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High Indoor Radon Case Study: Influence of Meteorological Parameters and Indication of Radon Prone Area

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Abstract: Indoor radon and meteorological parameters (temperature, humidity, pressure, precipitation, indoor dew point, wind direction, wind speed and heat index) were simultaneously monitored in an old residential house in a radon suspected area. Measurements were performed during the period from winter to summer (13 February 2021–15 August 2021). Indoor radon concentrations were measured with detectors, Airthings Corentium Home (alpha spectrometry method), and meteorological parameters were continuously monitored by the meteorological station WTH600–E (wireless weather station). The influence of geological characteristics in the study area was analyzed, as well as some observed variations and correlations with indoor/outdoor meteorological parameters. The results indicated that indoor radon levels are higher in the spring/summer season than in the winter season. Diurnal radon concentrations varied during measuring period from 303–1708 Bq/m^3 (average 949 Bq/m^3) and 427–1852 Bq/m^3 (average 1116 Bq/m^3) for the living room and bedroom, respectively. Indoor radon concentrations correlated with: outdoor/indoor temperature, indoor humidity (r = 0.45, r = 0.40, r = 0.32, r = 0.56, respectively); indoor dew point (r = 0.53); outdoor barometric pressure (r = −0.26); there were no clear correlation with precipitation and outdoor humidity. The health risk due to long-term, high radon exposure was assessed through the calculated inhalation dose.

Keywords: high indoor radon; meteorological parameters; radon prone area

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1. Introduction

People are mainly exposed to radon in their homes and workplaces. Radon is a serious health risk, and the second most prevalent cause of lung cancer after tobacco smoking [1]. Radon exposure may contribute to approximately 21,800 lung cancer deaths annually in the United States [2]. Some case–control studies of residential radon in North America showed a direct association between prolonged radon exposure and lung cancer [3]. A recent study noted that more than 150,000 men and women in the United States die from lung cancer each year, encouraging awareness about radon testing, mitigation, and ultimately a reduction in lung cancer deaths [4].

Earlier epidemiological studies confirmed the association between residential radon and lung cancer risk, pointing out that an increase of indoor radon concentration by 100 Bq/m^3 causes an increase of lung cancer risk by 16% [5–7]. On the other side, some authors reported that radon-related lung cancers could be prevented in 35–40% of cases if radon levels in residential buildings were reduced below 100 Bq/m^3 [8]. In Ireland, households who lived in an area where 10–20% of homes had indoor radon above 200 Bq/m^3 were 2.9–3.1 times more likely to report a lung cancer diagnosis in comparison to those who lived in areas where is less than 1% homes above 200 Bq/m^3 [9]. The relationship between radon exposure and chronic obstructive pulmonary disease is confirmed by increasing the mortality rate; the hazard ratio per 100 Bq/m^3, 1.13; 95% CI: 1.05–1.21 [10].

In Europe, according to the Directive 2013/59/EURATOM, EU member states should identify buildings (residential and working space) where the average annual radon concentration exceeds the reference level, and encourage a reduction of radon concentrations in these buildings, as well as provide information at the local and national levels on radon exposure and associated health risks [11].

As a natural, radioactive, and noble gas present in lithosphere, radon moves through soil pores by the diffusion process due to concentration gradient; also, the movement of pore fluids through the soil pores in convective transport occurs due to influence of external force [12]. Radon can move through interconnected pores in the soil and penetrate into buildings, regarding the relatively long half-life (3.8 days). Poor ventilation conditions or cracks in the construction systems can lead to radon accumulation in dwellings.

The factors responsible for the degree of radon hazard may exist from different sources, such as: geogenic radon potential, building construction, meteorological factors, lifestyle and habits. Contributions from geogenic and non-geogenic sources to indoor radon concentration are controlled by both, natural and anthropogenic factors. Natural (geogenic) factors are related to radon generation and transport from the ground [13–17], whereas anthropogenic factors relate to the construction characteristics of a building, including building materials and usage patterns [18–21]. Geogenic factors show a geographical trend and a spatial structure depending on geology, soil properties, and hydrology [17,22]. The studies of radon generation and transport through a porous medium and its influence on surface radon exhalation are important for prevention and best building practices due to action measures against radon [23]. Radon accumulation in dwellings does not just depend on radon exhalation from the subsoil/building materials or on forced ventilation. Meteorological factors may be considered with respect to both geogenic and anthropogenic impacts, insofar as they can influence radon transport in the ground, radon migration and accumulation indoors, and construction style and building occupancy patterns [24–28]. Local weather and meteorological conditions have also a strong influence [29,30], like soil moisture, rainfall, temperature, pressure, relative humidity and outdoor dew point [31–34], but the results reported in many studies are not consistent. High indoor temperatures cause upward flow, which decreases basement pressure, leading to radon entry in the building. Varying indoor/outdoor air pressure differences were revealed as key driver for the air exchange and, thus, for the variation of indoor radon concentration [26]. Radon concentrations fall as the relative humidity increases from 30% to 60% when temperature indoors is lower than that outside [35]. Wind speed and wind direction appeared to be the most significant predictors of radon concentration. Generally, both, geological and meteorological parameters likely present synergistic effects on indoor radon concentrations.

This study was conducted based on previous research in this mining/industrial area, which is suspected in elevated soil radioactivity, enhanced indoor radon levels and diverse geological characteristics [36,37]. The primary goals of the present paper were to analyze monthly and seasonal indoor radon variations in a typical ground-floor residential house, to assess the possible impact of geology (radioactivity from soil and stone used for construction) and relationships between indoor radon concentrations and main meteorological parameters, such as pressure, temperature, relative humidity and dew point. The results of this study would be useful to define methods for reducing high radon concentration in this specific region, contributing to an overall understanding of radon behavior.

2. Materials and Methods

2.1. Description of Study Location and Building

The residential house is located at the north part of Kosovo and Metohija, near Kosovska Mitrovica (42.9343° N and 20.8389° E). This family house was built in the 1970's; it is a typical one-story house with a basement excluding forced ventilation. Only natural ventilation is used; it implies periodic brief openings of windows, especially in the morning hours. The ground floor (used for housing) is built of carved stone with a thickness of about 50 cm; the front part is under the ground about 1 m. The stone was delivered from a nearby hill. The house does not have a floor concrete slab, while the ceiling is made of concrete. The walls in both studied rooms are plastered, and covered with paint. The windows are a double wooden with the height of 50 cm, at a mutual distance of 30 cm. There is laminate flooring in living room, and the bedroom has hardboard slabs with a gap of 0.5 cm at the joints. The entrance to the house is east-oriented and it is located 20 m from the foothill with few trees. The outer walls are oriented: North-East (living room) and East-South (bedroom), and both rooms of the house are on the leeward side. There are two other houses on the sides of the studied house at a distance of 15 m.

2.2. Methods of Measurements

Indoor radon levels were measured with the detector Airthings Corentium Home from 13 February to 15 August 2021. Radon detectors were placed on shelves away from doors and windows in the living room and bedroom. The measurement started at the moment when this region was hit by the coldest wave of winter, and completed in mid-summer when temperatures were the highest. The radon data were continuously recorded at the same time of each day (between 2 and 3 p.m.), as well as meteorological parameters.

Detector Airthings Corentium Home measures in range from 0–9999 Bq/m^3 [38]. Detection method is alpha spectrometry, based on the process of radon diffusion into the chamber. The accuracy of device at a typical concentration of 200 Bq/m^3 is 5–10% for measurement period from 7 days to 2 months, and uncertainty for one month measurement is less than 10%. The meter self-calibrates; a calibration was done prior to the measurement. The detector shows first result after 6–24h: long-term (LT) and short-term (ST) average radon concentration. The LT average represents average radon value for current measurement (updated once a day). The ST average shows last-day radon values.

Meteorological parameters were measured using a quality weather station with 5-in-1wireless transmitter sensor (WTH600–E-en– GB_v1.0). The sensor includes a self-emptying rain collector for measuring rainfall, an anemometer for wind velocity, a wind vane for wind direction, a temperature sensor, and a relative humidity sensor. The station was set up in the backyard, while the main monitor was in the bedroom and gave values for indoor temperature and humidity among other outdoor parameters.

In addition, the outdoor gamma dose rate was measured near the house at the height of 1 m above ground (four positions). Indoor gamma radiation levels in rooms were also measured by Geiger counter Radex model RD1503$^+$ supplied by QUARTA-RAD. The Radex monitor operates in range from 0.05 to 9.99 μSv/h with measuring uncertainty of $\pm$ 15%; it gives four values in each measuring circle which are averaged by counter itself. Calibration of the counter was done using 137Cs at 5 μSvh^{-1} (CE certificated from Germany).

2.3. Impact of Geology; Characteristics of Rocks and Soil

Overall, the tectonic Kosovo basin and Metohija valley stand out in the varied relief; the lowest altitude of 297 m, and the highest point is at an altitude of 2656 m. The rim of mountain basin is made up of Palaeozoic schist, Mesozoic limestone, volcanic and metamorphic rocks with layers of marl, sandstone, and coal deposits [39]. The study area is predominantly hilly; the lowest parts are located in the River Ibar valley. The studied house is located at the foothill, a hundred meters from the River Ibar, at an altitude of about 500 m. The industrial complex "Trepča", formerly the largest Pb-Zn mine in Europe is located in vicinity; the landfills are still the main source of air, water and soil contamination.

Strong volcanic activity produced large masses of intrusive materials in the past, including lead and zinc deposits. Process of ore formation is connected with volcanic breccia, which consists of common skarn minerals. Ultramafites have significant extensions; their complexes penetrated by Tertiary volcanic compounds in the southeast region of Kosovska Mitrovica, while volcanic rocks with pyroclastic materials formed mostly of quartz-latites and quartz-porphyries have been observed in Zvečan (hill, 797 m) near Kosovska Mitrovica [40].

According to particle size distribution, the soil nearby the house contains 11.16% sand (200–2000 mm), 43.00% fine sand (20–200 mm), 33.48% silt (<20 mm) and 12.36% clay (<2 mm) [36]. Also, according to some unpublished results of specific activities of radionuclides ^{238}U, ^{232}Th and ^{40}K in soil and stone, their elemental concentrations were calculated; observed values are two times higher than world average for Th and K, and four times for U; in comparison: the world average value for U and Th are 2.64 and 11.1 ppm respectively, whereas for K this value is 1.37% [41]. The soil near the house contains 10.85 ppm U, 20.44 ppm Th and 2.73% K, and stone used to build the house contains 10.53 ppm U, 30.78 ppm Th and 3.23% K (unpublished results).

2.4. Climate

The weather is quite variable from year to year, but commonly with cold winters with less and less snow, and dry/warm summers. Precipitation tends to be the heaviest during the end of the spring and beginning of the summer. The average winter and summer temperatures are 0.6 °C and 20.5 °C, respectively, while spring and autumn have average temperatures of 9.5 °C and 10.6 °C, respectively.

2.5. Dose Assessment

The average annual effective dose (in mSv y^{-1}) for residents of the investigated house was calculated according to the following formula [41]:

$$E = C \times F \times T \times D \quad (1)$$

where *C* is the mean radon concentration in house (in Bq/m^3), *F* is the equilibrium factor between radon and its decay products (taken equal to 0.4 according to the UNSCEAR, 2006) [42]; *T* is the indoor occupancy (taken equal to 7000 h y^{-1}), and *D* is the dose conversion factor for radon decay products assumed by ICRP (0.9 nSv $(Bq\ h\ m^{-3})^{-1}$).

3. Results and Discussion

Indoor radon measurements conducted in two rooms of the house are shown in Figure 1. Diurnal radon concentrations varied from 303–1708 Bq/m^3 in living room (R1) and from 427–1852 Bq/m^3 in bedroom (R2). The values of long-term average (LTA) radon for living room and bedroom were 949 Bq/m^3 and 1116 Bq/m^3, respectively. During the measurement period there were no measurements under reference level recommended by the EURATOM [11]. Although the trend of radon fluctuations in rooms is equable, there are different one-day changes that are noticeable in the first two months of measurements. This may be due to the different ways of heating the rooms (solid fuel and electrical) in the winter period. In spring/summer period, radon concentration tends to be more equal in both rooms, and increases faster in the living room (Figure 1). Another reason for differences in radon concentrations between rooms could be the different floor structures that influence radon diffusion from soil, as well as room ventilation, or living habits of residents. A larger gap thickness yields higher indoor radon concentration from the circumferential joint, as some authors reported that gap contribution to total indoor radon concentration varies from 31% to 62% for joint thickness from 1 to 20 mm, respectively [43].

These measurements are in good agreement with earlier annual radon measurements conducted with CR-39 detectors in two consecutive periods in this house. Ten years ago, radon concentration was 545 and 446 Bq/m^3 in living room, and 1216 and 1034 Bq/m^3 in bedroom, in two consecutive periods, respectively [44]. An average radon concentration of 810 Bq/m^3 (881 Bq/m^3 in December–May 2010/11, and 740 Bq/m^3 in May–December 2011) was reported, pointing out that soil and local stone pose important sources of indoor radon [37].

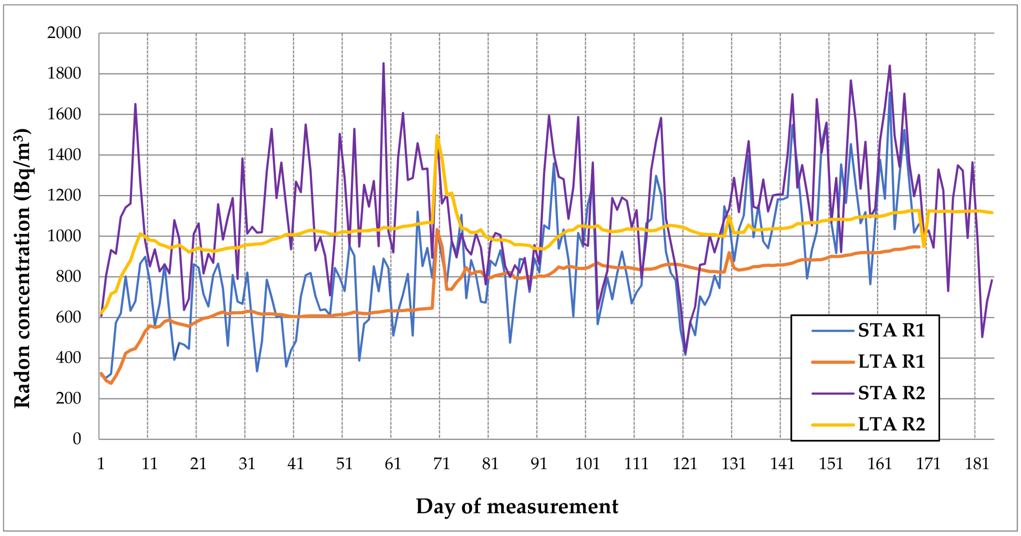

Figure 1. Short term average (STA) and longterm average (LTA) radon concentrations in living room (R1) and bedroom (R2).

The results of outdoor and indoor gamma radiation varied from 0.22–0.34 µSv/h and 0.22–0.38 µSv/h, respectively. The results of indoor gamma dose rate measurements near the walls were in the range of 0.22–0.34 µSv/h, which can indicate enhanced radiation from building materials (stone).

3.1. Influence of Meteorological Parameters

Besides the fact that the radon levels were very high during the measurement period (from winter to summer), a slight increase in radon concentration was observed towards the end of measurement. As shown in Figure 2, the highest average values of indoor radon in both rooms were during summer months, while lower values were measured in the winter months. During the summer, indoor temperature could be higher than soil temperature (a few degrees), causing an unexpected summer stack effect [45]. Building ventilation (induced by opening the windows) might strengthen due to generating air flow.

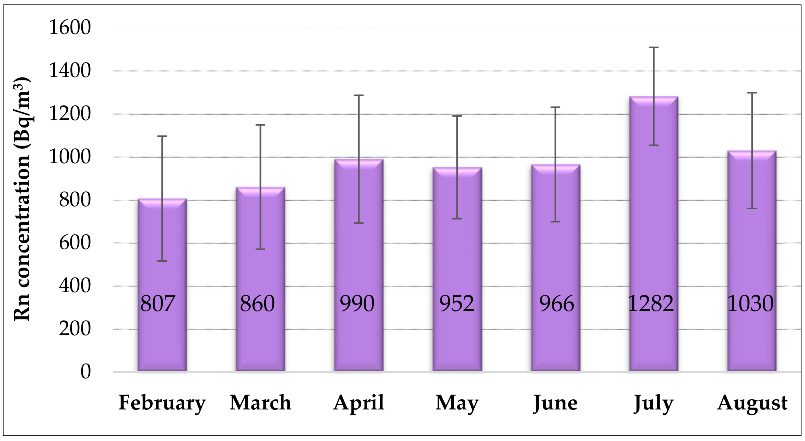

Figure 2. Monthly averaged radon concentrations for the house.

However, some authors reported mixed results about the indoor radon seasonality; the correlation of indoor radon concentration with specific environmental parameters is not consistent, and higher radon concentrations are most often observed in dwellings in the cold season of the year [34,46] and lower radon concentrations in the warm periods [47], while other studies found high indoor radon levels in the summer months [48,49]. The low radon flux in winter was caused by a combination of frozen ground and periodic snow melt, whereas low radon flux in spring most likely resulted from increased precipitation [50].

A sudden radon increase was recorded on the 69th day of measurement due to heavy rainfall a few days earlier (Figure 1). Precipitation saturates the soil and prevents radon from escaping due to a capping effect [31]. Radon exhalation from soil depends from precipitation amounts, and radon diffusion coefficient increase with temperature [51]. Kitto [52] found that radon flux from soil has a slight seasonal pattern, with the greatest exhalation occurring during the late summer months due to the lower moisture content and cracks in the clay soil. A similar pattern is observed in this study. Variations of each meteorological parameter (temperature, relative humidity, pressure, indoor dew point and precipitation) during the measuring period are presented in Figures 3–8, respectively.

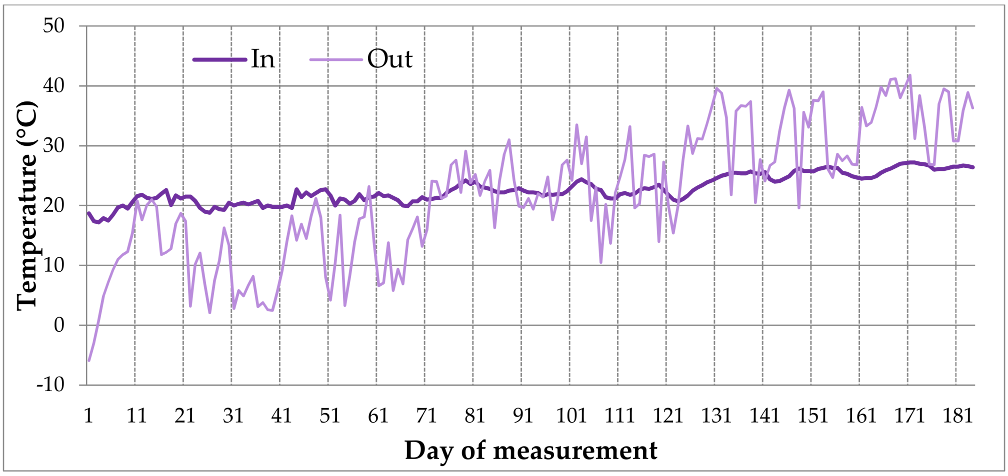

Figure 3. Daily variations of indoor/outdoor temperatures during the measurement period.

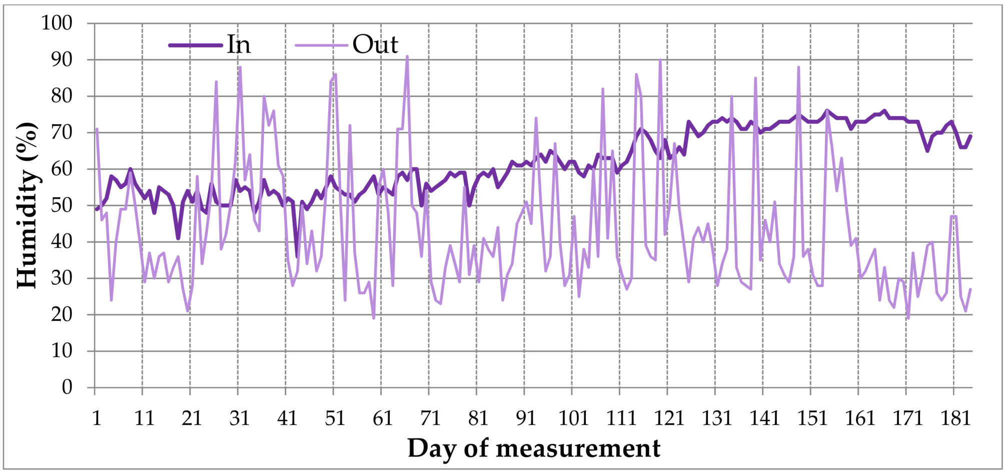

Figure 4. Daily variations of indoor/outdoor relative humidity during the measurement period.

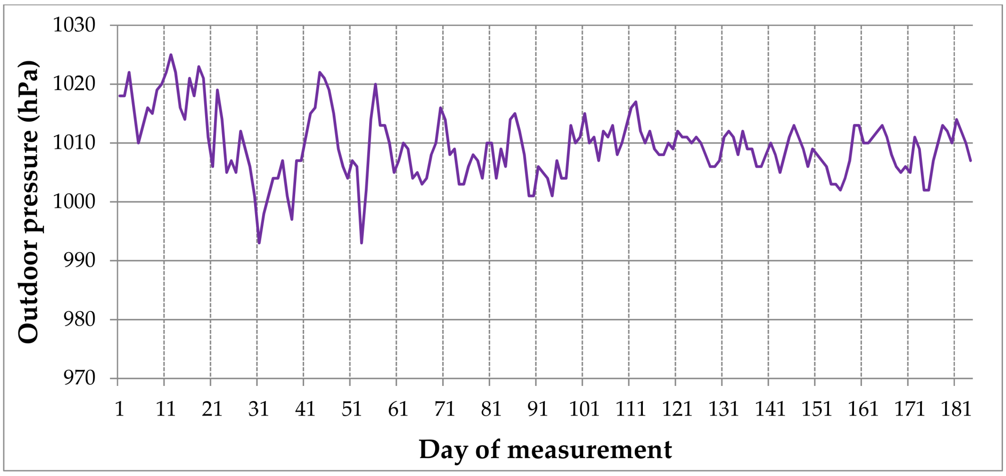

Figure 5. Daily variations of outdoor pressure during the measurement period.

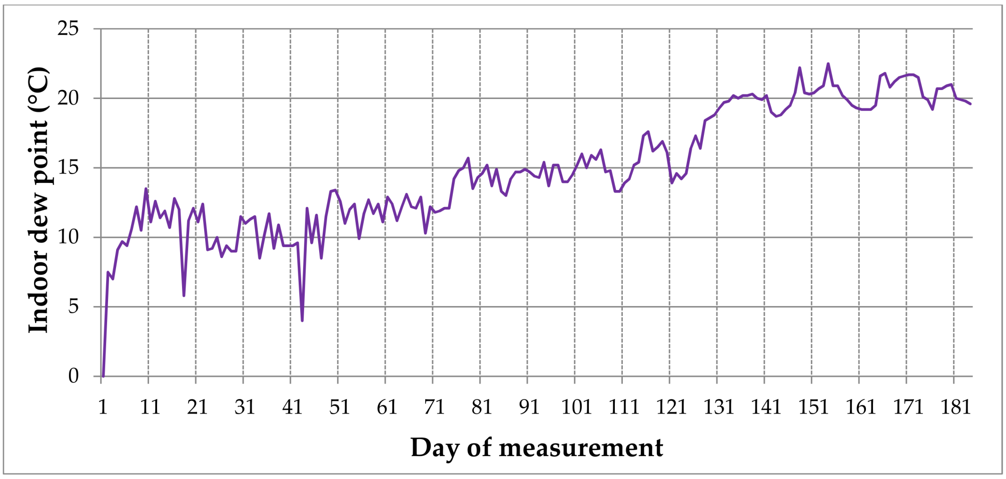

Figure 6. Daily variations of indoor dew point during the measurement period.

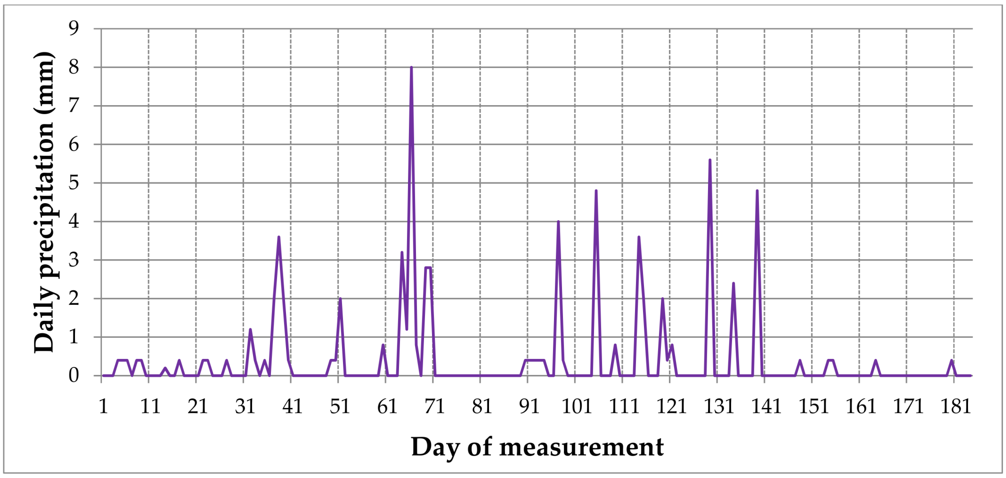

Figure 7. Daily variations of precipitation during the measurement period.

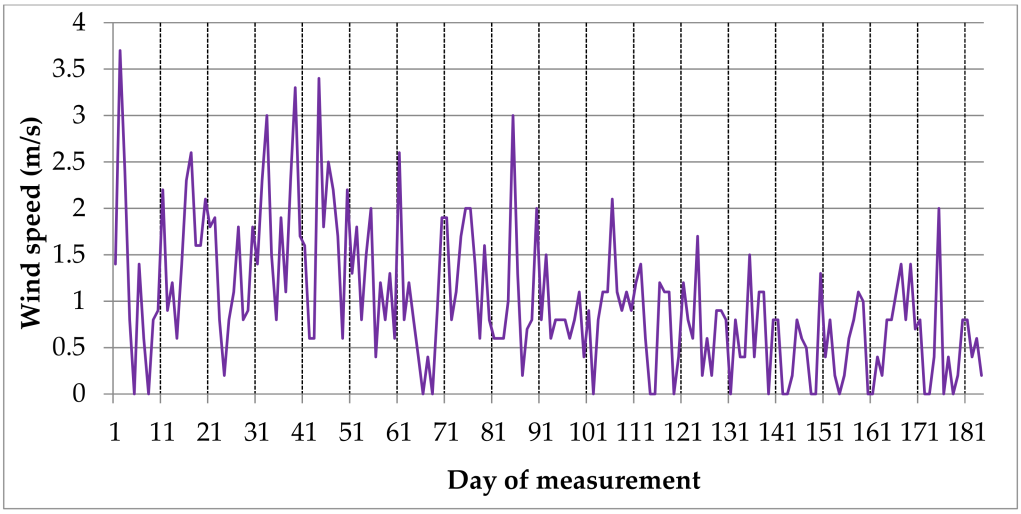

Figure 8. Daily variations of wind speed during the measurement period.

Increases in outdoor temperature, noticeable outdoor/indoor humidity differences, no precipitation, low wind speeds and stable pressure which occurred during the last 30 days (summer period) corresponded to an increase of indoor radon levels. It was found that the indoor radon level increased when relative humidity in air increased. High relative humidity enhances radon exhalation [53]. Although the radon increase is slight, it would be probably related to building materials. Generally, the radon exhalation from building materials increases in summer, when the mean temperature of the building materials is higher than in winter [54].

During the first 70 days of measurement, the outdoor temperature was lower than indoor temperature, and outdoor pressure was variable, which resulted in the lowest radon concentrations on average for the entire measurement period (Figure 2). Indoor radon concentrations showed elevation with increasing indoor/outdoor temperature differences [55]. In this study, it is noticeable from high fluctuations of STA values. Lower radon fluctuations occurred only between the 70th and 90th day of measurement, when the indoor/outdoor temperature differences were lower, and there was no precipitation.

Wind was mostly weak during the measurement period, with an average speed of 1 m/s (max speed 3.7 m/s) in the SSW-SW and NNW-NW prevailing direction. This can enhance radon levels in the house, since high wind speed reducing radon entry rate [56], and in addition wind direction can enhance sub-slab concentration, leading to an increase of radon level [57,58].

The Pearson coefficient of correlation, r, between indoor radon concentration and meteorological parameters provides information how the radon concentration is influenced by these parameters (Table 1). The daily averaged radon concentrations (STA R1 and STA R2) for all measurement period were used for correlation analysis.

Indoor radon concentrations were negatively correlated with outdoor pressure ($r = -0.26$) and wind speed ($r = -0.41$), but positively correlated with indoor/outdoor temperature ($r = 0.45$, $r = 0.40$, respectively), indoor humidity ($r = 0.56$) and indoor dew point ($r = 0.53$). The humidity difference is also positively correlated with indoor radon levels. No clear correlations were found between indoor radon concentrations and the other meteorological parameters (outdoor humidity and precipitation). Florea [59] found a negative correlation with wind speed, precipitation and temperature, while weak correlations with rainfall and mean daily temperature were identified in study of domestic radon in Northamptonshire, UK [60]. Opposite to our study, Xie [34] reported that indoor radon negatively correlates with indoor humidity ($r = -0.3$), outdoor temperature and dew point temperature ($r = -0.3$ and $r = -0.17$, respectively). Aquilina [27] found that indoor radon concentrations weakly correlates with outdoor temperature and relative humidity, but significantly positively correlates (+0.74) with indoor temperature.

Table 1. Daily minimum and maximum values of meteorological parameters, and Pearson correlation coefficient between indoor radon and meteorological parameters.

Meteorological Parameter	Minimum	Maximum	Rn Concentration (Bq/m^3)/Pearson Correlation Coefficient
Temperature (°C) indoors	17.2	27.2	0.45
Temperature (°C) outdoors	−5.9	41.8	0.40
Temperature(°C) differences	−15	24.6	−0.07
Humidity (%) indoors	36	76	0.56
Humidity (%) outdoors	19	91	0.03
Humidity (%) differences	−34	56	0.30
Pressure (hPa) outdoors	993	1025	−0.26
Dew point (°C) indoors	4	22.5	0.53
Precipitation (mm)	0	8	0.09
Wind speed (m/s)	0	3.7	−0.41

3.2. Annual Effective Dose and Health Risk

As indoor radon concentration exceed reference level of 300 Bq/m^3 in this house, the cancer risks from prolonged radon exposure for the inhabitants is emphasized. Based on mean radon concentration, the annual effective dose due to radon inhalation is calculated to 26 mSv; it is much higher than the world average of 1.15 mSv for radon and its decay products [41].

3.3. Indication of Radon Prone Area

Geogenic radon prone areas are regions in which, for natural reasons, elevated indoor radon concentrations must be expected [61]. Radon concentration was elevated (exceeds a recommended threshold) in two independent measurements conducted in recent years by different techniques. An indication of radon risk exists according to the calculated inhalation dose. Some available geological information is very useful when a radon hazard is suspected: gamma dose rate, high uranium activity concentration in soil and stone and, mostly, sand soil texture which allows easier radon transport. Elevated levels of natural radionuclides in the nearby soil are partly a consequence of technological activities [62], such as the mining operations in this area. An additional fact is the land-surface slope and elevation of house: hillsides usually have more permeable and rockier soils, which allows greater radon emanation [58,63]. In addition, high indoor radon in this house may be affected by the proximity of a deep fault zone that allows the radon transport through cracks of permeable soil, as well as the higher porosity of sand and gravel of the river bank that allows increased radon diffusion. All of the results confirmed that this is a radon prone area (RPA), as reported by a recent study [64].

This house needs urgent remediation and mitigation methods, and minimization of indoor radon levels inside the buildings. One of the best ways to reduce the radon concentration is pressurization. The introduction of fresh air and an increase in pressure diluted the radon concentration and prevented gas ingress [23,65]. Otherwise, the houses in whole area should be studied on a case-by-case basis.

4. Conclusions

In the present study, the radon data was recorded February–August 2021 and have been analyzed to correlate with meteorological parameters. According to the goal of this study, the obtained correlations are as follows: indoor radon concentrations were negatively correlated with outdoor pressure (r = −0.26) and wind speed (r = −0.41), but positively correlated with indoor/outdoor temperature (r = 0.45, r = 0.40, respectively), indoor humidity (r = 0.56) and indoor dew point (r = 0.53). No clear correlations were found between indoor radon concentrations and the other meteorological parameters (outdoor humidity and precipitation).

Radon levels are much higher than these proposed by the WHO (100 Bq/m^3), and exceed the recommended level by the EURATOM Directive 2013/59 of 300 Bq/m^3. A high risk of radon exposure was assessed, and a radon prone area was identified. Householders are strongly encouraged to take steps to remediate the situation, such as floor replacement, which could be one of the possible solutions. First of all, householders should enhance ventilation, as this is an important factor that decreases radon indoors. In particular, the properties of geological media to produce and transfer radon gas by mechanisms of diffusion and advection in interaction with the house have to be considered in more details. A public awareness of radon harmfulness must be widespread in this area by supporting and conducting measures for radon reduction, mitigation and remediation, with the aim of protection against exposure to radon.

Many of the residential single-family houses in this region have a comprehensive need to improve the indoor air quality due to the strong predominance of geology structure of the region, ages and constructions of the buildings. In order to obtain information on the long-term effectiveness of introducing requirements on radon prevention, studies in these homes should be continued in the future.

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