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National survey of indoor thoron concentration in FYR of Macedonia (continental Europe – Balkan region)

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HIGHLIGHTS

- The indoor ^{220}Rn concentrations show seasonal and regional variability.
- The amplitude of seasonal variability is smaller for ^{220}Rn, than for ^{222}Rn.
- Indoor ^{220}Rn map of FYR of Macedonia.
- The influence of house characteristics were analysed and discussed.
- Recommendations for future investigation were made.

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ABSTRACT

As a part of the national survey of natural radioactivity in The Former Yugoslav Republic of Macedonia, indoor thoron gas concentration was measured in 300 dwellings during one year, from December 2008 till December 2009 using passive discriminative radon–thoron detectors. Detectors were deployed at a distance of >50 cm from walls in order to be less sensitive to distance from walls. Altogether 532 measurements were performed: 53 in winter, 57 in spring, 122 in summer and 300 in autumn. The frequency distribution is well described by a log-normal function. The geometric means of indoor thoron concentration (with geometric standard deviations in brackets) in winter, spring, summer and autumn were obtained to be: 39 Bq m^{-3} (3.4), 32 Bq m^{-3} (2.8), 18 Bq m^{-3} (2.8), 31 Bq m^{-3} (2.9), respectively. Seasonal variations of thoron appear lower than those of radon. The seasonal corrected annual mean concentration ranges between 3 and 272 Bq m^{-3} with a geometric mean of 28 Bq m^{-3} (2.12). A detailed statistical analysis of the geogenic and building factors which influence the indoor thoron concentration is also reported. This survey represents the first national survey on indoor thoron in continental Europe.

1. Introduction

Radon (^{222}Rn) and its progenies are known to pose a major health hazard in indoor environments, as it is believed to be the second important cause of lung cancer after smoking. Epidemiological studies carried out in China, Europe, and North America gave robust evidence of increased lung cancer risk with increasing radon exposure in dwellings (Lubin et al., 2004; Darby et al., 2005; Krewski et al., 2005), and that adverse effects of radon occur also at relatively low concentration levels, i.e. up to 200 Bq m^{-3} (Darby et al., 2005). Moreover, recent studies on miners exposed at relatively low levels of radon showed that the risk is about twice the risk estimated on previous epidemiological studies on miners exposed at high radon concentration levels (Tomáček et al., 2008). As a consequence, recommendations and regulations of international organizations (reviewed by Bochicchio, 2011) have been

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revised for controlling radon exposure towards lower levels (e.g. WHO, 2009; EC, 2011; ICRP, 2011). WHO recommended for dwellings a reference level of 100 Bq m^{-3}, or, if such level was not feasible, a level not greater than 300 Bq m^{-3} (WHO, 2009). In its proposal for a new directive on basic safety standards, the European Commission reports that Member States shall choose a reference level not greater than 300 Bq m^{-3} to be implemented as national regulation (EC, 2011). ICRP has recently published a draft of a new recommendation on radon in dwellings and workplaces where the same reference levels for dwellings (not greater than 300 Bq m^{-3}) is recommended for normal workplaces, as a first step of a graded approach for workplaces (ICRP, 2011). The discussion about radon as health hazard has motivated many countries to carry out radon surveys for several decades. In some countries extensive datasets of indoor radon measurements have been collected and used for producing radon maps, which can be quite useful for planning radon control activities, especially if measurements are representative of population exposure (WHO, 2009). For some years a harmonized European radon map of measurements carried out at ground floor is under way to being created; so far (early 2012) 24 countries participate in the project. Preliminary results are reported e.g. in Tollefsen et al. (2011).

No comparable amount of surveys, data sets and maps exist for thoron (^{220}Rn). The main reason is that the effective dose from exposure in dwelling to thoron and its decay products is usually much lower than that from radon (^{222}Rn) and its decay products. This is due to its short half life (55 s, compared to 3.8 d of the ^{222}Rn) so that in most cases it infiltrates from the ground to a much lesser degree than radon (^{222}Rn). Therefore it is believed that geogenic contribution to thoron (^{220}Rn) concentration, and consequently to the one of thoron (^{220}Rn) progeny which are the main source of exposure, is small. Also, while the mean concentration ratio of ^{222}Rn progenies to ^{222}Rn gas (called equilibrium factor) is quite homogeneous within a room and it is often assumed to be about 0.4 for dwellings, such ratio is not homogeneous for thoron due to its steep gradient from the source, especially within few tenths centimetres from walls (Nuccetelli and Bochicchio, 1998; UNSCEAR, 2000; Meisenberg and Tschiersch, 2010).

During the last several years, however, a number of studies were performed with particular emphasize on thoron. We want to mention especially two international workshops on thoron: "Thoron 2010 in the Environment" held in Chiba, Japan (Radiation Protection Dosimetry, 2010) and "The new perspectives for thoron survey and dosimetry", held in Niska Banja, Serbia in 2005 (ECE II, 2005). These and publications of other authors (Martinez et al., 2004; Sreenath et al., 2004; Shang et al., 2005; Tschiersch et al., 2007) showed that sometimes high indoor thoron concentrations occur. Increasingly, experts conclude that also ^{220}Rn and its progeny should therefore be given more attention. For obtaining a more complete picture of inhalation dose it is thus necessary to have information on ^{220}Rn progeny levels in the environment as well (Porstendörfer, 1994; Steinhäusler et al., 1994). While in general it may be true that the contribution by ^{220}Rn is smaller than the one of ^{222}Rn, and respective progenies, it may be worthwhile to identify cases – regions or building conditions – where ^{220}Rn has to be considered if one does not want to severely underestimate total radon exposure.

The nationwide survey of indoor radon and thoron concentration has been implemented in the FYR of Macedonia under the National Project MAK 7002 of the International Atomic Energy Agency. Following the previous study of the indoor radon ^{222}Rn concentration in 437 dwellings across FYR of Macedonia (Stojanovska et al., 2011) which identified certain areas with higher average indoor radon concentrations, this study provides data about the indoor thoron concentrations in randomly selected dwellings over the whole territory of the country. Furthermore, seasonal and regional variability is investigated, as is influence of building characteristics (Stojanovska et al., 2011, 2012). This study is the first systematic survey on thoron in the dwellings of Republic of Macedonia and indeed one of the few country-wide ^{220}Rn surveys altogether. The surveys of ^{220}Rn/^{220}Rn-progeny have been carried out in Korea and Ireland (Kim et al., 2007; Mc Laughlin et al., 2010). This article does not pretend to be an assessment of ^{220}Rn/^{220}Rn-progeny caused exposure. Rather, it is an investigation of the occurrence of thoron (^{220}Rn) (its long-term mean concentration in dwellings, away from walls, to be precise) in dependence of several factors, namely season, region, geology and house characteristics.

The FYR of Macedonia is located on the central part of the Balkan Peninsula in South-eastern Europe. It covers an area of 25,713 km^2 and is divided into 8 administrative regions (Fig. 1). The total population is 2,022,547 and there are 564,296 households (State Statistical Office, Census, 2002). The entire territory has a transitional climate between Mediterranean and continental, quite mild in the valleys but harsher in the mountains. This implies certain construction styles and living habits which are among the factors that control indoor ^{222}Rn and ^{220}Rn.

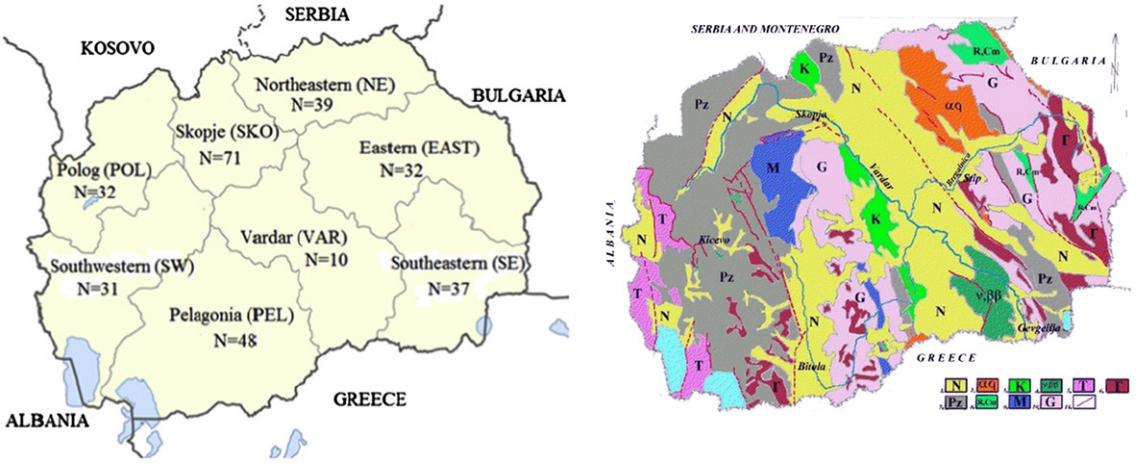

Fig. 1. Left: administrative units and number of samples; Right: geological map (Jovanovski et al., 2012); legend: 1 (N, yellow): neogene (upper tertiary–quaternary), 2 (αq, orange): vulcanites, 3 (K, green): cretaceous, 4 (ν ßß, green): gabbros and diabases, 5 (T, pink): Triassic, 6 (Γ, brown): granitoids, 7 (Pz, grey): Palaeozoic, 8 (R, Cm, green): Proterozoic, 9 (M, blue): marble, 10 (G, light pink): gneiss, red lines: faults. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2. Materials and methods

2.1. Design of the survey

Indoor thoron (^{220}Rn) gas was measured during the one year period from December 2008 to December 2009 in 300 randomly selected dwellings across the territory of the FYR of Macedonia in up to four consecutive seasons. The number of detectors which were exposed in the periods: December 2008–February 2009 (winter period), March 2009–May 2009 (spring period), June 2009–August 2009 (summer period) and September 2009–November 2009 (autumn period), were: 53, 57, 122 and 300 respectively.

The investigation area was divided into 8 regions according to administrative divisions. The number of dwellings in each area was determined, proportionally to the population density (number of households, more precisely). Fig. 1 presents the investigated regions together with the number of measured dwellings and a geological overview (Jovanovski et al., 2012).

Survey participants received detectors together with an explanation of the survey procedure, of detector handling and questionnaires asking for certain house characteristics. The categories and frequencies of occurrence, obtained from the questionnaires, are summarized in Table 1.

2.2. Placement of detectors

The detectors were set in the rooms with the highest occupancy time (either living rooms or the bedrooms) at a height of 1–1.5 m above the floor, at a distance greater than 0.5 m from each wall and at a minimum of 20 cm from any other object. This design has been chosen for the following reason. Given the short half life of ^{220}Rn, once exhaled from the wall, diffusion and advection within the room's atmosphere leads to a steeply decreasing concentration profile away from the wall, as shown e.g. in Meisenberg and Tschiersch (2010). In a distance from the wall of a few 10 cm one can usually expect a ^{220}Rn concentration level essentially due to mixing in the room, i.e. without direct influence of exhaling surfaces.

Table 1
Summary of data about thoron surveyed dwellings.

Variable	Categories	Frequencies	%
Age of the house	10–40	181	60.9
	<10	31	10.4
	>40	85	28.6
Floor	First	105	35.4
	Ground	182	61.3
	Second	10	3.4
Basement	No	128	43.1
	Yes	169	56.9
Room	Bedroom	31	10.4
	Living room	266	89.6
Building materials	Bricks	164	55.2
	Bricks/stone	38	12.8
	Concrete	55	18.5
	Concrete/bricks	9	3.0
	Concrete/stone	7	2.4
	Stone	21	7.1
	Wood	3	1.0
Type of windows	PVC	43	14.5
	Wood	254	85.5
Floor is covered with	Parquet	240	80.8
	Parquet/ceramic	5	1.7
	Ceramic	15	5.1
	Concrete	12	4.0
	Granite	6	2.0
	Laminate	18	6.1
	Marble	1	0.3
type of heating	Central heating systems	125	42.1
	Wood heating	172	57.9

2.3. Detectors

The measurements were performed with discriminative thoron/radon track-etch detectors which are ideal for long-term measurements of indoor thoron or radon (McLaughlin, 2010). The Raduet detector is a commercially available product of Radosys (Hungary), originally developed by the National Institute of Radiological Science (NIRS), Chiba, Japan (Tokonami et al., 2005). This type of detectors has been successfully used in a number of mostly regional surveys, such as in Korea (Kim et al., 2007), Serbia (Zunic et al., 2009; Milic et al., 2010), Hungary (Kovacs, 2010) and Slovenia (Vaupotic and Kavasi, 2010).

The Raduet consists of two diffusion chambers (Ø60 mm × 30 mm) with identical geometries but with different air-exchange rates. The detector's primary chamber has a high air exchange rate (low diffusion barrier) and alpha track on the CR-39 in this chamber is coming from both radon and thoron. The second diffusion chamber has low air exchange rate (high diffusion barrier), thus due to the very short half life of thoron (55.4 s), compared with that of radon (3.82 d), tracks recorded on the CR-39 in this chamber are practically only due to radon.

After exposition of the detectors, the CR-39 chips were detached from the diffusion chamber, and were chemically etched in 25% solution of NaOH at a temperature of 92 °C for 4.5 h. Counting the tracks was performed under an optical transmission light microscope, controlled by the RadoMeter 2000 analysis software. Etching and track counting of the detectors were performed in the Laboratory of the Institute of Public Health in FYR of Macedonia.

The ^{220}Rn concentration was determined from the track densities on CR-39 in the two chambers according to equation (1). The calibration factor (CF) and the adjusting factors (−0.02 and 1.21) were derived from response factors of the detectors determined in calibration chambers with controlled radon and thoron atmospheres, as explained in detail by Zhuo et al. (2002); Tokonami et al. (2005) and Sorimachi et al. (2012).

$$\begin{aligned}
\mathrm{Exp}(^{222}\mathrm{Rn}) &= \mathrm{CF}\cdot[1.00\cdot D(^{222}\mathrm{Rn}) - 0.02\cdot D(^{222}\mathrm{Rn})]\\
\mathrm{Exp}(^{222}\mathrm{Rn}) &= \mathrm{CF}\cdot[-1.00\cdot D(^{222}\mathrm{Rn}) + 1.21\cdot D(^{222}\mathrm{Rn})]\\
C(^{222}\mathrm{Rn}) &= \mathrm{Exp}(^{222}\mathrm{Rn})\ \frac{1000}{T\cdot 24}\left(\frac{\mathrm{Bq}}{\mathrm{m}^3}\right)\\
C(^{222}\mathrm{Rn}) &= \mathrm{Exp}(^{222}\mathrm{Rn})\ \frac{1000}{T\cdot 24}\left(\frac{\mathrm{Bq}}{\mathrm{m}^3}\right)
\end{aligned} \quad (1)$$

Here, $D(^{222}\mathrm{Rn})$ and $D(^{220}\mathrm{Rn})$ (tracks mm^{-2}) are the track densities recorded in the secondary and the primary chamber, respectively, with background subtracted. $\mathrm{Exp}(^{222}\mathrm{Rn})$ and $\mathrm{Exp}(^{220}\mathrm{Rn})$ are the respective radon and thoron exposures (kBq h), with CF = 36.06 (kBq h) (tr mm^{-2})$^{-1}$ the calibration factor, which together with the adjusting factors −0.02 and 1.21 is provided by the manufacturer (Radosys, Hungary). The values are specific to the series of detectors used in this project. T denotes exposure time (days) and $C(^{222}\mathrm{Rn})$ and $C(^{220}\mathrm{Rn})$ are the wanted radon and thoron activity concentrations.

2.4. Uncertainty

The results are affected by two important sources of uncertainty. Firstly, the random uncertainty associated with the number of tracks on the film resulting from a random Poisson process (including a contribution from subtraction of the background which is also affected by uncertainty); secondly, uncertainty of the calibration and adjusting factors. We face a random component of this second type of uncertainty, which stems from estimation of the response coefficients, and a systematic component due to slight physical

differences between series of detectors (if applicable). These differences are however randomly distributed between series.

This second type of uncertainty is more difficult to account for. The procedure of determining the calibration factors is described in Tokonami et al. (2005). Essentially it is based on estimating the response factors by linear regression and solving a system of two linear equations. The coefficients of this system are the response factors which, as estimates, contain uncertainty. The uncertainty propagates into the calibration factors.

More accurately, the procedure goes like this. The responses of the two detectors (low permeability or secondary, L, and high permeability or primary, H) are in terms of track densities D_L and D_H:

$$\begin{pmatrix} D_L \\ D_H \end{pmatrix} = \begin{pmatrix} B \\ B \end{pmatrix} + \begin{pmatrix} R_{Rn,L} & R_{Tn,L} \\ R_{Rn,H} & R_{Tn,H} \end{pmatrix} \begin{pmatrix} Exp_{Rn} \\ Exp_{Tn} \end{pmatrix}$$

$R_{Rn,L}$ etc. are the response factors (called CF_{Rn1} etc. in Tokonami et al., 2005), B the background (assumed the same for both detectors, but this does not matter), and D and Exp as above. This system is inverted to get eq. (1) which we use for evaluating actual measurements. CF × adjustment factors in eq. (1) are the elements of the inverse. The elements of the 2 × 2 response matrix $\boldsymbol{R}$ are uncertain, and so are consequently the elements of the inverse, $\boldsymbol{R}^{-1}$. If we call u_{11} the relative uncertainty of matrix element $\boldsymbol{R}_{11}$, etc., we find for the uncertainties of the elements of the inverse, u_{11}^{-1} etc., with a bit of algebra,

$$u_{11}^{-1} = \frac{1}{\det(\boldsymbol{R})}\sqrt{\boldsymbol{R}_{11}^2\boldsymbol{R}_{22}^2u_{11}^2 + \boldsymbol{R}_{12}^2\boldsymbol{R}_{21}^2(u_{12}^2 + u_{21}^2 + u_{22}^2)},$$

and so on. Possible covariances have been ignored here. If we assume all uncertainties u_{ij} the same equalling u, and writing the determinant explicitly, we find for the uncertainties of all elements of the inverse:

$$u^{-1} = u\frac{\sqrt{\boldsymbol{R}_{11}^2\boldsymbol{R}_{22}^2 + \boldsymbol{R}_{12}^2\boldsymbol{R}_{21}^2}}{\boldsymbol{R}_{11}\boldsymbol{R}_{22} - \boldsymbol{R}_{12}\boldsymbol{R}_{21}}$$

Using the elements of $\boldsymbol{R}$ given in Tokonami et al. (2005; Table 1, first and second row), we find $u^{-1} = 1.02\,u$. The ratio is near to 1 because the element $\boldsymbol{R}_{12}$ which is the response of the low-permeability detector to ^{220}Rn, is very small; therefore the terms with $\boldsymbol{R}_{12}$ almost disappear in the above ratio and the ratio reduces to approximately 1.

We do not know the uncertainties of the coefficients (also Tokonami et al., 2005 does not give them) but for illustrating the problem assume them as $u = 15\% \approx u^{-1}$, which is probably plausible (recent results, Sorimachi et al., 2012; however suggest that it might be higher). This can have a serious impact in particular to the second of the above equations, the one for ^{220}Rn exposure, because it includes the difference of two similar sized uncertain quantities. Assume $D(^{222}\text{Rn}) = 3$ tracks mm^{-2} and $D(^{220}\text{Rn}) = 4$ tracks mm^{-2}, 5% uncertainty for the track densities and 15% uncertainty of the calibration/adjustment factors. We find again by Gaussian propagation an uncertainty of the ^{220}Rn exposure of as much as about 49% (pertaining to: about 31 Bq m^{-3} for a 3 months exposure time; the background and its uncertainty have been neglected for the example). The lesson is that for high ^{222}Rn and low ^{220}Rn concentrations the uncertainty of ^{220}Rn can become very high.

In our investigation, the calibration/adjusting factors were taken from the manufacturer without uncertainties and were assumed to be constant within detector series, as certified by the manufacturer. In any case each series should be assigned individual calibration factors.

2.5. Decision threshold and detection limit

According to the final draft of the international standard ISO/FDIS 11665-4 (Measurement of radioactivity in the environment – Air: ^{222}Rn – Part 4: Integrated measurement method for determining average activity concentration using passive sampling and delayed analysis) decision threshold DT and detection limit LLD of the concentration for *individual* solid state detectors are given by the following equations (A.4 and A.6 of the cited reference):

$$\begin{aligned} \text{DT} &= \overline{C}^{*} = k_{1-\alpha}\cdot\tilde{u}(0) = k_{1-\alpha}\cdot w\cdot\sqrt{2\overline{n}_b} \\ \text{LLD} &= \overline{C}\# \approx \frac{2\cdot\overline{C}^{*} + k^2\cdot w}{1 - k^2\cdot u_{rel}^2(w)} \end{aligned} \quad (2)$$

and $w = (t\cdot S\cdot F_C)^{-1}$, where t = sampling time (90 d), S = detector area (46.2 mm^2), F_C = calibration factor, in (tracks cm^{-2})(Bq h m^{-3})$^{-1}$ (the inverse of the CF used by us), and $\overline{n}_b$ = mean number of background tracks. $u_{rel}(w)$ is the relative standard uncertainty of w, for which we assume 10% or 20%. Further we choose $k_{1-\alpha} = k_{1-\beta} = k = 1.65$ to $\alpha = 0.05$. The mean BG track density equals 28 tr cm^{-2}, which means a mean number of BG tracks = 12.9 for a 0.462 cm^2 detector. With $F_C = 0.00277$ (for ^{222}Rn) and $t = 2160$ h, we find:

DT = 3.0 Bq m^{-3} and LLD = 7.2 Bq m^{-3} assuming $u_{rel}(w) = 10\%$ and 7.9 Bq m^{-3} for $u_{rel}(w) = 20\%$.

The DT and LLD for the paired configuration as we use it are conceptually more difficult. As noted in Tokonami et al. (2005) the LLD of ^{220}Rn depends on the ^{222}Rn concentration and vice versa. Practically, also considering what has been said above about uncertainty propagation, detection limits are certainly higher than the values calculated above. Developing statistically sound equations for the two-detector configuration following the ISO procedure, which would yield reliable DT and LLD, must remain for further research. For now we propose the following, probably simplistic approach to estimate the LLD of thoron for the two-detector configuration: In the highly permeable detector a ^{222}Rn concentration C, but ^{220}Rn = 0 causes the number of tracks

$$n(C) = C\cdot S\cdot t\cdot F_C(^{222}\text{Rn}) + \overline{n}_b \quad (3)$$

This enters into the formulas (2) as ^{222}Rn – dependent background instead of $\overline{n}_b$, while in w the F_C (^{220}Rn) is used (i.e. accounting for the adjusting factor 1.21). $u_{rel}(w)$ is set 10% (perhaps over-optimistically). This leads to a dependency of the LLD (^{220}Rn) on the ^{222}Rn concentration as shown in Fig. 2.

We recognize a strong increase with increasing ^{222}Rn concentration, which may render low calculated ^{220}Rn values questionable in the presence of high ^{222}Rn concentrations. (The argument is however not correct for the LLD (^{222}Rn) in the presence of ^{220}Rn because the low-permeable detector has very little sensitivity for ^{220}Rn. Therefore the presence of ^{220}Rn would increase the LLD (^{222}Rn) only very little.)

2.6. General quality assurance

The above considerations show that QA which would include an uncertainty budget, confidence intervals of the results and decision thresholds and detection limits, is by no means trivial for this method. Also, so far, to our knowledge, only few and small-scale intercomparison exercises have been performed for this type of passive ^{220}Rn measurements so far (Sorimachi et al., 2012, who also give a list of references for other literature which is relevant to QA of ^{220}Rn measurement). Important results of the latter exercise are not satisfactorily explained differences of the response of Raduet detectors in two different calibration chambers, and possibly high uncertainties of the ^{220}Rn response factors.

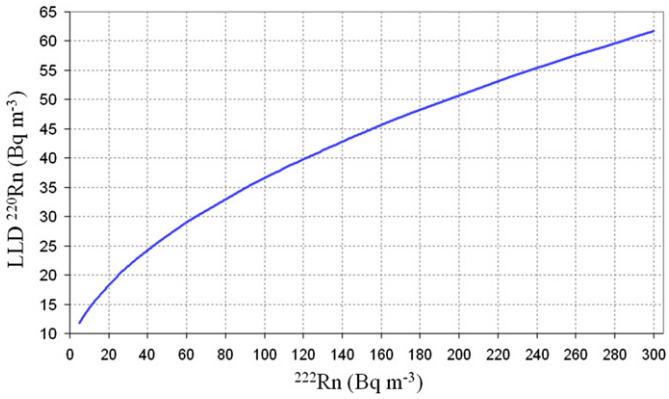

Fig. 2. Detection limit ($\alpha = \beta = 0.05$) of ^{220}Rn (thoron) in the presence of ^{222}Rn (radon).

Therefore we have to rely, for the time being, on the correctness of calibration and adjusting factors as provided by the manufacturer. We are however aware that this is a QA issue which has to be kept in mind, and one can only hope that more intercomparisons will be organized given the increasing awareness for thoron, similar to the frequent ones for radon, and that more research is being done to clarify existing discrepancies. (Some considerations of the last sections are based on recent discussions at NIRS/Chiba and RAD 2012/Serbia conferences, PB.)

3. Results and discussion

3.1. Summary statistics and seasonal variability

The summary statistics of indoor ^{220}Rn concentration measurements for each season are presented in Table 2.

Due to seasonal variations, a significant difference in concentrations between the four seasons can be observed in the results. Analysis of variance shows that the thoron concentrations were significantly higher in winter and spring than in autumn and summer. For comparison, the results from Korea showed similar temporal pattern, with the winter and spring seasons higher than those for the summer and autumn (Kim et al., 2007); on the other hand, Martinez et al. (2004) found the maximum concentration for Mexico City in the autumn season and the lowest concentration in summer.

The distribution of the indoor thoron concentrations measured in all season was accepted as log-normal by Kolmogorov–Smirnov test at a significance level of 95% (Fig. 3). We notice high geometric standard deviation up to over 3, indicating the large variability of ^{220}Rn concentration over measured rooms. Given approximate log-normality, all further statistical tests are performed only for log-transformed values of ^{220}Rn indoor concentrations, since this reduces the influence of extreme values on the statistics and also normality conditions for ANOVA are better fulfilled.

From Fig. 3 it could be seen that most of the values of thoron indoor concentration were lower than 150 Bq m^{-3}. Furthermore, the results showed that 7 dwellings in winter and 3 in autumn of the values were higher than 350 Bq m^{-3}.

Taking into account the seasonal variations of the indoor thoron concentration, ratios winter/autumn, spring/autumn and summer/autumn were estimated. The ratios of winter, spring and summer to autumn also follow a log-normal distribution with GM (GSD) 0.933 (2.86), 1.027 (2.38) and 0.676 (2.91), respectively.

Given the seasonality, we attempt to define a normalized or seasonally-corrected value of the ^{220}Rn concentration, as estimated annual average. Where measurements for all four seasons are available, the normalized value is simply the arithmetic mean of the four values. If one or more seasons are missing, the procedure is the following. For all rooms there is an "autumn" value; the values of missing seasons where estimated from this through a linear regression model of log(season) by log(autumn). The normalized annual mean is then the arithmetic mean of the values of measured and estimated seasonal values.

In order to refine the procedure it was tested if the regression models depend on other factors (Table 1). Technically, after testing of the homogeneity of variances of the groups with Bartlett's test, analysis of variance (ANOVA) and Fisher's LSD-test were applied to the seasonal ratios to determine the differences between mean values. It turned out that there is only one factor which significantly contributed at 95% confidence level (ANOVA, $p < 0.025$), namely the presence or absence of a basement for the summer/autumn ratio.

Taking into account a normal distribution of the log transformed data, the parametric linear regression analysis was applied to determined relationships between the decade logarithms ($_{10}$log or lg) of the ^{220}Rn concentrations measured in autumn to ^{220}Rn concentrations in winter, spring, as well as thoron concentration measured in autumn to measured in summer for the houses with and without basement separately. The parameters of linear fits together with Pearson coefficient of determination are presented in Table 3.

The seasonal dependence is shown graphically in Fig. 4 for thoron and radon, for comparison. The *y*-axis shows the arithmetic means and standard deviations (over all measured rooms) of $_{10}$log(seasonal value/estimated annual mean). We notice that seasonality is less pronounced for thoron, than for radon as the amplitude of its variability is smaller. The pattern appears however similar, with the difference in spring possibly insignificant given the large SDs.

Table 2
Indoor thoron (^{220}Rn) concentration measured in different seasons.

Season	N^a	^{220}Rn (Bq m^{-3})						
		Max[b]	Med[c]	AM[d]	SD[e]	SE[f]	GM[g]	GSD[h]
Winter	53	525	33	90	137	19	39	3.4
Spring	57	495	28	56	77	10	32	2.8
Summer	122	245	19	30	38	3	18	2.8
Autumn	300	395	34	52	64	4	31	2.9

[a] Number of measurements.
[b] Maximum.
[c] Median.
[d] Arithmetic mean.
[e] Standard deviation.
[f] Standard error of the mean.
[g] Geometric mean.
[h] Geometric standard deviation (dimensionless).

3.2. Regional variability of indoor thoron concentration

The annual mean indoor thoron concentration for a particular location is an average of the measurements performed in course of the survey, which were previously normalized with the corresponding seasonal correction factors as shown in Table 3. After this treatment, the resulting normalized thoron concentration data again follow a log-normal distribution with geometric mean of 28 Bq m^{-3} and geometric standard deviation of 2.12, and range 3–272 Bq m^{-3}. Note that the GSD is greatly reduced by seasonal normalization, which again shows the importance of seasonality.

The ANOVA tests showed that the mean values between the regions are statistically significant different ($p = 0.001$). The results of the estimated annual normalized indoor thoron concentration in whole regions are summarized in Table 4.

We used Fisher's LSD-test for grouping regions according to their GMs. In the following the significant higher thoron concentration in the Pelagonia versus Polog ($p = 0.001$), Skopje

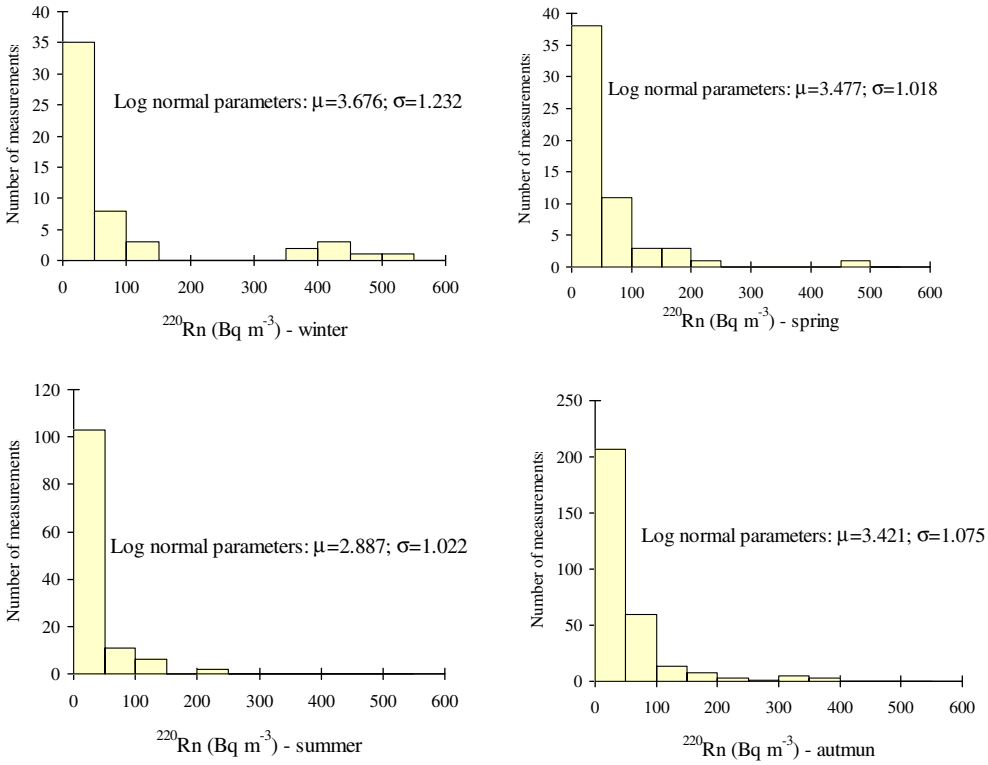

Fig. 3. Distribution of the indoor thoron concentration, measured in the winter, spring, summer and autumn.

($p = 0.0002$), South-Western ($p = 0.009$) and North-Eastern versus Polog ($p = 0.002$), Skopje ($p = 0.001$), South-Western ($p = 0.019$).

3.3. Building characteristics

Beside the regional differences, dependence of the concentrations of thoron on the characteristics of a building was examined. All categories presented in Table 1 were considered as grouping factors.

The ANOVA test results showed a statistically significant lower value in results measured in buildings with basement compared to buildings without basement ($p = 0.042$). This may indicate the presence of geogenic thoron. Differences between floor levels were not significant statistically (ANOVA, $p = 0.759$), which on the other hand suggests that building materials are mainly responsible for ^{220}Rn. A statistically significant difference was observed with buildings with different types of heating ($p = 0.037$). Buildings with a central heating system measured lower concentrations compared to stove-heated buildings (Fig. 5), for which we have no explanation at present.

ANOVA analysis showed a statistically insignificant difference between the concentrations of ^{220}Rn measured in buildings made of various buildings materials ($p = 0.062$). However, statistically significant differences can be identified between particular building materials: For instance, statistically significant higher values were measured in buildings made of stone compared to brick buildings (LSD, $p = 0.029$). In addition, higher concentrations were measured in concrete buildings compared to brick buildings (LSD, $p = 0.009$). Furthermore, concentrations of ^{220}Rn depending on construction materials grouped by the age of a building in different regions were measured, but we did not find statistically significant difference, which is perhaps because of the insufficient number of measurements in each group. As with indoor radon concentrations (Stojanovska et al., 2011), higher concentrations relate to older-aged stone homes in PEL and SE regions, and they are mostly without basement, which also significantly impacts ^{220}Rn concentrations.

Table 3
Results of the regression analysis of seasonal variation of indoor thoron (in Bq m^{-3}). Regression model: $y = Ax + B$ (lg denotes $_{10}$log).

y	x	A	B	R^2
lg(^{220}Rn), winter	lg(^{220}Rn), autumn	0.648 ± 0.109	0.542 ± 0.186	0.410
lg(^{220}Rn), spring	lg(^{220}Rn), autumn	0.590 ± 0.074	0.626 ± 0.118	0.534
lg(^{220}Rn), summer; house with basement	lg(^{220}Rn), autumn	0.472 ± 0.093	0.640 ± 0.137	0.269
lg(^{220}Rn), summer; house without basement	lg(^{220}Rn), autumn	0.468 ± 0.121	0.504 ± 0.185	0.488

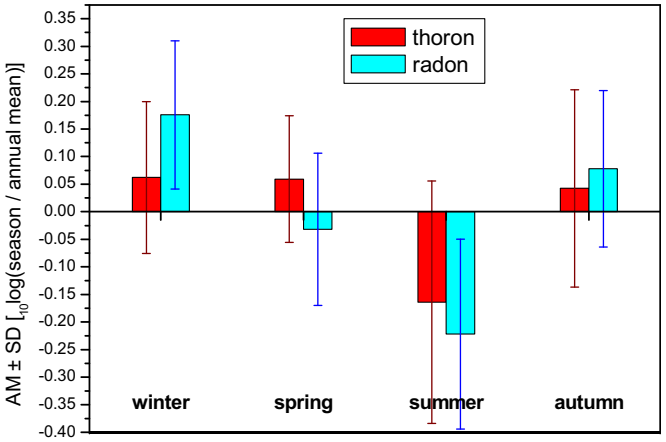

Fig. 4. Seasonality of indoor thoron and radon concentration.

Table 4
Summarized results of annual thoron concentrations in different regions.

Region	N[a]	^{220}Rn (Bq m^{-3})						
		Min[b]	Max[c]	Med[d]	AM[e]	SD[f]	GM[g]	GSD[h]
EAST	32	6	126	30	36	27	28	2.16
NE	39	10	272	36	51	50	37	2.19
PEL	48	9	197	33	54	51	38	2.29
POL	32	5	137	23	29	25	22	2.18
SKO	71	3	242	23	29	30	23	1.98
SE	37	6	105	28	34	20	28	1.86
SW	31	7	74	26	29	18	24	1.91
VAR	10	12	109	27	34	28	29	1.80
Total	300	3	272	27	37	36	28	2.12

[a] Number of measurements.
[b] Maximum.
[c] Median.
[d] Arithmetic mean.
[e] Standard deviation.
[f] Standard error of the mean.
[g] Geometric mean.
[h] Geometric standard deviation (dimensionless).

In any case one must be cautious with simple interpretations of the statistical findings. Factors may be contingent, which means that effects cannot easily be attributed to a factor in some cases, as it may in reality be the concealed effect of another factor which is contingent to the first. Separating effects of convoluted factors such as seasonal and possibly spatial geogenic variability, construction type and floor level, and possibly other controlling factors (apparently heating system) which may themselves show regional trends, by statistical means, is complicated and requires many more data than could be generated by our gross survey.

3.4. Geology

To investigate a possible role of geology ground floor data were assigned to geological units according to the map in Fig. 1. At present we do not have a more detailed geological map except for the Cenozoic basins, Dumurdzanov et al. (2004). Moreover, the assignment of sampling points is inaccurate since no digital map is available. Most samples lie in the unit labelled neogene, which covers the mostly inhabited valleys. Some samples are located near the border of neogene and Palaeozoic and neogene and granitoid, and a few in the Proterozoic zone. The remaining units essentially form comparatively sparsely inhabited mountain ranges. The "neogene" unit is not further stratified according to the genesis of the rock which may often be derived from surrounding rocks, such as the Palaeozoic or granitoid units, and is therefore probably not homogeneous with respect to ^{222}Rn and ^{220}Rn.

It appears that ground floor rooms tend to have slightly higher ^{220}Rn concentration in granitic and proterozoic units (GM: 43 Bq m^{-3}, GSD: 2.4, $n = 15$) than over Neogene and Palaeozoic (GM = 28 Bq m^{-3}, GSD = 2.2, $n = 159$), but ANOVA test (for the logarithms) is not really conclusive (F-test: $p = 0.049$, Kruskal–Wallis: $p = 0.054$).

For ^{222}Rn, neogene, Palaeozoic and Proterozoic put together have GM = 90 Bq m^{-3}, GSD = 1.7 ($n = 164$), while granitoid has GM = 138 Bq m^{-3}, GSD = 2.2 ($n = 10$). The difference is only questionably significant (F-test: $p = 0.019$ but variance test fails, K.W.: $p = 0.090$).

The relevance of geology for indoor ^{222}Rn is well known, while little is known about the relation of geology with indoor ^{220}Rn. Further elucidations must be left to further studies, requiring higher stratified geological map and a sampling scheme tailored for studying such relation. Since – due to the short half life of ^{220}Rn – only a small fraction of geogenic ^{220}Rn can be expected to contribute to indoor ^{220}Rn, compared to geogenic ^{222}Rn which is often the main source of indoor ^{222}Rn, influence of geology can be assumed of minor importance in general. It is difficult to decide at present, whether the observed statistical differences by geology are due to direct influence of geogenic ^{220}Rn, or rather to building materials preferentially made from local material; in such case geology would indirectly influence ^{220}Rn, as building material and geology would be contingent factors.

3.5. An indoor thoron map of FYR of Macedonia

The estimated mean ^{220}Rn concentration in ground floor rooms, understood as spatial random variable, shows a spatial correlation structure. In the following, the log transformed values were analysed. For local replicates (referring to rooms in the same house or in nearby houses which had been assigned the same coordinates) medians were calculated. The empirical variogram, Fig. 6, shows a distinct structure; modelled as exponential for this purpose (the relatively bad fit for high lags matters little because of the shielding effect). The variability between local replicates is however large and using all data instead of local medians would result in a high nugget effect (local variance), but still a visible structure.

The data were interpolated on a 20 km grid by ordinary block kriging. The map, Fig. 7, shows the estimated geometrical means over grid cells. The map was blanked with kriging – SD = 0.22 ($_{10}$log(Bq m^{-3})), chosen deliberately, in order to limit estimation uncertainty.

Although thoron concentration does show a distinct spatial structure, the pattern is difficult to explain. Reasons may be regional preferences of building materials or other relevant house characteristics, or indeed geogenic factors, although the pattern does not appear to coincide with the geological map. Our data are however not sufficient to clarify this at this point and the matter must be left to further investigation.

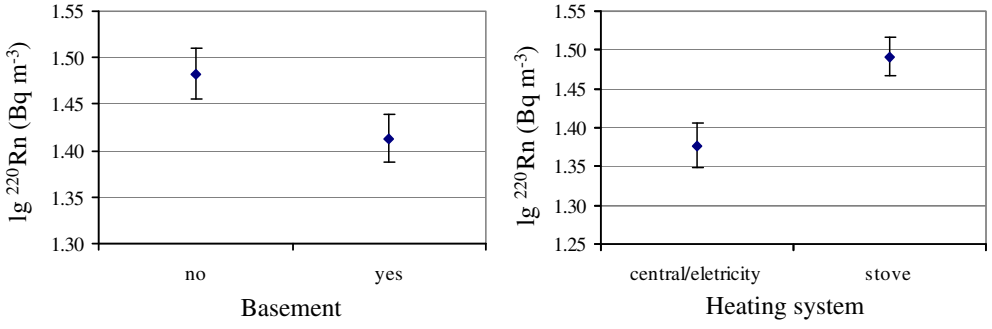

Fig. 5. Indoor ^{220}Rn concentration in building: with and without basement (left), central/electricity and stove heating (right).

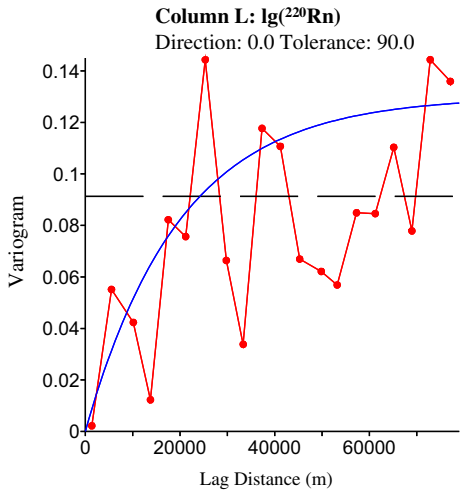

Fig. 6. Variogram of the local medians of the log-transformed estimated annual mean thoron concentrations. Blue curve: exponential model; dashed line: sample variance. Unit of *y*-axis: $({}_{10}\log(\text{Bq m}^{-3}))^2$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.6. Small scale variability

A relatively high number of observations are available for the city of Skopje (capital of the county, to be distinguished from the homonymous province) which extends over about 10–15 km, located in the Valley of Vardar River, essentially over neogene ground. The valley is surrounded by mountains belonging to Palaeozoic rocks to the West, North and South, and opens to the East. The river cuts through the mountains from the NW, so that one can assume that the neogene has been derived from these Palaeozoic rocks which are known for generally low radon potential. More details are given in Dumurdzanov et al. (2004), who also mention travertine deposits (a kind of limestone).

Table 5 gives some statistics of indoor ^{222}Rn and ^{220}Rn concentrations of ground floor rooms in Skopje. We find geometrical standard deviations (measures of variability) of 1.88 and 1.82 for ^{220}Rn and ^{222}Rn, respectively, which matches perfectly the median GSD 1.85 for ^{222}Rn within 10 km × 10 km cells over Europe (Tollefsen et al., 2011).

Table 5
Annual means of ^{220}Rn and ^{222}Rn concentrations (Bq m^{-3}) in the city of Skopje – only ground floor rooms.

	^{220}Rn	^{222}Rn
All		
n	53	53
GM (Bq/m^3)	24.3	86.9
GSD	1.88	1.82
With basement		
n	34	34
GM (Bq/m^3)	22.2	71.3
GSD	1.66	1.76
Without basement		
n	19	19
GM (Bq/m^3)	28.4	123.7
GSD	2.23	1.63

The heteroscedastic *t*-test for the logarithms of the concentrations indicates significant difference for ^{222}Rn between houses with and without basement ($p < 0.001$; hypothesis: difference = 0; one- and two-sided tests), but no such difference ($p > 0.1$) for ^{220}Rn. Presence of a basement mitigates infiltration of Rn into above-surface parts of houses, in most cases, as is well known from many studies, and also confirmed here. The finding for ^{220}Rn (no or only insignificant reduction of ^{220}Rn in houses with basement) may be a hint that geogenic ^{220}Rn contributes little, if any, to indoor ^{220}Rn in Skopje.

GSD values for other major towns (all smaller than Skopje) are given in Table 6, without further distinguishing between houses with and without basement, for lack of sufficient data. We notice that in all cases the variability of ^{220}Rn is higher than the one of ^{222}Rn. Without being able to prove it, we think that a major contribution to ^{220}Rn variability comes from the high relative uncertainty in determining the ^{220}Rn concentration, as explained in Section 2.4, and is therefore not one related to the true variability of ^{220}Rn. All values are however in the same order of magnitude.

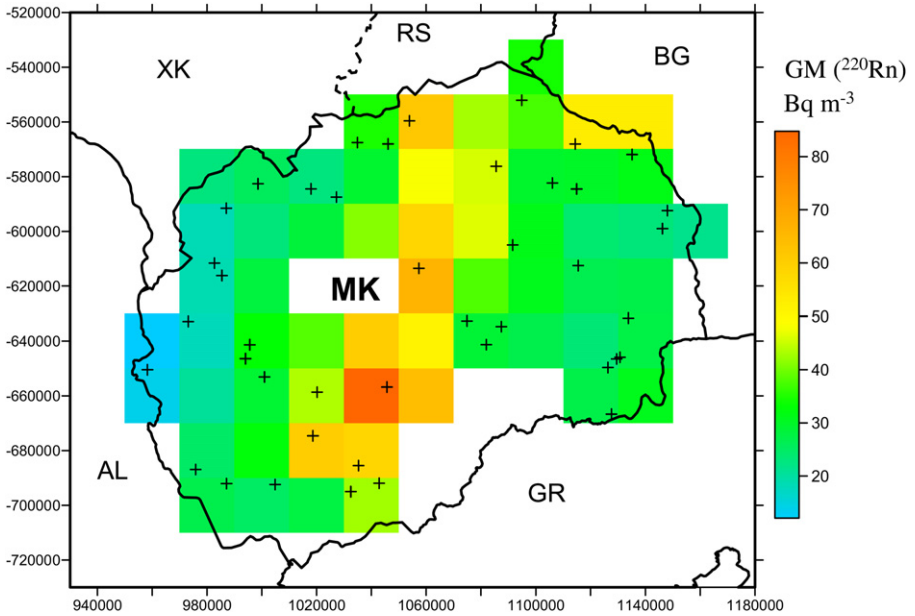

Fig. 7. Estimated geometrical means of annual mean thoron concentrations in ground floor rooms over 20 km × 20 km cells. Coordinates: GISCO Lambert azimuthal equal-area projection centred at 9°E, 48°N, unit: m. Crosses: sampling locations.

Table 6
Geometrical standard deviations of ^{220}Rn and ^{222}Rn concentrations in various cities.

	^{220}Rn	^{222}Rn
Bitola	2.07	1.37
Gostivar	2.74	1.85
Kicevo	2.18	1.74
Kumanovo	2.49	1.57
Skopje	1.88	1.82
Stip	2.21	1.84
Strumica	2.10	1.65
Median	*2.18*	*1.74*

The general message is that considerable small-scale variability remains, which has to be kept in mind when interpreting maps such as the one in Fig. 7.

3.7. Thoron and radon correlation

Finally correlation between indoor thoron and indoor radon concentration was investigated. The linear regression analysis shows a weakly positive correlation with Spearman coefficient of $R = 0.335$. This survey showed that thoron concentrations are not negligible in comparison to radon concentrations, in many cases. The ratio thoron/radon concentrations ranged between 0.05 and 3.38, and again follow a log-normal distribution, approximately. The arithmetic mean of thoron/radon equals 0.51 (SD = 0.49) and geometric mean 0.37 (GSD = 2.19).

4. Conclusions

The results show that in the FYR of Macedonia, in most instances the thoron (^{220}Rn) concentrations are not negligible compared to those of the radon (^{222}Rn). The indoor thoron concentrations show seasonal and regional variability and depend on house characteristics, notably the presence of a basement and kind of heating system, but not significantly the building material in general (differently from one might have expected). However, certain types of building materials could be indeed significantly associated with higher or lower thoron concentrations (stone and concrete vs. brick).

It is however difficult to identify the sources of thoron. It is mostly assumed that the main source is exhalation from building material, while transport from the ground is little relevant due to the short half life of thoron. Also our data do not clarify this. Considering the occurrence of higher indoor thoron concentrations, we suggest that it may be worthwhile to identify situations in which the geogenic contribution could be indeed relevant and allocate them geographically. Reasons may be particularly high thoron concentration in the ground and poor insulation of the house against the ground which would facilitate thoron infiltration by advective transport which is in general more efficient than diffusive transport, despite its short half life.

The identified spatial dependence of thoron concentration which results in the thoron map can have two sources. Firstly: regional prevalence of building materials which have different thoron exhalation rates; secondly: a contribution of geogenic thoron, related to geology (since rocks have different thoron content and thoron exhalation potential). Both factors can however be contingent if building materials tend to be made of local material. The available data are not sufficient to decide this at present.

Another issue which requires further attention is establishing an uncertainty budget which accounts for calibration uncertainty, defining detection limit for the two-detector configuration and more generally, QA achievable only through more practical experience, additional research on the behaviour of thoron in solid-state detectors and intercomparisons.

Acknowledgment

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