MEASUREMENT OF THORON AND ITS PROGENY IN TRADITIONAL AND MODERN EARTHEN BUILDINGS IN <u>GERMANY: METHODOLOGY AND RESULTS</u>

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Abstract

In the past, the radon isotope ²²⁰Rn (thoron) was often paid little attention in the context of radiation protection at home. Measurements in buildings on the Central-Chinese Loess Plateau have shown, however, that in dwellings built from unfired earthen material, thoron and its progenies can significantly contribute to the inhalation dose of the dwellers. In Germany, such buildings mainly comprise traditional half-timbered houses and modern ecological low-energy houses with clay boards and plaster. Measurements of thoron in several such houses show concentrations of up to 150 Bq/m (at 20 cm distance from the wall). Radon and thoron progenies were measured with a newly constructed device which makes use of aerosol precipitation in an electric field upon solid-state nuclear track detectors. Thoron progeny concentrations were in the range from 100 to 1000 nJ/m resulting in a dose contribution of up to about 4 mSv per year.

Introduction

The radioactive noble gas radon is a well-known health risk in the indoor environment. Exposure to radon and its progeny can cause significant dose to the lungs and other tissue. Historically, most studies have focused on the exposure from the decay chain of the isotope ²²²Rn (radon), with little attention paid to the isotope ²²⁰Rn (thoron) (e.g. Stranden, 1980; Steinhäusler, 1996). The rationale being that its short half-life (55.6 seconds) is insufficient for it to diffuse from soil or rock under the building and into the indoor environment. However, this assumption is not true if the thoron emanates from building material or in cases where it can concentrate under a structure with a tight foundation. For example, increased thoron concentrations have recently been found in residences made of unfired mineral building material as adobe and mud bricks (Sreenath Reddy, 2004), or which are dug directly into the soil as on the Central-Chinese Loess Plateau (Wiegand, 2000; Shang, 2008). In these cases, thoron contributes up to 50% of the inhalation dose (Shang, 2005). It is therefore safe to conclude, that other homes built with similar materials may also have similar thoron concentrations and merit more detailed investigation.

The use of unfired mineral building materials is common throughout most parts of the world. For example, in Germany, traditional buildings are mostly half-timbered houses, erected with a timbered frame construction. The panels between the timbers are filled directly with mud or with boards of straw or wood and then covered with mud. Modern earthen architecture is also

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popular in ecological and health-conscious habitation. In this style of construction mud render or panels on conventionally built walls are most common. In both traditional and modern construction only the walls and in some cases the ceiling are decorated with mud with the floor being excluded. Currently about two million buildings in Germany contain earthen building material (Schreckenbach, 2004). Therefore, surveillance of the occurrence of thoron in houses built of such material is warranted.

Like radon, the inhaled radiation dose received by thoron is not caused by the parent, but by the subsequent decay of it decay products. However, unlike radon where a single measurement is usually adequate, to accurately measure the inhalation dose of thoron, many measurements need to be taken at different locations. The reasons for this are:

- The spatial distribution of the thoron concentration is highly inhomogeneous with high thoron concentrations close to earthen walls, floor, and ceiling. However, the decay products are distributed homogeneously throughout the house.
- Thoron emanation from the building materials might also vary within different rooms of a house because of differences in the building material properties. Also, the concentration of the decay products are influenced by the indoor transport within the whole house-the equilibrium factor of thoron is also influenced by indoor atmospheric parameters such as ventilation more strongly than that of radon.

For a dose assessment from a single-point thoron measurement, a homogeneous exhalation rate and a specific spatial distribution of thoron must be assumed. Even if thoron measurements at a variety of positions in a room or a house are performed, the equilibrium factors between the progenies and the gas must be known. Therefore for dose assessments, direct measurements of the thoron progenies might be preferred to measurements of the gas.

Because of their longer half-lives, thoron progenies cause an inhalation dose of about 14 times greater than that of radon progenies at the same concentration in Bq/m (Porstendörfer, 1994). Although progeny measurements are frequently reported using working level (WL), comparing levels of radon and thoron can be difficult because of the inhomogeneous distribution of thoron and the different equilibrium factors. A more accurate representation of the energy released in air is measured by using the potential alpha energy concentrations (PAEC). Briefly the PAEC is the sum of alpha energies emitted by the progenies contained in unit volume until decay to a stable nuclide reported in μ J/m

Results from measurements of thoron and its progenies with active and passive measurement devices in 17 traditional and modern houses with earthen building material are presented.

Methods

Passive measurements of thoron progenies

For passive thoron progenies measurements a newly developed measurement device was applied (Figure 1). These unattended, battery-operated progeny measurement devices (UBPMs) make use of the precipitation of aerosol particles onto a CR39 solid-state nuclear track detectors as sampling substrates within an electric field. The substrates are kept at a potential of + 7 kV compared to a hemispheric wire-mesh covering the substrates, which is on the ground potential. The UBPMs are an enhancement of a device first described by Bi et al. (2011). These devices were calibrated against working level monitors inside the HMGU thoron experimental house (Tschiersch and Meisenberg, 2010). Further details about the UBPMs can be found in Gierl et al. (2013).



Figure 1: Photo of one of the passive UBPM thoron progeny measurements devices used in this study.

Passive radon and thoron measurements

CR39 solid-state nuclear track detectors (SSNTD) were used for passive, integrating measurements of radon and thoron in indoor air: A 1 cm CR39 plastic polymer platelet is placed at the bottom of a 3.6 cm tall plastic box. For measurements of both radon and thoron, the screw cap of the box contains several holes, which are covered with a membrane filter and conductive foam through which the gases but not their progenies can enter the box. The boxes screw caps, which are used for measurements of radon only, are solid; the gas enters the box through the cap thread. For measurements of thoron concentration one device or one set of devices for radon and thoron are used together with at least one device for radon only. The measurement devices are calibrated against a RAD7 active radon and thoron measurement device (Durridge Inc.) in a 1 m calibration chamber. The quality of the measurement results is assured by frequent participation in laboratory intercomparison measurements (e.g. Janik et al. 2010).

Measurements were performed at different distances from a clay wall depending on the number of devices which could be deployed (at distance of 20 cm only or at 10 and 50 cm). All other detectors were placed at least 50 cm from any other wall, floor, and ceiling. The plastic boxes containing the CR39 detectors were placed in cardboard holders which were fixed at the wall or in wooden stands (Figure (2)) or were placed on a desktop. In several measurements, additional passive detectors were placed at different positions 20 cm from the wall, but not in a direct line between any other detectors and the wall. In addition, detectors were also deployed in the middle of the rooms.



Figure (2): Placement of a combined radon and thoron and a thoron-only passive measurement device at a distance of 20 cm from a clay paneled and rendered wall.

Results

Concentrations of radon, thoron and thoron progeny were measured for one to two months in

Figure (3): Information about houses in which measurements were performed. Clay render refers to a conventional construction on which clay render was applied. The other types of earthen architecture might also feature clay render on the respective underlying clay structure.

			Measurement of	
			radon/thoron	progenies
No.	Architecture	Usage	(distance in cm)	
1	half-timbered	residential	20	Х
2	clay render	residential	20	Х
3	half-timbered	residential	20	Х
4	half-timbered	town hall	20	Х
5	half-timbered	residential	20	Х
6	clay render	residential	20	Х
7	half-timbered	residential	10/50	Х
		(unoccupied)		
8	clay render	residential	20	Х
9	clay render	rescue	20	Х
		station		
10	clay panels	residential	10/50	—
11	clay panels	residential	10/50	—
12	clay panels	residential	10/50	_
13	clay panels	residential	10/50	—
14	clay panels	office	10/50	—
15	clay panels	residential	10/50	
16	mixed: adobe,	residential	10/50	_
	render, panels			
17	adobe	office	10/50	_

late winter 2013 within 17 houses in which unfired mineral material were extensively used (Figure (3)). Other than this criteria, the houses were selected randomly, i.e. they were not chosen according to risk factors such as a known high thorium content in the building material. The houses in the study comprised of traditional half-timbered residential and administrative buildings (Figure (4)), one traditional house with clay render and several modern houses with earthen building material such as adobes, clay render, and clay panels.



Figure (4): Photo of a traditional half-timbered (House no. 7). Such houses are common in most parts of Germany. Buildings with similar architecture can be found throughout northern Europe.

Concentrations of radon and thoron progeny

Figure (5) presents the average concentrations of 212 Pb in the houses in which UBPMs were applied during their exposure time. The concentrations ranged from 2 to 10 Bq/m . Applying published inhalation dose coefficients for 212 Pb (Bi et al., 2010) and assuming an exposure to the dwellers of 10 hours/day (occupancy factor of 40%) with an unattached fraction of 5% (Meisenberg and Tschiersch, 2011) these concentrations result in an annual inhalation doses of up to 4 mSv from 212 Pb alone.



Figure (5): Average concentration of ²¹²Pb measured with the UBPM over one to two months in late winter 2013. The measurement uncertainty (k = 1) is mainly caused by the uncertainty contribution of the calibration factor with an additional contribution from the number of measured CR39 tracks.

Radon and thoron concentrations

Thoron concentrations at a distance of 20 cm ranged from below the detection limit to 90 Bq/m (Figure (6)). The low thoron concentrations in the houses 1 and 4–6 cannot be correlated with the progeny concentrations. However in houses no. 1 and 6, relatively high progeny concentrations were found. Three explanations are possible: Firstly, the thoron concentration profile could decrease more rapidly with increasing distance so that no thoron reaches the measurement device at 20 cm from the wall. Secondly, the progeny concentration could be caused by thoron exhalation from other parts of the room or the building in which clay was used. Thirdly, the equilibrium factor between thoron and its progenies could be particularly high e.g. because of weak ventilation with the outdoor air.



Figure (6): Average radon and thoron concentrations in houses 1 to 9 at a distance of 20 cm from a wall. In the houses no. 1 and 5, the thoron concentration was below the detection limit.

At a distance of 10 cm from the clay walls, houses 7 and 10-17 showed slightly greater concentrations (Figure (7)). Conversely, the concentrations in the other houses were similar to those found in the houses shown in Figure 6. In all cases, concentration of up to 50 Bq/m could still be found in the middle of the rooms. This gives rise to the assumption that similar progeny concentrations can also be found in houses 1–6, 8, and 9. , However a precise statement cannot be given because of the lacking correlation between thoron and its progeny, which were mentioned above.

Another observation is that considerably different concentrations were measured in front of different types of clay walls. In the houses 11–13 and 15, clay is applied at a uniform thickness of 4 cm whereas it is 2 cm in house 14. However, the relatively low concentration of 24 Bq/m at 10 cm in house 15 was measured in front of a wall with a clay render of only 0.5 cm thickness applied to a structure of different material. Also the slopes of the concentration profiles differ from each other, e.g. with moderate concentrations at 10 cm and still considerable concentrations at 50 cm in house 15.



Figure (7): Minimum, average and maximum concentrations of thoron at distances of 10 and 50 cm from different walls and the concentration of radon in the ground floor in Bq/m in different rooms of the houses 7 and 11–17. For better clarity, the thoron concentrations are arranged around the 10 and 50 cm marks.

Conclusions

The measurements of thoron progenies under normal conditions show that increased concentrations with annual inhalation doses of a few mSv occur frequently in houses in which mud and clay is extensively used as a building material. The measurements of thoron gas in such houses suggest similar results. Thus, thoron should be considered as an issue for radiation protection in houses with earthen architecture.

The comparison of thoron and its progeny measurements confirm that a deduction of inhalation doses from gas measurements alone is generally not possible. In particular, the measurement of thoron at different distances indicates that there is no representative distance which can be used for a single-point thoron measurement, from which an average concentration can be deduced. However, it is suitable for a first assessment, if thoron is present at a high concentration.

The steep decrease of the thoron concentration from the wall to the middle of the room makes the need for measurement devices suitable for non-uniform concentrations necessary. In the design of such devices, especially for thoron passive measurement devices, a realistic thoron profile rather than a homogeneous concentration should be taken into account. These devices should also be calibrated and intercompared with these nonhomogeneous profiles in mind.

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