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THORON IN SWEDISH BUILDINGS*

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ABSTRACT

Measurements of thoron (^{220}Rn) have been performed in some 90 buildings during 1992 and 1993, as parts of two different investigations. The measurements were performed in order to form a basis for assessing the risk for high thoron levels indoors in Sweden. In the first study thoron progeny measurements were undertaken in 53 dwellings and 7 workplaces at 13 different locations in south and central Sweden. Most buildings were situated in areas with elevated natural thorium concentrations. In each dwelling two measurements were made; in the living-room or a bedroom and in the basement. The radon (^{222}Rn) progeny concentration was measured at the same time and the thorium and uranium activty concentrations in the rock and soil surrounding the buildings were measured with a portable gamma spectrometer. Typical thoron progeny values (Equilibrium Equivalent concentration of Thoron, EET) in the living areas were in the range 0.1 to 1.0 Bq.m^{-3} (8 to 76 nJ.m^{-3}, 0.4 to 3.6 mWL, Potential Alpha Energy Concentration, PAEC and Working Level for thoron progeny) with a mean of 0.9 Bq.m^{-3} (68 nJ.m^{-3}, 3.3 mWL). The measurements in the basements had a mean of 1.5 Bq.m^{-3} (114 nJ.m^{-3}, 5.5 mWL). A few measurements of thoron gas were performed in basements and showed typical values between 10 and 80 Bq.m^{-3}, with a maximum of 430 Bq.m^{-3}. As part of a study of long-term variations of ^{222}Rn in dwellings located in different parts of Sweden, 24 hour continuous measurements of thoron gas were performed in 28 of these dwellings. In this study the buildings were chosen regardless of natural thorium concentrations. The mean of these measurements was 13 Bq.m^{-3}. One high value of 150 Bq.m^{-3} was found. The estimated population mean for thoron progeny in Swedish homes is 0.5 Bq.m^{-3} (38 nJ.m^{-3}, 1.8 mWL). This would give an approximate annual effective dose to the average Swede of 0.1 mSv.

INTRODUCTION

Thoron (^{220}Rn) has a short half-life, 55.6 seconds, compared to radon (^{222}Rn) with a half-life of 3.8 days. This means that the distance that the thoron gas atoms can migrate in the ground and inside building materials and buildings before it decays is much shorter than for radon gas and also that it is comparatively easily stopped by wall paper and other surface sealants. Therefore the risk for high thoron levels indoors can be expected to be low, at least much lower than the risk for high levels of radon. However, in buildings with an ineffective barrier between soil and indoor air the entry of thoron could be significant, especially if the gravel or sand filler or the soil itself immediately under the building has a high concentration of ^{232}Th. Soil as a significant source of indoor thoron has been demonstrated by Li and coworkers (Li et al., 1992). The release of thoron from building materials has also to be considered, especially where unsealed surfaces are present, such as bare walls, floors or ceilings. Under certain circumstances outdoor air can probably also be a source of high thoron levels indoors. The concentration of thoron outdoors has been measured as high as 200 Bq.m^{-3} at 1 m above ground (Porstendörfer, 1994), and is strongly dependent on height. A few cm above ground the thoron concentration can be an order of magnitude higher. In Sweden, as part of this study the thoron gas concentration outdoors at a site near one of the office buildings of our institute was measured for about two months beginning September 1991. The thoron concentration was measured at 10 cm above ground and was found to vary between 20 Bq.m^{-3} and 330 Bq.m^{-3} with an approximate mean of 90 Bq.m^{-3}. If the outdoor thoron concentration is high and the air inlet ducts are placed near the ground, outdoor air could be a source of indoor thoron.

In 1990-1991 a pilot study on thoron progeny in dwellings and workplaces was performed in Sweden (Mjönes et al., 1992). In order to further assess the risk for high thoron levels indoors, the Swedish Radiation Protection Institute (SSI), has performed measurements of thoron and thoron progeny in some 90 buildings. Most measurements were made during 1992 and 1993, as parts of two different investigations. In the first study, here called study No. 1, thoron progeny measurements were undertaken in some 45 single-family houses and in a small number of apartments in multi-family buildings and workplaces situated in south and central Sweden. Most buildings in study No. 1 were situated in areas with elevated natural thorium concentrations. Sweden has several areas with natural thorium concentrations of 200-500 Bq.kg^{-1}. As part of an ongoing study of long-term variations of radon in 158 single-family houses located in different parts of Sweden, 24 hour continuous measurements of thoron gas were performed in 28 of these houses. In this study, called study No. 2, the buildings were chosen regardless of natural thorium concentrations (Hubbard et al., 1995)

ENVIRONMENTAL LEVELS OF THORON

Data on indoor thoron are limited. Surveys from different countries show mean thoron progeny values (Equilibrium Equivalent concentration of Thoron, EET) from 0.3 Bq.m^{-3} to 3.5 Bq.m^{-3} (23 nJ.m^{-3} to 265 nJ.m^{-3}, 1.1 mWL to 12.7 mWL, Potential Alpha Energy Concentration, PAEC and Working Level for thoron progeny), with most means below 1.0 Bq.m^{-3} (76 nJ.m^{-3}, 3.6 mWL) (Mjönes et al., 1992, Steinhäusler et al. 1994). The most extensive surveys have been performed in the UK, Germany and France. In the UK, measurements have been made in 390 dwellings. The mean of these measurements was 0.6 Bq.m^{-3} (46 nJ.m^{-3}, 2.2 mWL). According to the authors, results from measurements in 150 dwellings in the Pennines, with a mean of 0.3 Bq.m^{-3} (23 nJ.m^{-3}, 1.1 mWL), are more likely to be typical of the concentrations in UK dwellings (Wrixon et al., 1988). This has been confirmed by measurements in 50 dwellings in five major cities in the UK (Cliff et al., 1992). In Germany Keller and coworkers (Keller et al., 1982) have published results from measurements in 148 homes with a median of 0.3 Bq.m^{-3} (23 nJ.m^{-3}, 1.1 mWL). From two departments in a granitic area in Brittany in western France medians of 0.7 and 0.8 Bq.m^{-3} (53 and 61 nJ.m^{-3}, 2.5 and 2.9 mWL) have been reported (Rannou et al., 1988). Schery has measured ^{212}Pb at 68 indoor locations in 18 states in the United States and found a mean of 0.28 Bq.m^{-3} (21 nJ.m^{-3}, 1.0 mWL) (Schery, 1985). In Elliot Lake in Canada 95 dwellings have been investigated with a mean of 1.5 Bq.m^{-3} (114 nJ.m^{-3}, 5.5 mWL) (Gunning and Scott, 1982). Thoron progeny investigations have also been reported from, for example, Norway and Hong Kong (Stranden, 1980, Tso and Li, 1987).

In recent years several investigations concerning thoron have been performed by japanese research groups. They have found relatively high levels of thoron progeny and thoron gas in traditional japanese buildings with one or more walls covered with a widely used soil-based plaster. Guo and coworkers (Guo et al., 1992) have found a mean thoron progeny concentration of 3.5 Bq.m^{-3} (265 nJ.m^{-3}, 12.7 mWL) in 4 houses with plaster walls in the Nagoya area. A mean thoron gas concentration of 394 Bq.m^{-3} was found from measurements in 9 such houses (Guo et al., 1993).

Doi and coworkers have measured activity concentrations of thoron gas in 21 houses in the Hiroshima prefecture with a mean of 85 Bq.m^{-3} and a maximum of 550 Bq.m^{-3}. The measurements were made 20 cm from a wall (Doi et al., 1994b). Several investigations show that the thoron concentration decreases exponentially with the distance from a soil-plastered wall (Doi et al., 1994a, Guo et al., 1993). Measurements of the concentration of ^{232}Th in the soil have shown values of about 90 Bq.kg^{-1} in the Hiroshima area (Doi et al., 1994b) and about 35 Bq.kg^{-1} in the Nagoya area (Guo et al., 1992).

The Swedish preliminary study included thoron progeny measurements in nine dwellings, eight basements and four workplaces in 12 different buildings. In this study the mean thoron progeny concentration in the measured dwellings was 0.3 Bq.m^{-3} (EET) (23 nJ.m^{-3}, 1.1 mWL) and in the basements 1.5 Bq.m^{-3} (114 nJ.m^{-3}, 5.5 mWL).

measured with a gamma meter and a portable gamma spectrometer.

In study No. 2, continuous measurements of thoron gas were performed. In this study the buildings were chosen regardless of natural thorium concentrations. 24 hour measurements were made in one room, mostly the living-room or a bedroom, in the ground floor or in the basement. The thoron gas meter was placed on the floor close to one of the walls. Simultaneous radon gas measurements were performed with the same equipment as in study No. 1.

Thoron gas measurements

The thoron gas measurements were performed with the Falk-Möre-Nyblom multiple time analysis method (Falk et al., 1992). The technique is based on selective alpha-counting of the short-lived nuclide ^{216}Po, half-life 0.145 s. The alpha particle from the decay of a ^{220}Rn atom will be followed, within a fraction of a second, by an alpha particle from the decay of ^{216}Po. By measuring the time interval between succesive alpha counts and a following multiple time analysis, the ^{216}Po activity and thereby the ^{220}Rn activity can be calculated.

The detector is a commercial radon monitor using a 160 ml Lucas type scintillator cell working in flow-through mode. Air is sucked through a filter to remove any airborne particles and decay products from ^{220}Rn and ^{222}Rn before entering the sensitive volume. The flow rate was 1.5 liter per minute.

Thoron progeny measurements

The progeny measurements have been performed using alpha spectroscopy with two postsampling counting periods. The general outline of this method has been presented by several authors, for example Martz and coworkers (Martz et al., 1969).

At the sampling place air is pulled through a 180 mesh wire screen (to measure the unattached fraction of radon and thoron progeny) and a 0.8 μm millipore filter at a rate of approximately 15 $l.min^{-1}$. The air volume is measured with a gas meter. The radiation on the filter and the wire screen is counted with a surface barrier silicon alpha detector and a PC-based spectroscopy system. The sampling time was 60 minutes and the filter and the wire screen were first counted for 22 minutes followed by a 35 minute interval and then a second count was taken for 61 minutes. Using three energy channels and the two counting periods the concentrations of ^{218}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC), ^{212}Pb (ThB) and ^{212}Bi/^{212}Po (ThC/ThC') and consequently the equilibrium equivalent concentrations of radon and thoron (EER and EET) can be calculated. The equipment has been calibrated in one of the radon chambers at the Swedish Radiation Protection Institute (Falk et al., 1994).

Other measurements

The radon gas measurements were made with a commercial continuous radon gas monitor (Baltzer et al., 1992). A portable NaI-based 256 channel gamma spectrometer was used for the measurements of thorium and uranium concentrations in the rock and soil surrounding the buildings and a commercial NaI gamma meter was used for the gamma radiation measurements.

RESULTS

Thoron gas measurements

Thoron gas measurements were made in 45 single-family houses. The mean of these measurements was 30 $Bq.m^{-3}$. The mean of the thoron gas measurements from study No. 1, where most buildings were situated in areas with high natural thorium concentrations in the ground, was 55 $Bq.m^{-3}$. Here two high values were found 430 $Bq.m^{-3}$ and 290 $Bq.m^{-3}$, both measured in closets in the basement. If these two values are excluded the mean will be 20 $Bq.m^{-3}$. The mean from study No. 2, where the locations were chosen regardless of natural thorium concentrations, was 13 $Bq.m^{-3}$. Here one high value of 150 $Bq.m^{-3}$ was found, measured in a living-room in the ground floor. The mean for measurements made in the ground floor was 15 $Bq.m^{-3}$ while the mean for basement measurements was 40 $Bq.m^{-3}$. The thoron gas measurements are summarized in Table 3. The frequency distribution of all the thoron gas measurements from studies 1 and 2 is presented in Fig. 1.

Regression analysis show only a very weak correlation (r=0.25) between thoron gas and radon gas when applied to all thoron gas measurements (studies 1 + 2). When the analysis is restricted to the thoron gas measurements from study No. 2 no such correlation can be found (r=0.03).

Thoron progeny

Thoron progeny measurements have been performed in 45 single-family houses, in 5 apartments in multi-family buildings and in 9 workplaces (in 7 different buildings). The mean equilibrium equivalent concentration

Table 3. Thoron gas measurements in Swedish dwellings

Type of site.	Number of samples	Arithmetic mean (Bq.m^{-3})	Range (Bq.m^{-3})	Standard deviation (Bq.m^{-3})
All measurements	47	30.3	1-430	76.4
All single-family houses	45	31.3	1-430	79.9
High Th areas[a]	19	55.3	1-430	112.3
Random Th areas[b]	28	13.3	2-151	28.2
Ground floor	18	14.8	2-151	34.6
Basements	29	39.8	1-430	92.8

[a] Most buildings were situated in areas with elevated natural thorium concentrations
[b] The buildings were chosen regardless of natural thorium concentrations

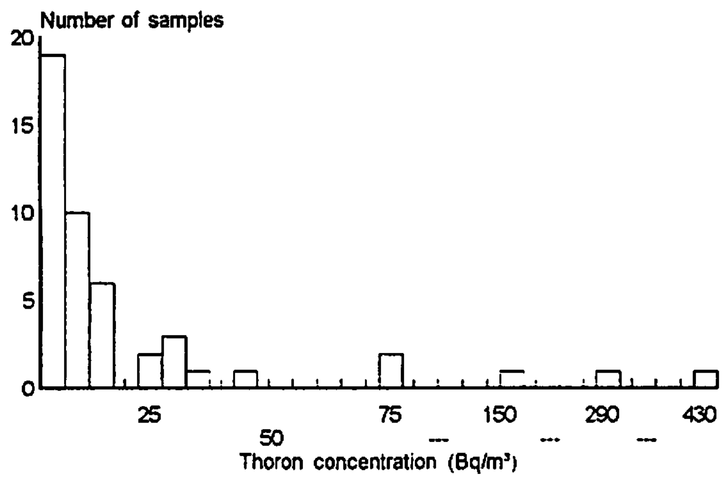

Fig. 1. Frequency distribution of thoron gas measurements indoors in Sweden

of thoron (EET) for the measurements in the ground floor in single-family houses was 0.9 Bq.m^{-3} (68 nJ.m^{-3}, 3.3 mWL). The maximum value, 16 Bq.m^{-3}, (1212 nJ.m^{-3}, 58 mWL) was measured in a wooden, two-storey house in central Sweden. The radon progeny level was also high in this house, 2 700 Bq.m^{-3} EER (Equilibrium Equivalent concentration of Radon) (15 000 nJ.m^{-3} , 0.7 WL). Measurements in the basements of the single-family houses had a mean of 1.5 Bq.m^{-3}. The mean of 5 measurements in multi-family buildings was 0.6 Bq.m^{-3} (46 nJ.m^{-3} , 2.2mWL) and in 9 workplaces 0.2 Bq.m^{-3} (15 nJ.m^{-3} , 0.7 mWL), Table 4a. The corresponding results for the radon progeny measurements are presented in Table 4b.

Regression analysis was performed to seek correlations between the measured thoron progeny values and the two parameters ^{232}Th concentration in the ground outside the building and the measured indoor radon progeny values. Surprisingly no such correlations were found. The reason for this is not known and will require further research.

The unattached fraction of thoron progeny was measured in the single-family houses. For the ground floor the results varied from less than the detection limit up to 0.8 Bq.m^{-3} EET (61 nJ.m^{-3}, 2.9 mWL) with a preliminary mean of 0.03 Bq.m^{-3} EET (2.3 nJ.m^{-3}, 0.1 mWL). For the basements the maximum was 6.3 Bq.m^{-3} EET (477 nJ.m^{-3}, 22.9 mWL) with the preliminary mean 0.2 Bq.m^{-3} EET (15 nJ.m^{-3}, 0.7 mWL). The details of these measurements will be presented elsewhere.

Table 4a. Thoron progeny measurements indoors in Sweden. Most measurements were made in houses situated in areas with high natural thorium concentrations.

Type of site	No. of samples	Arithm. mean (Bq.m^{-3})^{a}	Arithm. mean (nJ.m^{-3})	Range (Bq.m^{-3})^{a}	Range (nJ.m^{-3})	Standard deviation (Bq.m^{-3})^{a}	Standard deviation (nJ.m^{-3})
Single-family houses: Ground floor	45	0.9	68	0.02-16	2-1212	2.5	189
Single-family houses: Basements	37	1.5	114	0.06-12	5- 909	2.3	174
Multifamily houses	5	0.6	46	0.4 -0.9	30- 68		
Workplaces	9	0.2	15	0.02-0.9	2- 68		

^{a} Equilibrium Equivalent concentration of Thoron, EET
1 Bq.m^{-3} (EET) = 3.64 mWL

Table 4b. Radon progeny measurements at the same sites as in Table 4a.

Type of site	No. of samples	Arithmetic mean (Bq.m^{-3})^{a}	Arithmetic mean (nJ.m^{-3})	Range (Bq.m^{-3})^{a}	Range (nJ.m^{-3})	Standard deviation (Bq.m^{-3})^{a}	Standard deviation (nJ.m^{-3})
Single-family houses: Ground floor	45	104	575	1-2657	6-14 680	396	2188
Single-family houses: Basements	37	194	1072	3-3636	17-20 088	617	3409
Multifamily houses	5	7	39	5- 11	28- 61		
Workplaces	8	7	39	1- 29	6- 160		

^{a} Equilibrium Equivalent concentration of Radon, EER
1 Bq.m^{-3} (EER) = 0.00027 WL

Frequency distributions of the thoron progeny measurements are presented in Figures 2 and 3.

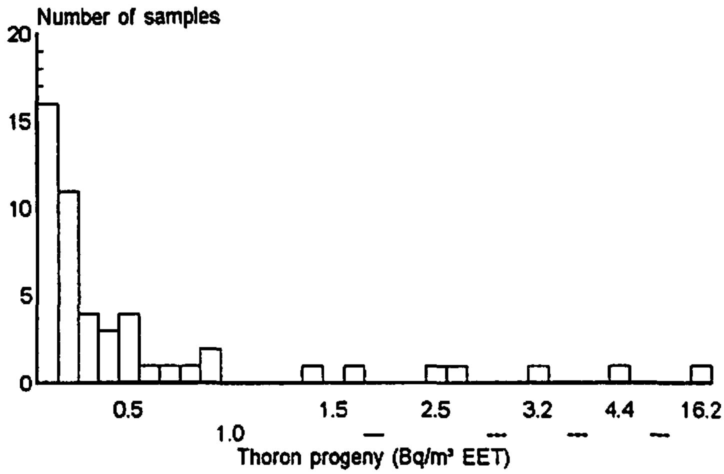

Fig. 2. Frequency distribution of thoron progeny measurements indoors in Sweden, ground floor measurements. (1 $Bq.m^{-3}$ EET = 75.8 $nJ.m^{-3}$, 1 $Bq.m^{-3}$ EET = 3.64 mWL).

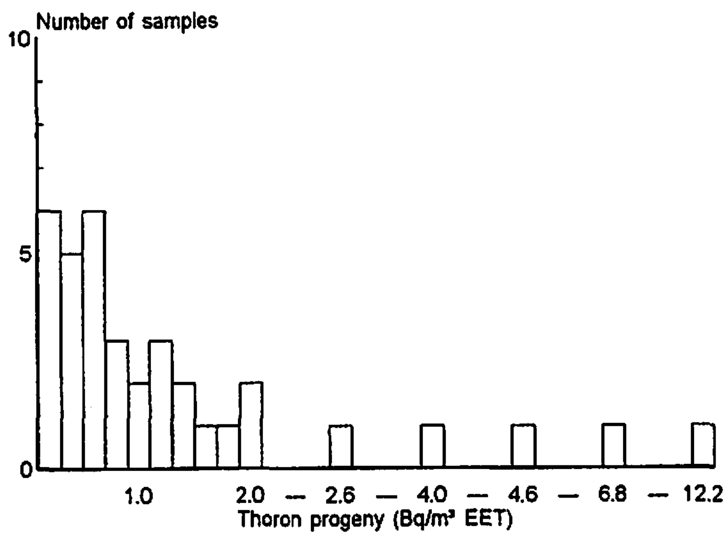

Fig. 3. Frequency distribution of thoron progeny measurements indoors in Sweden, basement measurements. (1 $Bq.m^{-3}$ EET = 75.8 $nJ.m^{-3}$, 1 $Bq.m^{-3}$ EET = 3.64 mWL).

Other measurements

In one of the houses in study No. 2 some follow-up measurements were performed in 1995. The 24 hour thoron gas measurement from 1993 had a mean value of 150 $Bq.m^{-3}$. The house is a two-storey building with a basement, situated in a rural area outside Stockholm. The ^{232}Th concentration outside the house varied between 80 and 110 $Bq.kg^{-1}$. Continuous measurements of thoron and radon gas were performed for eight days and thoron progeny measurements were made on three occasions during this time. The mean thoron gas concentration, measured 40 cm above the floor and 30 cm from an interior wall, was 46 $Bq.m^{-3}$ and the radon gas concentration 110 $Bq.m^{-3}$. The variation of the thoron gas concentration is shown in Fig. 4.

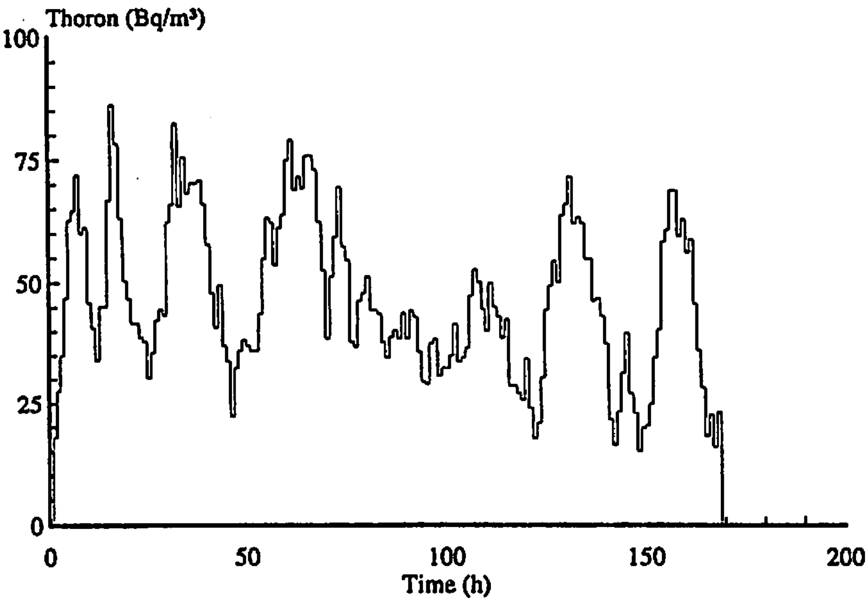

Fig. 4. The variation of the thoron gas concentration for eight days in a detached two-storey house

Thoron progeny was measured on three occasions at the same spot as the thoron gas, and varied between 0.2 and 0.3 $Bq.m^{-3}$ which would give an average equilibrium factor for thoron of about 0.005. However, as the japanese groups have shown, the thoron gas concentration is very much dependent on where in the room it is measured. The thoron progeny concentration, on the other hand, should be much more homogeneuos. This limits the practical use of the F-factor in calculating the thoron progeny concentration from thoron gas measurements and vice versa.

DISCUSSION AND CONCLUSIONS

The aim of the studies has been to find an approximate mean and range of thoron and thoron progeny in buildings in order to assess the possible health hazards from thoron indoors in Sweden. The thoron progeny measurements in single-family houses have a mean of 0.9 $Bq.m^{-3}$ EET (68 $nJ.m^{-3}$, 3.3 mWL). Taking into account that these measurements were made in selected areas with a high natural ^{232}Th concentration and that measurements in multi-family buildings show lower levels this value is probably an overestimation of the average thoron progeny level in Swedish dwellings. A level of 0.5 $Bq.m^{-3}$ (38 $nJ.m^{-3}$, 1.8 mWL) is probably a better estimation.

According to UNSCEAR the dose coefficient corresponding to inhalation of thoron progeny can be taken to be 32 nSv per $Bq.h.m^{-3}$ (UNSCEAR 1993). Assuming an occupancy factor of 0.8 indoors this would give an approximate annual effective dose to the average Swede from indoor thoron progeny of 0.1 mSv. The corresponding mean annual effective dose from radon progeny is about 2 mSv in Sweden. At the maximum thoron progeny level found so far in Sweden, 16 $Bq.m^{-3}$, the annual effective dose would be slighly over 3 mSv.

The mean thoron gas concentration in Swedish dwellings can be estimated to be 10 to 15 $Bq.m^{-3}$. The contribution to the mean annual effective dose from inhalation of thoron gas is very small. The dose coefficient according to UNSCEAR is 0.1 nSv per $Bq.h.m^{-3}$. Thus, an average of 10 $Bq.m^{-3}$ would contribute less than 0.01 mSv to the annual effective dose.

Large spatial variations of thoron gas indoors have ben demonstrated by several researchers. This means that it is difficult to assess the mean thoron gas concentration in a room and consequently the exposure of the residents to thoron gas. It also means that it is difficult to use an estimated F-factor for thoron to determine the thoron progeny concentration from thoron gas measurements. The contribution from thoron gas itself to the effective dose is small. Therefore, for dose calculation purposes, it seems better to rely on direct measurements of thoron progeny

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