house in June can be explained by residual moisture that derived from the compaction of wet terra rossa earlier in November 2017 (Figure 6) when the house was newly constructed.

Forced turbulence of room air in a mud house by an oscillating ceiling fan reduced the residence time of ^{210}Rn -laden air close to mud surfaces, mixed the air from near walls and floors with overall room air on the order of seconds, and thus effectively diluted the ^{210}Rn near mud surfaces (Figure 7). Without forced turbulence, the air flow close to mud surfaces tends to be laminar and better retains exhaled ^{220}Rn near mud surfaces. The large short-term variability of ^{210}Rn concentrations near a mud wall that was observed regardless of forced turbulence may be due to the convective flow of room air as influenced by meteorological parameters, such as temperature gradients between mud surfaces and outdoor and/or indoor air, as well as wind forcing air through cracks into the room. Forced turbulence of room air by electrical fans is not useful for remediation of the thoron geohazard in room air because (i) it merely dilutes thoron in air near mud surfaces with overall room air and (ii) it does not diminish the health hazard from inhaling metallic radioactive

progeny of radon isotopes from room air. Exhalation of radon isotopes from porous earthen substrates can only be stopped by impermeable surface treatments that need to be economically and socially acceptable. Additional research is needed for different types of earthen dwellings in different climates to devise suitable mitigation strategies.

CONCLUSION

Thoron exhalation rates from terra rossa bricks with different densities and moisture contents in conjunction with concentrations of prominent γ -emitting radionuclides call into question the radiological safety of terra rossa and possibly other unfired earthen materials for uses as building material. The average ^{226}Ra and ^{232}Th activities of the studied terra rossa samples are 52.0 and 68.5 Bq kg⁻¹, respectively, which are above the average values of 35 Bq kg⁻¹ for ^{226}Ra and 30 Bq kg⁻¹ for ^{232}Th in average soil around the world (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1982, 2000). The relatively high activities of ^{226}Ra and ^{232}Th radionuclides in terra rossa explain the significant exhalation of radon from terra rossa and the associated α -radiation health hazard in indoor air when terra rossa is used for the construction of earthen dwellings. Exhalation of radon isotopes from interior mud walls and mud floors into room air of mud houses is controlled by multiple characteristics of earthen materials and meteorological parameters, i.e., the mineralogical composition of mud, residual moisture, porosity, permeability, temperature, and water VPD in room air. Especially the moisture content of terra rossa mud is strongly affecting ^{220}Rn exhalation rates. Starting from a low exhalation rate in waterlogged terra rossa from northern Việt Nam, subsequent drying gradually increases the ^{220}Rn exhalation rate until peaking at ~10 kBq m⁻² h⁻¹ at an intermediate moisture content of ~5–10 wt. %, followed by a decline of the exhalation rate toward dryer mud surfaces. Lowered density and associated macroporosity as a result of cracks in drying bricks effectively add surface area for exhalation and increase the thoron exhalation rate. Seasonal variations of temperature and moisture content in ambient air influence the VPD of air and the potential of mud surfaces to adsorb/desorb moisture. Adsorption of moisture into interior mud walls and floors from air at lower temperature and higher relative humidity can temporarily increase the thoron exhalation rate. The present knowledge of the physical and meteorological parameters affecting the exposure of inhabitants of earthen buildings in northern Việt Nam to radiation makes it unlikely that mud houses can be structurally modified to eliminate the radiation geohazard. Our ongoing efforts focus on affordable treatment of mud surfaces to at least limit the transport of exhaled ^{220}Rn from mud walls into room air.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

NN: investigation, design of the study, data curation, and writing. DN-T and HN-V: conceptualization, investigation, design of the study, data curation, writing, review, and editing. NN-H: data curation and writing. AS: conceptualization, investigation, design of the study, writing, review, and editing. All authors contributed to the article and approved the submitted version.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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