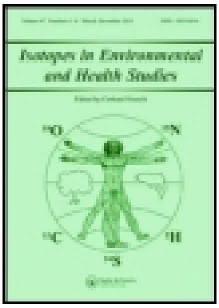

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Zsuzsanna Szabó[ab], Gyozo Jordan[c], Csaba Szabó[a], Ákos Horváth[d], Óskar Holm[e], Gábor Kocsy[f], István Csige[g], Péter Szabó[h] & Zsolt Homoki[h]

[a] Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry

[b] Environmental Physics Department, Centre for Energy Research, Hungarian Academy of Sciences, Budapest 114, Hungary

[c] Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, Budapest, Hungary

[d] Department of Atomic Physics, Eötvös University, Budapest, Hungary

[e] Icelandic Radiation Safety Authority, Reykjavík, Iceland

[f] Radosys Ltd., Budapest, Hungary

[g] MTA Atomki, Debrecen, Hungary

[h] National Research Institute for Radiobiology and Radiohygiene, Budapest, Hungary
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Radon and thoron levels, their spatial and seasonal variations in adobe dwellings – a case study at the great Hungarian plain

Zsuzsanna Szabó[a,b*], Gyozo Jordan[c], Csaba Szabó[a], Ákos Horváth[d], Óskar Holm[e], Gábor Kocsy[f], István Csige[g], Péter Szabó[h] and Zsolt Homoki[h]

[a]*Lithosphere Fluid Research Lab, Department of Petrology and Geochemistry;* [b]*Environmental Physics Department, Centre for Energy Research, Hungarian Academy of Sciences, Budapest 114, Hungary;* [c]*Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, Budapest, Hungary;* [d]*Department of Atomic Physics, Eötvös University, Budapest, Hungary;* [e]*Icelandic Radiation Safety Authority, Reykjavík, Iceland;* [f]*Radosys Ltd., Budapest, Hungary;* [g]*MTA Atomki, Debrecen, Hungary;* [h]*National Research Institute for Radiobiology and Radiohygiene, Budapest, Hungary*

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Radon and thoron isotopes are responsible for approximately half of the average annual effective dose to humans. Although the half-life of thoron is short, it can potentially enter indoor air from adobe walls. Adobe was a traditional construction material in the Great Hungarian Plain. Its major raw materials are the alluvial sediments of the area. Here, seasonal radon and thoron activity concentrations were measured in 53 adobe dwellings in 7 settlements by pairs of etched track detectors. The results show that the annual average radon and thoron activity concentrations are elevated in these dwellings and that the proportions with values higher than 300 Bq m^{-3} are 14–17 and 29–32% for radon and thoron, respectively. The calculated radon inhalation dose is significantly higher than the world average value, exceeding 10 mSv y^{-1} in 7% of the dwellings of this study. Thoron also can be a significant contributor to the inhalation dose with about 30% in the total inhalation dose. The changes of weather conditions seem to be more relevant in the variation of measurement results than the differences in the local sedimentary geology. Still, the highest values were detected on clay. Through the year, radon follows the average temperature changes and is affected by the ventilation, whereas thoron rather seems to follow the amount of precipitation.

Keywords: adobe; indoor radon and thoron; natural radioactivity; radon-220; radon-222; statistics

1. Introduction

Adobe, made from clay, sand (or simply from soil), water and organic material, is a typical building material of cultural heritage in Hungary's rural areas. After mixing the raw materials and pressing them into shapes, the adobe blocks are dried out in sunshine without any burning procedure. Therefore, adobe dwellings are considered to have low energy costs and a small environmental impact. For this reason the tradition of building adobe houses is being revived and, considering all potential health hazards, such as indoor radon and thoron activity concentrations, is becoming more important.

*Corresponding author. Emails: szabo.zsuzsanna@energia.mta.hu; zsszabo86@gmail.com

Radon (^{222}Rn, Rn) and thoron (^{220}Rn, Tn) isotopes are responsible for approximately half of the average annual effective dose to humans, and their elevated concentrations can increase the risk of lung cancer. In the past, exposure to thoron was often ignored due to its short half-life (55.6 s), but it is now known that thoron and its daughters can significantly contribute to the radiation dose in some environments [1]. In these environments, several studies [2–6] show elevated thoron concentrations in dwellings built of soil and mud as are Hungarian adobe dwellings. From a methodological point of view, Tokonami [7] has also pointed out the importance of parallel thoron measurements for the correct radon inhalation dose estimation and for risk evaluation purposes.

Although the naturally occurring radioactive material content of adobe is close to the average background levels in Hungarian soils based on our parallel study [8], its porous and permeable structure can lead to increased radon and thoron exhalation and indoor accumulation. The indoor radon and thoron activity concentrations are influenced by weather conditions, the properties of the source materials (soil and building material) and the wall coatings (lime, paint or wallpaper). For example, the intensity of heating in the room during the winter period (pressure difference driven airflows) [9], the natural air exchange processes (cracks around the windows, doors and roof) or the habits of residents (opening windows or doors to ventilate the house) can all change the measurable activity concentration of either radionuclide. However, while radon is approximately homogeneously distributed in the indoor air, thoron concentration drops drastically towards the centre of the room [10], and only its decay products are evenly distributed. Both the gaseous radon and thoron and their solid decay products can be inhaled, and they can affect human health.

The need to compare the annual levels with international action levels is obvious. The World Health Organization (WHO) [11] proposes an annual indoor radon activity concentration action level of 100 Bq m^{-3} and declares that the chosen national action level of any country should not exceed 300 Bq m^{-3} [12]. No such action levels exist for thoron gas but for thoron decay products. Annual thoron levels at a given distance from the walls can instead be compared with several published national surveys.

The objective of this paper is to present the seasonal and spatial variations of radon and thoron activity concentrations and annual averages in adobe dwellings in the rural area of the Great Hungarian Plain. The paper considers the health impact on the residents, and our findings help to forecast expected levels of radon and thoron in adobe dwellings via analysing the affecting environmental factors.

2. Studied area

The location of the studied area of Békés County, SE Hungary in the Carpathian–Pannonian region is shown on a geographical map in Figure 1. Since the raw material of adobe is collected locally, the target area selection took into account the possible geological and geographical differences to represent the building material variability. The sampling is considered to be representative for adobe dwellings of the Great Hungarian Plain. Seven settlements within a 2000 km^2 study area were chosen for the indoor radon and thoron activity concentration measurements. The selected settlements are Gyula, with more than 30,000 inhabitants, Gyomaendrőd, Sarkad, Vésztő and Kondoros with a population between 5000 and 15,000 and Sarkadkeresztúr and Újiráz with fewer than 2000 inhabitants. Subsequently, these settlements are discussed in alphabetical order and indicated by capitals: Gyomaendrőd by A, Gyula by B, Kondoros by C, Sarkad and Sarkadkeresztúr together by D, Újiráz by E and Vésztő by F (Figure 1). Sarkad and Sarkadkeresztúr are in close proximity to each other and considered together.

Most of the studied adobe houses were built between 1930 and 1960. At each settlement the clayey soil for adobe was typically collected from the same location at the edge of the populated

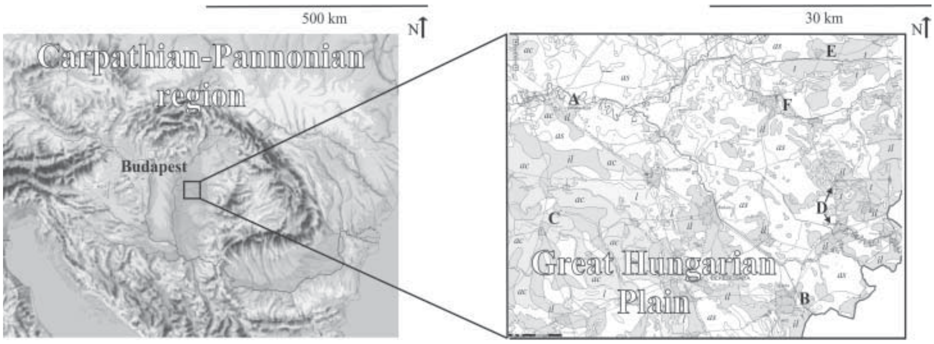

Figure 1. The location of the studied part of the Great Hungarian Plain, SE Hungary in the Carpathian–Pannonian region and the location of the studied settlements indicated by capitals on the geological map [14]. The different grey tones with the following notations indicate different Quaternary geological formations: ac – alluvial clay, as – alluvial sediments, il – infusion loess, l – loess (mixed with sand) and t – turf.

area. The major raw material of local adobe building materials is the prevailing fluvial sediments of Körös and Berettyó rivers, tributaries of Tisza river. These sediments are mainly Quaternary alluvial clay, infusion loess and rarely turf and aleurolite formations [13,14] originating from the Carpathians. Based on the most probable geological source of adobe (Figure 1), the selected settlements can be divided into three groups: clay (ac and as, Figure 1) – A, C and F settlements, loess (il and l, Figure 1) – B and D settlements and turf (t, Figure 1) – E settlement.

This rural, agricultural area lies at 80–180 m above sea level. It has a European continental climate with an annual average temperature of 10–11 °C, with warm summers (20 °C), relatively cold winters (0 °C) and mild springs and autumns (10 °C). The annual average precipitation is 500–550 mm. The precipitation falls mostly during the summer months (especially June) and the least in the winter season (especially February) based on the Hungarian Meteorological Service (OMSZ) database [15]. The landscape is dominated by grassland shaped by extensive agriculture.

3. Methods

3.1. *Sampling*

Indoor radon and thoron activity concentration measurements were performed in 53 adobe dwellings in the above-mentioned settlements (Figure 1), 5–11 studied buildings in each. In detail, 11 dwellings were studied at A, 10 at B, 5 at C, 11 at D, 5 at E and 11 at F settlements.

For the success of the project, the help of local people to find appropriate houses and owners volunteering to participate in the measurement campaign was indispensable. The selection criterion was that the room had to have at least one adobe wall, and preferably be in daily use or at least offer the possibility to sleep, live or work in. The type of coating and the level of heating were easy to report, whereas passive or active air exchange could not be easily measured. It was not possible to measure the radon and thoron activity concentrations at all locations during the whole measurement period. In some cases, the owners refused having the detectors or some loss or damage occurred.

Raduet type etched track detector pairs produced by Radosys Ltd., Budapest, Hungary were used for the radon and thoron measurements. They were placed at a 10 ± 1.5 cm distance from the adobe walls. This distance was chosen to match earlier studies [16–18] so that a comparison of the results can be made. However, some recent studies, e.g. by Stojanovska et al. [19], placed the detectors at a distance of at least 50 cm. Note that these measurements are always expected to show lower thoron activity concentrations than at 10 cm distance because of the inhomogeneous distribution of thoron in the room [10]. For the same reason, special care was taken to avoid double or multiple-sided effects at wall edges and corners, as well as any influence of electronic devices [20]. Detectors were placed at a height between 60 and 240 cm from the ground, while adjusting to the conditions provided by the residents.

The measurements were carried out in three-month periods for one year, from December 2010 to November 2011, in order to represent the four seasons typical of the climate [15]: winter from December 2010 to February 2011, spring from March to May 2011, summer from June to August 2011 and autumn from September to November 2011.

3.2. *Detector reading*

For the analysis of detectors, the chemical etching of inner plastic films was performed in the laboratory of the National Research Institute for Radiobiology and Radiohygiene (OSSKI), Budapest, Hungary with 6.25 M NaOH solution at 90 °C for 5 h. The counting of α-tracks was carried out by Radosys automatic microscopes in the lab of OSSKI and partly in the lab of the manufacturer, and each plastic film was counted in at least three sequential runs.

Radon and thoron activity concentrations for the three-month periods were calculated from the track density of detector pairs. The background track density and the calibration factors were provided by the manufacturer. In the uncertainty calculations, we took into account the uncertainty of both the track densities and the calibration factors. For more details about applied calculation procedures (activity concentrations, uncertainty, decision threshold and detection limit) and quality assurance see the publication of Stojanovska et al. [19] and the manuals of the manufacturer [21,22].

Annual averages were only determined when measurement data were available in all seasons in the given building.

3.3. *Statistical analysis*

Several distributions are analysed in this study. Two main groups are considered: annual average activity concentrations ($c_{av.}$) and seasonal measurement results (c). The annual statistics are analysed as $Rn_i(c_{av.})$ and $Tn_i(c_{av.})$, where $i =$ all, A, B, C, D, E and F settlements. The direct three-month period measurement results were considered as $Rn_j(c)$ and $Tn_j(c)$, where $j =$ winter, spring, summer and autumn.

In the statistical analysis of the distributions, robust statistics were used and Tukey's resistant five-letter summary statistics [23] containing the minimum, lower quartile, median, upper quartile and the maximum were calculated and visualised by box-whisker plots. Median and upper quartiles played important roles in the data evaluation of this study. Average and standard deviation values are also presented, as well as the standardised skewness and standardised kurtosis. Relative variabilities were analysed by the robust median absolute deviation/median (MAD/median) measure because this is not sensitive to outlier values.

The Mann–Whitney (Wilcoxon) non-parametric homogeneity test (MW test), based on the comparison of medians, was applied to test homogeneity and observed significant differences between radon and thoron activity concentrations and among seasons. Significant deviation from lognormal distribution of radon data was described by Bossew [24] and Tóth et al. [25] when non-homogeneous regions with uniform geology, building style and living habits were sampled. Statistical distributions, hence, were tested for lognormality by Shapiro–Wilk test (SW test). The data were also tested for normality as it is a condition of most parametric tests. All statistical tests were carried out at the 95% confidence level (α). The best-fitting distribution was also found in some cases by the log likelihood statistics (LL test). The standardised skewness and standardised kurtosis values were also taken into account in the distribution analysis, the values of these distribution-shaped statistics outside the range of −2 to +2 indicate significant departures from normality (or lognormality). Statistically significant non-zero relationships between measured parameters were studied with the Pearson's linear correlation coefficient at the 95% confidence level.

For data processing, this study used the Microsoft Excel, Statgraphics Centurion, Golden Software Surfer and Microcal Origin software packages.

4. Results

In the 53 adobe dwellings of the studied settlements, 190 radon and 189 thoron measurements are available for the 4 seasons in the measurement period from December 2010 to November 2011. One thoron data are missing, beside radon, due to the damage of one detector in the detector pair. For radon and thoron activity concentration results, the overall uncertainties are determined to be 20 and 30%. The annual average radon and thoron activity concentrations were determined in 43 and 42 adobe dwellings, respectively, only where measurement data were available in all seasons.

4.1. *Annual activity concentrations and their spatial variation*

Figure 2 summarises the statistics of $Rn_{all}(c_{av.})$ and $Tn_{all}(c_{av.})$. It is seen that the annual radon activity concentration has a median of 188 Bq m^{-3}, and it is 232 Bq m^{-3} for thoron. Based on the MW test, there is a statistically significant difference between these medians. The MAD/median values are similar, 0.29 and 0.35 for radon and thoron, respectively (Figure 2).

The SW tests are consistent with the standardised skewness and the standardised kurtosis values (Figure 2). They reject the normality but do not reject the lognormality for annual average radon data. However, for thoron the same tests cannot reject the idea of normal or lognormal distributions. Results of LL statistics show that for radon, the best-fitting distribution is the lognormal distribution and for thoron, normal distribution fits better than lognormal. In the further analysis of the results we accept that annual radon data come from a lognormal and annual thoron data come from a normal distribution.

Spatial variation of annually averaged radon and thoron activity concentrations for each studied settlement ($Rn_A(c_{av.})$, $Tn_A(c_{av.})$, $Rn_B(c_{av.})$, $Tn_B(c_{av.})$, $Rn_C(c_{av.})$, $Tn_C(c_{av.})$, $Rn_D(c_{av.})$, $Tn_D(c_{av.})$, $Rn_E(c_{av.})$, $Tn_E(c_{av.})$, $Rn_F(c_{av.})$ and $Tn_F(c_{av.})$) are presented by box-whisker plots (Figure 3). Radon has annual average medians ranging from 102 (E settlement) to 212 Bq m^{-3} (C settlement) at the studied settlements. At the same time, annual thoron activity concentration medians range from 172 (D) to 320 Bq m^{-3} (E, Figure 3).

Always one dwelling at each settlement showed an elevated (i.e. higher than annual upper quartile) radon activity concentration, except settlement E with no anomalous concentrations and F with four. Thoron data are not elevated at the same places except for two dwellings at F settlement. Beyond that, two dwellings at A and one at B, D and E settlements show elevated thoron activity concentrations.

4.2. *Seasonal variation*

Seasonal variation of radon and thoron activity concentration in the studied adobe dwellings are shown by box-whisker plots in Figure 4. Table 1 summarises the robust statistics for the data in the four seasons ($Rn_{winter}(c)$, $Tn_{winter}(c)$, $Rn_{spring}(c)$, $Tn_{spring}(c)$, $Rn_{summer}(c)$, $Tn_{summer}(c)$, $Rn_{autumn}(c)$ and $Tn_{autumn}(c)$). Some relevant measures and the Pearson's linear correlation coefficients are analysed (Table 2).

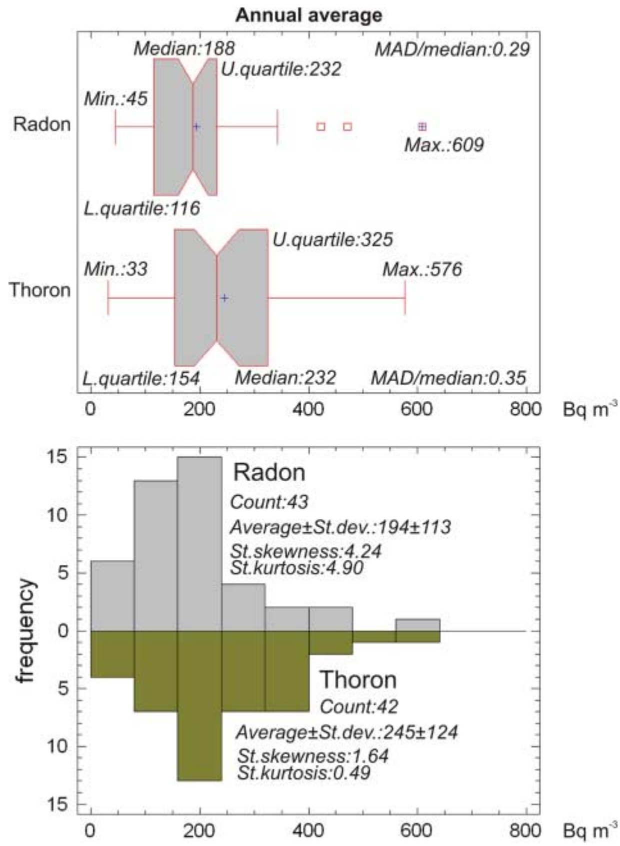

Figure 2. Box-whisker plots and frequency histograms of all determined annual average radon and thoron activity concentrations ($Rn_{all}(c_{av.})$ and $Tn_{all}(c_{av.})$) showing the count (sample number by the piece), minimum (Bq m^{-3}), lower quartile (Bq m^{-3}), median (Bq m^{-3}), upper quartile (Bq m^{-3}), maximum (Bq m^{-3}), average (Bq m^{-3}), standard deviation (Bq m^{-3}), standard skewness, standard kurtosis and MAD/median of the statistics.

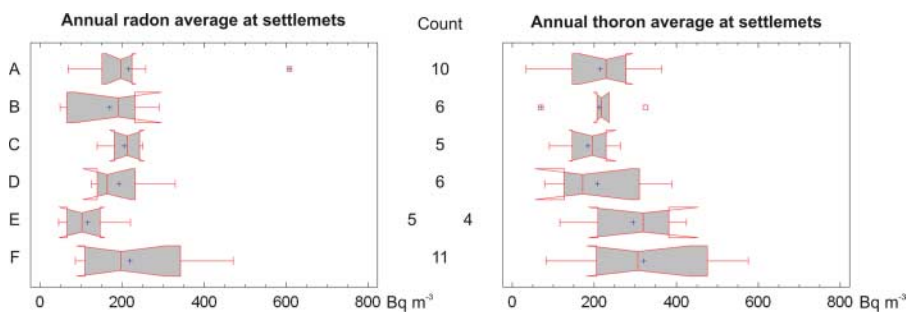

Figure 3. Box-whisker plots of determined annual average radon and thoron activity concentrations at the studied settlements indicated by capitals ($Rn_A(c_{av.})$, $Tn_A(c_{av.})$, $Rn_B(c_{av.})$, $Tn_B(c_{av.})$, $Rn_C(c_{av.})$, $Tn_C(c_{av.})$, $Rn_D(c_{av.})$, $Tn_D(c_{av.})$, $Rn_E(c_{av.})$, $Tn_E(c_{av.})$, $Rn_F(c_{av.})$ and $Tn_F(c_{av.})$).

4.2.1. *Seasonal medians*

Figure 4 and Table 1 show that radon median displays a close to typical seasonal variation with high values in winter and autumn, smaller values in spring and low values in summer. Winter and autumn medians are three to four times higher than that of summer. The MW tests show that there

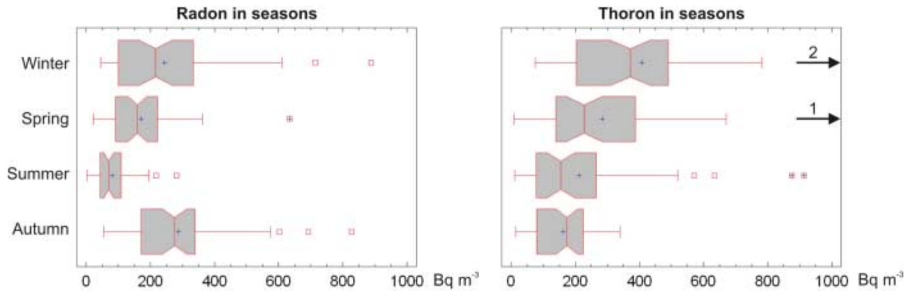

Figure 4. Box-whisker plots of measured radon and thoron activity concentrations in the four seasons ($Rn_{winter}(c)$, $Tn_{winter}(c)$, $Rn_{spring}(c)$, $Tn_{spring}(c)$, $Rn_{summer}(c)$, $Tn_{summer}(c)$, $Rn_{autumn}(c)$ and $Tn_{autumn}(c)$). In case of thoron, three outlier values are not shown on the figure for better visibility and comparable scale to radon. More details about statistics are in Table 1.

Table 1. Count (sample number), minimum (Bq m^{-3}), lower quartile (Bq m^{-3}), median (Bq m^{-3}), upper quartile (Bq m^{-3}), maximum (Bq m^{-3}), average (Bq m^{-3}), standard deviation (Bq m^{-3}), standard skewness, standard kurtosis and MAD/median for radon and thoron activity concentration measurement data in the four seasons ($Rn_{winter}(c)$, $Tn_{winter}(c)$, $Rn_{spring}(c)$, $Tn_{spring}(c)$, $Rn_{summer}(c)$, $Tn_{summer}(c)$, $Rn_{autumn}(c)$ and $Tn_{autumn}(c)$).

		Count	Min.	L. quartile	Median	U. quartile	Max.	Average	Std dev.	Std skewness	Std kurtosis	MAD/ median
Radon	Winter	50	45	100	217	334	888	244	179	4.45	4.20	0.54
	Spring	48	22	92	159	222	634	171	108	4.91	8.30	0.40
	Summer	46	3	43	70	109	281	81	57	4.02	3.63	0.45
	Autumn	46	55	171	276	339	827	287	160	3.57	3.18	0.29
Thoron	Winter	50	75	203	372	491	2306	407	344	11.00	27.92	0.36
	Spring	47	8	139	228	388	1264	284	218	5.97	10.85	0.54
	Summer	46	11	78	156	266	914	212	210	5.15	4.92	0.65
	Autumn	46	14	79	174	225	340	163	86	−0.10	−1.21	0.35

Table 2. Correlation coefficients of radon and thoron activity concentrations among the four seasons ($Rn_{winter}(c)$, $Tn_{winter}(c)$, $Rn_{spring}(c)$, $Tn_{spring}(c)$, $Rn_{summer}(c)$, $Tn_{summer}(c)$, $Rn_{autumn}(c)$, $Tn_{autumn}(c)$).

r	Winter–spring	Winter–summer	Winter–autumn	Spring–summer	Spring–autumn	Summer–autumn
Radon	.89	.32	.90	.50	.90	.50
Thoron	.77	.18	.44	.54	.55	.36

are statistically significant differences among the seasonal median radon activity concentrations, except the winter and autumn pair. At the same time thoron median is steadily decreasing during the measurement period (Figure 4, Table 1). Winter median is more than twice as high as that of summer and autumn. MW tests show that there are statistically significant differences between the thoron activity concentration medians between seasons except the summer–autumn pair.

Medians of thoron activity concentration data tend to exceed those of radon in winter, spring and summer. However, in autumn, radon exceeds almost twice the thoron median (Figure 4, Table 1). Based on MW tests, there are statistically significant differences between the radon and thoron activity concentration medians in all seasons.

4.2.2. *Seasonal statistical variabilities*

The overall variability of radon activity concentration data is represented by the MAD/median values (Table 1) and shown to be similar in winter, spring and summer, but the values of the measure drop in autumn. Similarly, the variability of thoron activity concentration data increases from winter to summer and then drops again in autumn.

These results suggest that radon is most variable and thus least predictable in winter, and thoron varies the most in summer. For both isotopes, autumn seems to be the most stable and predictable season.

4.2.3. *Seasonal statistical distributions*

The SW tests verify what standardised skewness and the standardised kurtosis values (Table 1) suggest, namely that only autumn thoron activity concentrations come from a normal distribution and that winter, spring and autumn radon and winter thoron results are lognormally distributed. However, the SW tests reject both normality and lognormality in the cases of summer radon and spring and summer thoron measurements.

4.2.4. *Correlation analysis*

Pearson's linear correlation coefficients (r) among different seasons were studied for both radon and thoron (Table 2). Radon shows $r \approx 0.9$ among winter, spring and autumn seasons in different dwellings but summer has lower r values (Table 2). However, all of these indicate statistically significant non-zero correlations among seasons. Thoron generally shows lower correlation coefficients (Table 2). These also indicate statistically significant non-zero correlations among seasons, except winter and summer.

The Pearson's linear correlation coefficients between the two studied isotopes are low and all of them statistically insignificant ($r = 0.12$, 0.00, -0.04 and 0.23 for winter, spring, summer and autumn seasons, respectively).

5. Discussion

5.1. *Health impact*

5.1.1. *International comparison of annual activity concentrations*

Table 3 summarises an international comparison of our results and shows the types of building materials and the measurement distances from the walls. The geometric means of our results are also presented for comparison: 166 and 211 Bq m^{-3}, respectively, for radon and thoron.

Only three studies show high radon activity concentrations comparable with our results. These were performed at Kővágószőlős, Hungary [26,27], which is known by its former uranium mine and in Kosovo and Metohija, Serbia [28]. Thoron generally has low activity concentrations ($<$50 Bq m^{-3}) [16,17,19,26–31]. However, elevated thoron activity concentrations were detected in Indian mud dwellings [29] and rural dwellings of Balkans [32]. Only in Chinese cave dwellings [5,18] were thoron levels measured to be as high as in Hungarian adobe dwellings.

It is seen that the annual radon and thoron levels measured in adobe dwellings in the studied part of the Great Hungarian Plain show elevated levels compared with results in other studies (Table 3).

5.1.2. *Proportion of dwellings above action levels*

The annual average radon activity concentration follows a lognormal distribution, whereas we accepted that the annual thoron data are normally distributed. In Figure 5 we chose the axes

Table 3. Average (av.), minimum–maximum (min–max.) and geometric mean (GM) of determined annual average radon and thoron activity concentrations (Bq m^{-3}) in this study ($Rn_{all}(c_{av.})$ and $Tn_{all}(c_{av.})$) in comparison with international action levels.

Reference	Country	Building material	Distance	Measure	Radon	Thoron
This study	Hungary – Great Hungarian Palin	Adobe	10 cm	Av. Min-max. GM	194 (43) 45–609 (43) 166 (43)	245 (42) 33–576 (42) 211 (42)
[26]	Hungary – Kővágószőlős	Various – summer	15–20 cm	Av.	154 (72)	98 (72)
[27]				GM	107 (80)	53 (74)
[18]	China	Various	10 cm	Av.	29 (100)	184 (100)
[5]	China	Cave dwelling (adobe)	5–30 cm	GM	81 (102)	261 (102)
[29]	India – Andhra Pradesh	Mud (adobe)	–	–	–	143 (8)
		Stone	–	–	–	34 (60)
		Mosaic	–	–	–	31 (10)
		Concrete	–	–	–	33 (11)
[17]	India – Assam	Various	10 cm	Min-max.	40–215 (46)	13–38 (46)
[30]	India – Chhattisgarh	Various	20 cm	GM	26 (210)	18 (210)
[16]	India – Kerala	Various	10 cm	GM	23 (200)	24 (200)
[28]	Serbia – Kosovo and Metohija	Various	–	GM	224 (63)	43 (63)
[19,31]	Macedonia	Various	50 cm	GM	82 (437)	28 (53–300)
[32]	Serbia and Bosnia-Hercegovina	Rural dwellings	20 cm	GM	82 (183)	109 (183)

Note: The sample numbers were indicated in brackets following the activity concentration values.

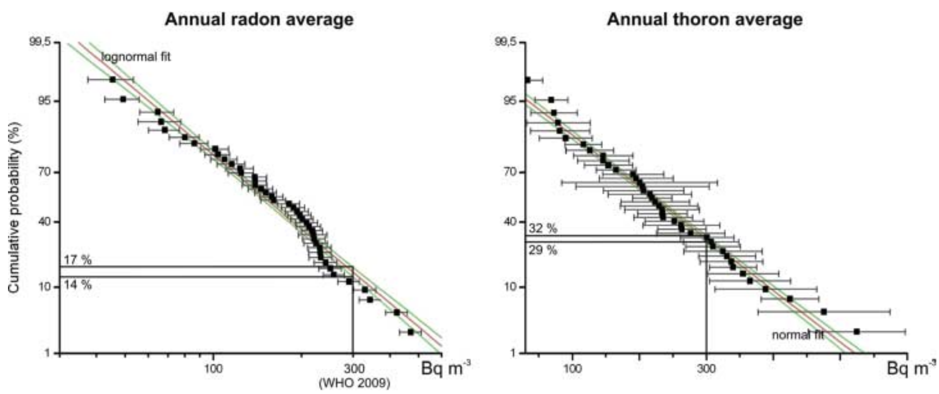

Figure 5. Cumulative probabilities of annual average radon and thoron activity concentrations ($Rn_{all}(c_{av.})$ and $Tn_{all}(c_{av.})$). The activity concentrations are presented on a log scale for radon, and on a linear scale for thoron. The lines represent the fitted lognormal (radon) and normal (thoron) distribution functions with their 95% confidence intervals.

and scales in such a way that the plotted annual activity concentration values vs. the cumulative probability describe linear functions. For this purpose, the activity concentrations are presented on a log scale for radon, but on a linear scale for thoron.

For radon, we consider 300 Bq m^{-3} as the highest action level recommended by WHO [11]. Choosing an action level for thoron is not as obvious as for radon because it exists only for thoron decay product concentration. In this study, for a comparison, we determine the proportion of dwellings above the same activity concentration as for radon, 300 Bq m^{-3}. However, it is emphasised that the same thoron effective dose must come from a much higher activity concentration, probably around 1000 Bq m^{-3}.

Based on Figure 5 and the fitted linear functions representing the lognormal (radon) and normal (thoron) distributions, the following can be stated:

(1) 14–17% of the adobe dwellings in the Great Hungarian Plain have higher radon activity concentration than the reference level of 300 Bq m^{-3} by 95% probability. In this study, 12% of the dwellings were above this action level (Figure 5, left side).
(2) 29–32% of the adobe dwellings in the Great Hungarian Plain have higher thoron activity concentration than 300 Bq m^{-3} by 95% probability. The empirical proportion for thoron in this study is determined to be 33% (Figure 5, right side).

5.1.3. *Inhalation dose estimation and evaluation*

Based on the determined annual levels, residential inhalation dose estimation is carried out for radon isotopes using the average radon equilibrium factor of 0.4, the dose conversion factor of 9 nSv (Bq h m^{-3})$^{-1}$ [1] and the indoor occupancy time of 7012.8 h y^{-1} [33]. The calculated radon inhalation dose median of 4.74 mSv y^{-1} and average of 4.90 mSv y^{-1} (Figure 6) are both significantly higher than the world average value ($\sim$1.15 mSv y^{-1}) [1], which is equal to the minimum in our statistics. In three dwellings, i.e. 7% of the studied dwellings with estimated values, the inhalation dose exceeds 10 mSv y^{-1}, which still falls into the low dose range. However,

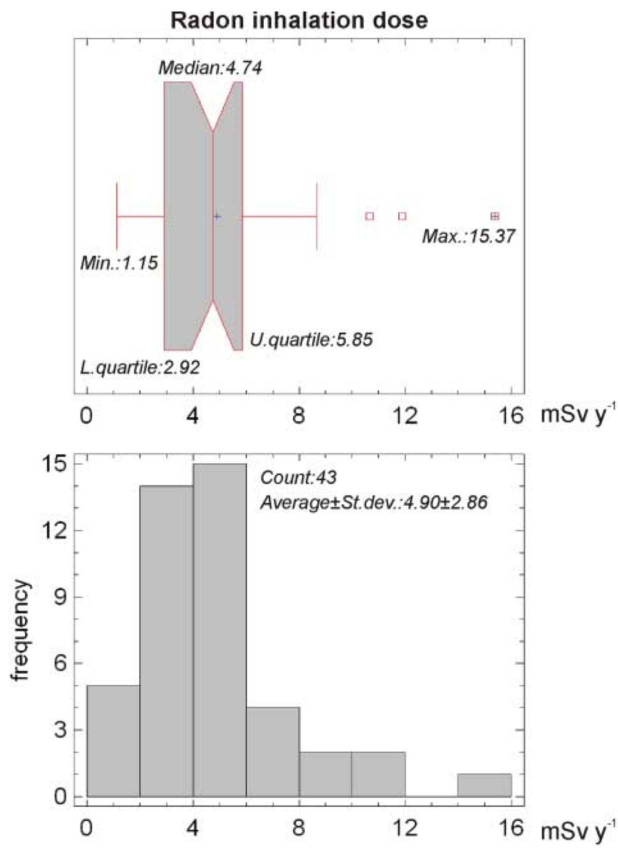

Figure 6. Box-whisker plot and frequency histogram of estimated radon inhalation doses showing the count (sample number by the piece), minimum (mSv y^{-1}), lower quartile (mSv y^{-1}), median (mSv y^{-1}), upper quartile (mSv y^{-1}), maximum (mSv y^{-1}), average (mSv y^{-1}) and standard deviation (mSv y^{-1}) of the statistics.

recent models [34] describe a much higher local tissue dose resulting from the determined effective dose because of its inhomogeneous distribution in the lungs. This means that 10 mSv y^{-1} from radon cannot be considered identical to 10 mSv y^{-1} from γ-radiation because it can lead to a more significant health impact resulting in the elevation of lung cancer risk.

Similar reliable dose estimation is not possible to be carried out for the thoron isotope (for more information about behaviour of thoron and its daughters see [35]). However, we made a crude estimation based on the thoron activity concentration data and international experiences. For this, the equilibrium factor of 0.03, the dose conversion factor of 40 nSv $(\mathrm{Bq\,h\,m^{-3}})^{-1}$ [1] and the indoor occupancy time of 7012.8 h y^{-1} [33] were used. The equilibrium factor was chosen based on a few test measurements of thoron decay products in the spring period (see Acknowledgements) and Harley et al. [36]. This calculation should be considered carefully but shows that thoron gives about 30% of the inhalation dose in the studied dwellings.

5.2. *Environmental factors affecting radon and thoron variations*

5.2.1. *Geological environment*

Minda et al. [37] pointed out that indoor radon shows dependence on geological formations in Hungary. Moreover, the radon and thoron mother nuclide content of adobe is a consequence of the soil concentrations at the settlement as this building material is always made locally. Therefore, in this section we discuss the dependence of annual radon and thoron activity concentrations on the local geological environment (Figure 1, [13,14]).

Summarising the results, E settlement shows lower radon levels than the others while having the highest median thoron activity concentration. This settlement lies next to a Quaternary turf formation. Meanwhile, adobe building material at C settlement showing the highest radon and the lowest thoron median (Figure 3) most probably is made of Quaternary alluvial clay. The statistically insignificant Pearson's linear correlation coefficients between radon and thoron isotopes (see Results, $-0.04 < r < 0.23$) are consistent with the observation that radon and thoron data are generally not elevated in the same dwelling. All these results refer to a difference between the amount of radon and thoron in different geological backgrounds. Note that radon also originates from the soil below the dwellings.

Based on the geological environment of settlements (see Studied area), new distributions are defined as $\mathrm{Rn}_k(\mathrm{c_{av.}})$ and $\mathrm{Tn}_k(\mathrm{c_{av.}})$, where $k =$ clay, loess and turf. Figure 7 summarises the statistics for these distributions, i.e. for clay, loess and turf environments ($\mathrm{Rn_{clay}}(\mathrm{c_{av.}})$, $\mathrm{Tn_{clay}}(\mathrm{c_{av.}})$, $\mathrm{Rn_{loess}}(\mathrm{c_{av.}})$, $\mathrm{Tn_{loess}}(\mathrm{c_{av.}})$, $\mathrm{Rn_{turf}}(\mathrm{c_{av.}})$ and $\mathrm{Tn_{turf}}(\mathrm{c_{av.}})$). The radon activity concentration median values are 199, 175 and 102 Bq m^{-3}, whereas the thoron medians are 234, 211 and 320 Bq m^{-3} for clay, loess and turf, respectively. All distributions strongly overlap and the sample numbers are

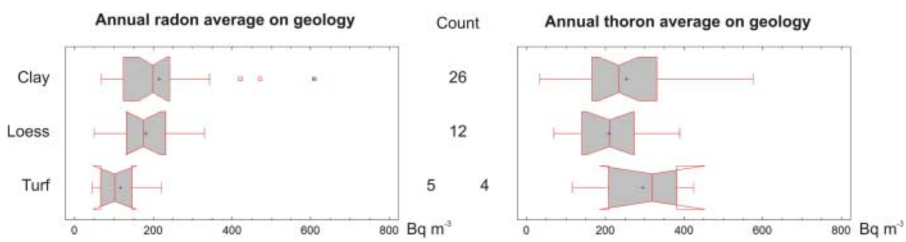

Figure 7. Box-whisker plots of determined annual average radon and thoron activity concentrations grouped based on different geological environments: clay, loess and turf ($\mathrm{Rn_{clay}}(\mathrm{c_{av.}})$, $\mathrm{Tn_{clay}}(\mathrm{c_{av.}})$, $\mathrm{Rn_{loess}}(\mathrm{c_{av.}})$, $\mathrm{Tn_{loess}}(\mathrm{c_{av.}})$, $\mathrm{Rn_{turf}}(\mathrm{c_{av.}})$ and $\mathrm{Tn_{turf}}(\mathrm{c_{av.}})$).

low (Figure 7), but MW tests still show that radon annual average median on clay (199 Bq m^{-3}) is statistically significantly higher than on turf (102 Bq m^{-3}).

The highest values for both radon and thoron are detected on clay formations (Figure 7). On clay the highest, on loess medium and on turf the lowest annual radon activity concentrations were detected, whereas in case of thoron the highest median was on turf although the maximum values were again on clay. It is seen that the local geology differently affects the radon and thoron levels resulting in different spatial variations. However, for both isotopes the clay formations showed to be the highest risk localities.

5.2.2. *Weather conditions*

Based on the results, MW tests satisfy the criteria of significant difference between seasons more frequently than between geological environments. Therefore, the weather conditions seem to be the more relevant radon and thoron activity concentration affecting factors than the local geology. Below, the results are evaluated considering the environmental parameters changing through the seasons of the measurement period.

Radon median seasonal pattern (Figure 4, Table 1) basically follows the typical seasonal temperature changes of the studied area [15]. It is explained by the outdoor/indoor temperature gradient [38] and the resulted pressure gradient since radon is always leaving the adobe walls and the soil towards the warmer, i.e. lower pressure side [9]. However, in case of the thoron isotope, the median values decrease during the whole measurement period (Figure 4, Table 1), i.e. they do not increase again in autumn when outdoor temperature drops and indoor heating usually starts. Variances (MAD/median) of both radon and thoron levels are much lower in autumn (Table 1) compared with other seasons. These results all indicate the strong influence of another environmental parameter, which has the maximum effect at the end of the measurement period.

We experienced during the field campaigns that the studied area received an extremely high amount of precipitation in 2010, but an extremely low amount in 2011 (Figure 8; [15]). Since the moisture content of building materials influences their radon and also their thoron emanation and exhalation [39,40], it indirectly influences the indoor activity concentration values. Adobe

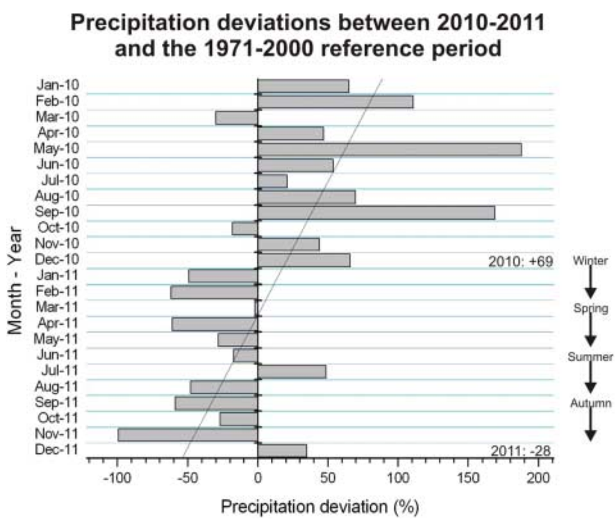

Figure 8. Deviation of monthly averages of measured precipitation (%) in 2010 and 2011 from the average of 1971–2000 period, Hungary [15]. The radon and thoron activity concentration measurements started in the winter of 2010, earlier months before the start of measurements are able to influence the results.

building material, especially, tends to absorb water from the ground and release the moisture during dry spells. Therefore, the strongly decreasing amount of precipitation (Figure 8; [15]) and consequently decreasing moisture content of adobe is considered to possibly cause the decreased thoron results in autumn. This is possible if the decrease is all happening below the optimal value of moisture content in the exhalation process (determined to be 8% in [39]). Other studies seem to be consistent with our thoron results [41–43]. If we accept the reasoning above, it has to be assumed that indoor radon data in this study are not as strongly affected by the moisture content of the building material because of the isotope's much longer half-life and its additional source: the soil below the dwellings.

The statistical distributions of measured radon and thoron activity concentrations also show seasonal variations. In case of radon, the results generally show a lognormal distribution but not in the hot summer period. Since it is known [24,25] that deviation from the lognormal distribution is connected to sampling heterogeneity, it is assumed that at least two types of dwellings are formed by summer, i.e. well and poorly ventilated ones. Consequently, the level of ventilation via opening the windows is considered to have a significantly reducing but unpredictable effect on the radon activity concentration. The low Pearson's linear correlation coefficients for only the summer season (Table 2) also support this generally accepted process. The effect of ventilation on the thoron activity concentration is more complicated than on that of radon.

The effects of three weather conditions were connected to our data. The seasonal variations of radon and thoron activity concentrations seem to be influenced by temperature, precipitation and ventilation weather conditions. Radon follows the average outdoor temperature changes and affected by the increased ventilation in summer; however, thoron moved together with the amount of precipitation, i.e. the moisture content of adobe building material.

6. Conclusions

The annual radon and thoron activity concentrations in adobe dwellings in the Great Hungarian Plain are elevated compared with results in other national and international surveys. The present study shows that 14–17 and 29–32% of the dwellings have annual activity concentrations higher than 300 Bq m^{-3} for radon and thoron, respectively. The estimated radon inhalation dose of residents is significantly higher than the world average value, and in 7% of the dwellings of this study it exceeds 10 mSv y^{-1}. This study suggests that thoron can also be a significant contributor to the inhalation dose with about 30% in the total inhalation dose, additionally. Therefore, Hungarian and also East-Central European adobe dwellings should be considered for the thoron problem beside the generally recognised Chinese and Indian cave and the so-called mud dwellings. Further surveys are needed together with informing residents about easily available radon and in some cases optional thoron mitigation methods.

The spatial variation of annual average radon activity concentrations suggests that on clay the highest, on loess medium and on turf the lowest values are expected in adobe dwellings of the Great Hungarian Plain. In case of the thoron isotope, no significant differences were observed based on the type of sedimentary geological environment, although the maximum values were measured in adobe dwellings on clay and in contrast to radon, the highest median was determined on turf. However, the weather conditions such as the temperature, precipitation and ventilation seem to be more relevant than the local geology for both radon and thoron resulted in characteristic but different seasonal variations of activity concentrations. Radon follows the average temperature changes and is affected by the increased ventilation in summer. However, in this study thoron moved together with the amount of precipitation, i.e. the moisture content of adobe building material through the one-year measurement period.

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