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Radon and thoron inhalation doses in dwellings with earthen architecture: Comparison of measurement methods

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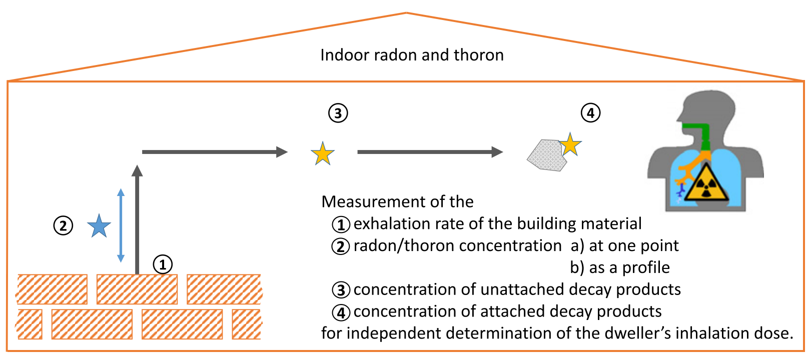
HIGHLIGHTS

- A significant contribution from thoron exposure to the inhalation dose was found.
- Thoron decay product measurements are necessary for reliable dose assessments.
- The unattached thoron decay products are relatively unimportant.
- Active/time-resolved or passive/integrating methods are precise and efficient.
- Advice on the proper measurement method in similar exposure situations is given.

GRAPHICAL ABSTRACT



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ABSTRACT

The radioactive noble gas radon (^{222}Rn) and its decay products have been considered a health risk in the indoor environment for many years because of their contribution to the radiation dose of the lungs. The radioisotope thoron (^{220}Rn) and its decay products came into focus of being a health risk only recently. The reason for this is its short half-life, so only building material can become a significant source for indoor thoron. In this study, dwellings with earthen architecture were investigated with different independent measurement techniques in order to determine appropriate methods for reliable dose assessment of the dwellers. While for radon dose assessment, radon gas measurement and the assumption of a common indoor equilibrium factor often are sufficient, thoron gas has proven to be an unreliable surrogate for a direct measurement of thoron decay products. Active/time-resolved but also passive/integrating measurements of the total concentration of thoron decay products demonstrated being precise and efficient methods for determining the exposure and inhalation dose from thoron and its decay products. Exhalation rate measurements are a useful method for a rough dose estimate only if the exhalation rate is homogeneous throughout the house. Before the construction of a building *in-vitro* exhalation rate measurements on the building material can yield information about the exposure that is to be expected. Determining the unattached fraction of radon decay products and even more of thoron decay products leads to only a slightly better precision; this confirms the relative unimportance of the unattached thoron

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decay products due to their low concentration. The results of this study thereby give advice on the proper measurement method in similar exposure situations.

1. Introduction

For many years the radioactive noble gas radon (^{222}Rn) and its progeny have been considered a health risk in the indoor environment because of their contribution to the radiation dose of the lungs and their potential for inducing lung cancer (WHO, 2009). In contrast, the isotope thoron (^{220}Rn) was object of only a few exposure surveys in the past (e.g. Stranden, 1980; Steinhäusler, 1996; Chen et al., 2011). Because of its short half-life of 55.6 s it is unable to penetrate reasonably airtight building foundations. However, thoron gained attention after increased concentrations were found in cave dwellings dug into clay soil and houses with earthen architecture in China and India (Wiegand et al., 2000; Sreenath Reddy et al., 2004; Shang et al., 2005; Yamada et al., 2006; Shang et al., 2008; Ramola et al., 2012; Mishra et al., 2015; Omori et al., 2016). In the following years thoron was identified as a significant contributor to the dose also in houses in which unfired earthen material was used e.g. in Germany, Hungary and Japan (Gierl et al., 2014; Szabó et al., 2014; Yonehara et al., 2005). There, the building material is the source of thoron and the gas can enter the indoor air without any diffusion barrier. Therefore, monitoring strategies for the exposure both to radon and to thoron are required for the health protection of the inhabitants of such houses. The aim of such monitoring must be the reliable assessment of the contributions to the equivalent lung dose and the effective dose as the quantities of radiation protection considerations.

The largest contribution to the inhalation dose is caused by the progeny and not by the gases for both decay chains, radon and thoron. Exposure to radon and its progeny is, however, usually monitored by measurements of the gas. This is possible because of the equilibrium factor being within a narrow range of 40 ± 12% (UNSCEAR, 2006). The applicability of thoron gas measurements for dose assessments is considered as limited because of the inhomogeneous concentration, the variant equilibrium factor and the variant unattached fraction (UNSCEAR, 2006). Furthermore, suggested mitigation measures mainly refer to the decay products (Wang et al., 2011; Tschiersch et al., 2012; Khandare et al., 2016). However, measurements of thoron gas are commonly performed in many studies and easy to conduct because of the availability of passive thoron measurement devices whereas passive measurement devices for thoron progeny are rare (Mishra et al., 2014; Zhuo and Iida, 2000). Additionally, for the planning of houses it is desirable that civil engineers assess thoron inhalation doses of the future dwellers from measurements of the exhalation rate of the building material, which can be tested *in-vitro* before the construction of the house.

To test the suitability of different measurement techniques, a variety of measurement devices was exposed in two dwellings in southern Germany where increased thoron concentrations were found. Measurement devices of Bhabha Atomic Research Centre (BARC) and Helmholtz Zentrum München (HMGU) employing measurement techniques for the gas and for the decay products, with active and passive sampling, active/time-resolved and passive/integrating measurement, and for short-time and long-time measurements were applied. Besides, the exhalation rate of the building material was measured in-situ. The results of each single measurement technique were used on their own, i.e. without regard to the results of the other techniques, to calculate the inhalation dose of the dwellers.

The aim of the study was to identify appropriate measurement techniques that allow assessing reliably the exposure of the dwellers of radon and thoron prone homes. Irrespective of the inherent problems of each method, the inhalation dose was determined autonomously as end point of each measurement technique. On the base of the determined dose, a decision on the method of choice can be taken for similar exposure situations, independent from the dwellings tested as examples in this study. Finally, the inter-comparison performed in the present study shall establish a sound protocol for inhalation dose assessment of thoron.

2. Materials and methods

2.1. Measurement objects

The first house in which the measurements were performed is a half-timbered house from the 18th century in a village in southern Germany (house A). The ground floor is built of natural stones, whereas the first floor and the attic are timber-framed. The house does not feature a basement. In one part of the first floor, the panels between the timber beams of the inner walls are filled with unfired mud, whereas the other part of the upper floor was renovated and features a different filling material. Gamma spectroscopy of the applied mud showed specific activities of ^{226}Ra, ^{232}Th, and ^{40}K of 37.5 ± 3.4 Bq/kg, 60 ± 14 Bq/kg, and 727 ± 93 Bq/kg, respectively. This is well in the range of typical activity concentrations for clay bricks in the EU (50 Bq/kg ^{226}Ra, 50 Bq/kg ^{232}Th, and 670 Bq/kg ^{40}K; European Commission, 1999). Thus, the mud is neither excessively rich in ^{226}Ra nor in ^{232}Th. The house was uninhabited at the time of investigation, just after renovation before further use. The render and wallpaper have been removed from the walls in several rooms. Passive measurements were carried out in the different parts of the house from September to December 2012. Additionally, active measurements were performed during two weeks within that period. Rooms 1–4 are situated in the upper floor in the older part where unfired mud is present, room 5 is located in the renovated part of the upper floor and room 6 is located on the ground floor.

The second investigated house is a modern one-family dwelling with basement, ground-floor and one upper floor in a small town in the south of Germany (house B). It is erected from natural stones and bricks. The indoor walls are rendered with clay at thicknesses between 1 and 3 cm. The applied clay was purchased from industrial production and its quality with regards to low radionuclide content was assured by the producer. To avoid damaging of the newly built house, no sample of the clay plaster for gamma spectroscopy was taken. The house was constructed according to energy saving aspects. Thus, it is equipped with a ventilation system with three air exchange rates (ca. 0.12 h^{-1}, 0.25 h^{-1}, 0.4 h^{-1}), which are manually adjusted according to the number of inhabitants and their activities. Inlet and outlet of the system are located in the open staircase, with the inlet being in the basement and the outlet in the first floor. The house is heated by a wall heating system, i.e. several walls in each floor contain pipes for hot water circulation. During the measurements, which were performed from December 2013 to March 2014 and from May to June 2014, the house was unoccupied. During other periods it is used as holiday and week-end house.

Measurements in that house were performed in the ground floor living room, which is open to the ventilated staircase and to several adjacent rooms, and in a 31 m^3 unventilated guestroom in the basement, which was separated from the staircase by a closed door. The air exchange rate between the guestroom and the staircase was determined as 0.07 h^{-1} using a puff of CO_2 as trace gas.

2.2. Outline of the applied methods

Airborne radon and thoron gases were measured both with actively sampling, time-resolving measurement devices (Section 2.3) and with passive, integrating devices (Sections 2.4 and 2.5). To trace the concentration gradient of thoron measurements, time-resolving as well as integrating devices were deployed at different positions within the rooms. For radon and thoron progeny measurement, both, actively sampling, time-resolving devices (Section 2.6) as well as passive, integrating devices (Section 2.7) were applied. The latter allow discriminating between the unattached and the total concentration (HMGU and BARC devices) or between the attached and the total concentration (BARC devices). In addition, spot measurements of the radon and thoron progeny concentration were performed with actively sampling and passively measuring devices (Section 2.8). The exhalation of thoron from the building material was measured in situ in an accumulation chamber (Section 2.9).

2.3. Active radon and thoron measurements (HMGU)

For time-resolved measurements of radon concentrations, an electronic measurement device Alphaguard (Saphymo, Germany) was used. This device uses the diffusion of ambient air into an ionization chamber to count radon decays and thus to calculate the concentration. It was calibrated at PTB (German metrology agency) with traceability to a primary standard. Additionally, newly developed radon exposure meters (prototypes of AlphaE, Saphymo, Germany) were used (Irlinger et al., 2014).

Time-resolved measurements of thoron concentrations were performed using a RAD7 (Durridge, USA). This device samples air at a flow rate of about 1 l/min. Inside its measurement chamber the originating, positively charged decay products are deposited via an electric field onto a silicon alpha detector with subsequent spectrometer. Their decay on the detector is used for the calculation of the thoron concentration in air. The device was calibrated at PTB; the calibration is traceable. Because of the active sampling, thoron within a certain distance can reach the inlet. On average, this distance can be estimated from the mean lifetime of thoron and the air flow rate as 6 cm.

2.4. Passive radon and thoron measurements (HMGU)

Passive, integrating measurements of the concentrations of radon and thoron were performed with CR39 solid-state nuclear-track detectors (SSNTD). In these instruments, a 1 cm^2 piece of CR39 is placed at the bottom of a 3.6 cm tall plastic cup. Two different types of instruments were used: For measurements of radon and thoron the cups are closed with a screw cap with several holes, which allows both gases to enter the box. The holes are covered with a membrane filter and conductive foam to prevent progeny from entering the cup. For measurements of only radon solid caps were used. Radon can still enter the cups by diffusion along the cap thread whereas this is not possible for the short-lived thoron. Thoron concentrations can be measured with these instruments by placing a cup for radon and thoron together with at least one for radon in the same room.

The devices were calibrated to the concentration at the surface of the cap using a RAD7 active radon and thoron measurement device in a 1 m^3 calibration chamber. Frequent participation in intercomparison measurements assured the quality of the measurement results (e.g. Janik et al., 2010).

Thoron measurement devices were deployed at five to eight different distances from a wall in order to trace the thoron concentration profile. The devices for each profile were positioned in close proximity to each other but not in a straight line one after the other to avoid shadowing of the more distant detectors. All detectors were positioned at least 50 cm from other walls, the floor and the ceiling. Cardboard holders, which were fixed at the wall, or wooden stands were used to set up the detectors at their position.

The measured concentration $c(d)$ can be described as an exponential function of the distance d from the wall with an additional homogeneous contribution c_a:

$$c(d) = c_a + c_b \cdot \exp(-d/d_0)$$

with c_b as the additional concentration directly at a thoron-exhaling surface and d_0 as the characteristic distance for the decrease of the concentration.

The average concentration $\bar{c}_{Tn}$ of thoron within the room was calculated using the integral of the thoron concentration profile throughout the room:

$$\bar{c}_{Tn} = c_a + \frac{S}{V} \cdot \int_0^{\text{middle of room}} c_b \exp(-d/d_0)\,\mathrm{d}d$$

with S as the surface area of the clay surfaces in the room (other surface were assumed as not thoron-exhaling and thus were not taken into account) and V as the volume of the room (Meisenberg and Tschiersch, 2010).

2.5. Passive radon and thoron measurements (BARC)

Passive radon and thoron gas measurements were carried out using pin-hole dosimeters. They consist of two chambers separated by a plate, which is airtight except of four pinholes in it (Sahoo et al., 2013). The first chamber allows both radon and thoron gas to pass through to the nuclear track detector by natural diffusion, whereas the pinholes selectively allow only radon to pass to the second chamber. As detectors LR-115 SSNTDs were used. The pin-hole dosimeters were deployed in line at five different distances from a wall.

2.6. Active decay product measurements with active sampling (HMGU)

A working level monitor BWLM-PLUS-2S (Tracerlab, Germany) was used for time-resolved progeny measurements. This device samples aerosol particles at an air flow rate of 100 to 150 l/h onto sampling substrates and measures the activity of sampled alpha-particle emitters online with a solid state alpha detector with subsequent spectrometer. Concentrations of radon and thoron progeny in the air are calculated from the measured activities of the alpha-particle emitting decay products. The device comprises of two sampling and measurement units: A membrane filter is used as the sampling substrate for aerosol particles of all sizes whereas a wire-mesh is used for aerosol particles in the size range of unattached radon and thoron progeny. The device was calibrated at the German Federal Office for Radiation Protection, Berlin for the total concentration of radon progeny; this calibration is traceable. There, a calibration for unattached radon progeny was also performed. This calibration is not traceable because no standardized definition of the size range of unattached progeny exists. The counting efficiencies for radon progeny are applied for the measurement of total and unattached thoron progeny.

2.7. Passive decay product measurements with passive sampling (BARC)

Passive time-integrated measurements of radon and thoron progeny were carried out with so-called Direct Radon and Thoron Progeny Sensors (DRPS and DTPS). These detectors are absorber-mounted LR-115 SSNTDs, which record the alpha particles emitted from the deposited progeny atoms (Mishra and Mayya, 2008). The detectors make use of the deposition of progeny atoms by natural diffusion. Alpha particles of different energies are discriminated using aluminized polyethylene foils of different thickness. After exposure, the LR-115 detectors are chemically processed in 2.5 N NaOH at 60 °C for 90 min followed by alpha particle track counting using a spark-counter. The number of tracks registered in the SSNTD is used to calculate the time-averaged equilibrium equivalent concentration (EEC) of radon and thoron progeny using appropriate sensitivity factors (Mishra et al., 2014). The thoron progeny interference in the case of DRPS is subtracted to estimate the EEC of radon progeny (Mishra et al., 2009). The DRPS/DTPS were calibrated against grab filter-paper sampling and alpha-counting in controlled conditions as well as in real indoor environments.

For passive measurement of attached progeny only, wire-mesh capped deposition sensors are used (Mayya et al., 2010). They consist of DRPS/DTPS detectors capped by a 200 mesh wire-screen. These detectors have been calibrated against integrated sampling with deposition on a wire-screen followed by alpha-particle spectrometry.

2.8. Passive decay product measurements with active sampling (BARC)

For passive short-time measurements of radon and thoron progeny with active sampling so-called integrated samplers were applied (Mishra et al., 2009). Integrated samplers comprise of a wire-mesh and a filter-paper arranged in an array. Two LR-115 SSNTD progeny sensors are placed in such a way that one is facing the wire-mesh and the other the filter paper. The sensor facing the wire-mesh records the alpha particles emitted from the unattached progeny atoms deposited on the wire-mesh; the sensor facing the filter paper records the attached fraction of progeny concentration (Mishra et al., 2010). These detectors are calibrated against wire-mesh and filter-paper sampling and alpha-particle spectrometry. Sampling with these devices was performed for 3 to 4 h.

2.9. Exhalation rate measurements (BARC)

The measurement of thoron exhalation rates from walls was carried out using the so-called accumulator technique (Schery et al., 1989; Mayya, 2004). The accumulator had a diameter of 7 cm and a height of 8 cm and was fixed in an air-tight way to the wall to prevent leakage of thoron. The measurement of the accumulated thoron concentration was carried out by pumping air from the accumulator into a Pylon scintillation counter in closed loop. A delay of 8 to 10 min was allowed for the thoron concentration in the closed loop to reach steady state. The detector was calibrated using a standard scintillation cell provided by the manufacturer. The thoron exhalation rate E_{Tn} was calculated from the thoron concentration c in steady state as follows:

$$E_{\mathrm{Tn}} = \frac{cV\lambda}{A}$$

where λ is the thoron decay constant, $V = 6.5 \times 10^{-4}\ \mathrm{m}^3$ is the total volume of the closed loop and A is the surface area of the wall enclosed by the accumulator.

2.10. Dose calculation

Comparing the different applied measurement techniques which provide different measured quantities such as airborne concentrations of the gas, of the decay products or the exhalation rate, a translation into one joint quantity is necessary. As the health impact on the dwellers in radon and thoron prone homes is of concern, the inhalation dose is taken as the quantity to be compared. Therefore all results of the individual methods were transferred independently into an inhalation dose.

The inhalation dose of adult dwellers from exposure to radon and thoron progeny was calculated as effective dose (averaged over doses for males and females) from the concentrations of the unattached and attached progeny. The effective dose resulting from the inhaled short-lived decay products is expressed in terms of a dose conversion factor, which is given as effective dose per unit potential alpha exposure. The calculation of these dose conversion factors is based on biokinetic and dosimetric models as it is suggested by the ICRP (2010). They consider the size of the inhaled progeny and the inhaled main radionuclides which are ^{218}Po, ^{214}Pb and ^{214}Bi for radon and ^{212}Pb and ^{212}Bi for thoron, respectively, and their decay chains in the body. The dose conversion factors published by Brudecki et al. (2014) for radon progeny and by Bi et al. (2010) for thoron progeny were applied. An occupancy factor of the dwellings of 40% (i.e. 10 h per day) and a breathing rate of 0.78 m^3/h were assumed (ICRP, 2002).

The measured concentrations were used for measurement methods giving immediate information about the progeny concentrations in their unattached and attached state (i.e. active progeny measurements with a WLM and passive progeny measurements with DRPS/DTPS). For measurements of the gross concentration of radon and thoron progeny, of radon and thoron concentrations and thoron exhalation rates the following assumptions were made in order to calculate the concentration of unattached and attached radon and thoron progeny:

- The unattached fraction was assumed to be 13 ± 9% for radon progeny (UNSCEAR, 2006) and as 3.0 ± 1.5% (compiled from UNSCEAR, 2006 and Meisenberg and Tschiersch, 2011) for thoron progeny.[4]

- The equilibrium factor between the progeny and the gases was assumed to be 40 ± 12% (UNSCEAR, 2006) for radon and as 3.5 ± 2.0% (compiled from UNSCEAR, 2006 and Meisenberg and Tschiersch, 2012) for thoron. For this purpose, the equilibrium factor F of the thoron decay chain is defined as the equilibrium-equivalent concentration (EEC) of thoron progeny relative to the average concentration of thoron within the room:

$$F = \frac{\mathrm{EEC}}{\bar{c}_{\mathrm{Tn}}}$$

With these assumptions, average thoron concentrations were calculated from thoron exhalation rate measurements. Unattached and attached radon and thoron progeny concentrations were calculated from radon and average thoron concentrations.

Because of a lack of further information, equilibrium between the decay products of each decay chain was assumed. Since this applies to the dose calculations for all investigated measurement techniques, no bias between the results is introduced by that.

3. Results and discussion

3.1. Radon and thoron concentrations

Thoron concentrations of several 100 Bq/m^3 close to the clay walls were found, both in house A and in house B. This is significantly more than what was measured in previous studies in which the focus was not on buildings with earthen architecture (Stranden, 1980; Steinhäusler, 1996).

In house A the thoron concentrations featured clear profiles with a decrease at larger distances (Fig. 1). There, exponential functions could be fitted to the profile to calculate the average thoron concentration in each room. A comparative measurement series with the actively sampling device RAD7 in front of the same position yielded concentrations slightly larger but within the confidence interval (Fig. 2). Walls where the clay was replaced by a different building material showed distinctly smaller concentrations.

In house B strong variations of the concentrations at different distances were measured with passive dosimeters (Fig. 3). Therefore, an evaluation of the average concentration was not possible here. This apparently different behaviour of thoron might be attributed to air turbulence in front of the wall where the measurements were performed. This turbulence may have been caused by the wall heating system which exists inside the wall. In contrast, a passive measurement of the thoron concentration profile with pin-hole dosimeters showed a distinct decrease of the concentration with increasing distance. It was

[4] All uncertainties are presented with a coverage factor of k = 2.

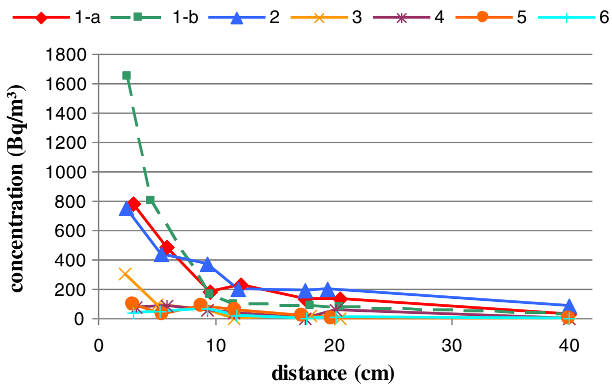

Fig. 1. Thoron concentration profiles for different rooms of house A. The concentrations were measured with passive devices of HMGU over 74 days. The uncertainty is in the order of 10% for 100 Bq/m^3. Labels 1-a and 1-b denote two walls within the same room 1. Room 5 in the renovated part of the upper floor and room 6 in the ground floor do not contain any clay surfaces.

used for an estimation of the average thoron concentration in the room and thus also for a dose assessment. This measurement series was performed at another part of the same wall, but without wall heating system. The influence of strong air turbulence on the thoron concentration profile, leading to a homogenization of the thoron concentrations, whereas static air causes a distinct profile, was shown by Ma et al. (1997).

Effective doses for the inhalation of radon, thoron and their progeny were calculated from the profiles found in house A. In the rooms where no clay was present anymore, upper limits of 0.62 mSv per year or less were calculated under the assumptions given above. In house B the thoron measurements could not be used for a calculation of the progeny concentrations and were therefore not usable for a dose estimation either.

3.2. Decay product concentrations

In house A significant concentrations of thoron progeny were found also in rooms 5 (room in a part of the first floor where the clay was replaced) and 6 (room in the ground floor, which is completely built of natural stones). They can be considered to originate from transport within the house from the rooms with clay walls to the other rooms rather than from thoron exhaled in the respective room. Consequently, higher thoron progeny concentrations were found in the room in the ground floor when the door to the room was open; in contrast, the radon concentration at that time was lower because of stronger ventilation of the room.

In house B the progeny concentration was measured in the ground floor at three different ventilation rates. A decrease of the total concentration of thoron progeny (Fig. 4) and of radon progeny at increased ventilation could be observed: to 85% and 95% of the value during low ventilation at medium ventilation rate and to 35% and 40% at strong ventilation rate for radon and thoron progeny, respectively. At the same time no influence on the unattached concentrations was found, leading to an increase of the unattached fraction of radon and thoron progeny from 6.2% and 2.4% at low ventilation to 8.7% and 3.0% at medium ventilation and to 22% and 7.6% at strong ventilation rate. Thus, the measured unattached fractions of radon were within the range reported by UNSCEAR (2006) whereas those of thoron were significantly larger during strong ventilation. Unattached fractions of thoron progeny in this range, and at values up to 15 to 20% at low aerosol concentrations, were found in other studies (Meisenberg and Tschiersch, 2011; Singh et al., 2016).

A comparison of progeny and gas measurements in the guestroom of house B yielded equilibrium factors of the radon decay chain, i.e. the ratio of the equilibrium-equivalent concentration of the radon progeny to the concentration of the radon gas, of 80 ± 31% (measured with passive gas and active progeny devices by HMGU) and of 117 ± 40% (measured with passive gas and progeny devices by BARC). These values are significantly larger than 40 ± 12%, which is assumed by UNSCEAR (2006). Both values show that in this room the decay products are close to equilibrium with the gas, the reason for which can be the low air exchange rate in that room. The calculational best estimate of 117% might not reflect the true value of the equilibrium factor as for radon (in contrast to thoron) significantly higher concentrations of the decay products than of the gas are not realistic. The true value might be at the lower end of the confidence interval.

3.3. Exhalation rate measurements

In-situ measurements of the thoron exhalation rate were performed in several rooms in house A. Those walls in rooms 1 and 2 with earthen building material showed exhalation rates between 14 and 25 Bq/(m^2 s). Walls in room 3, which are in the same part of the building

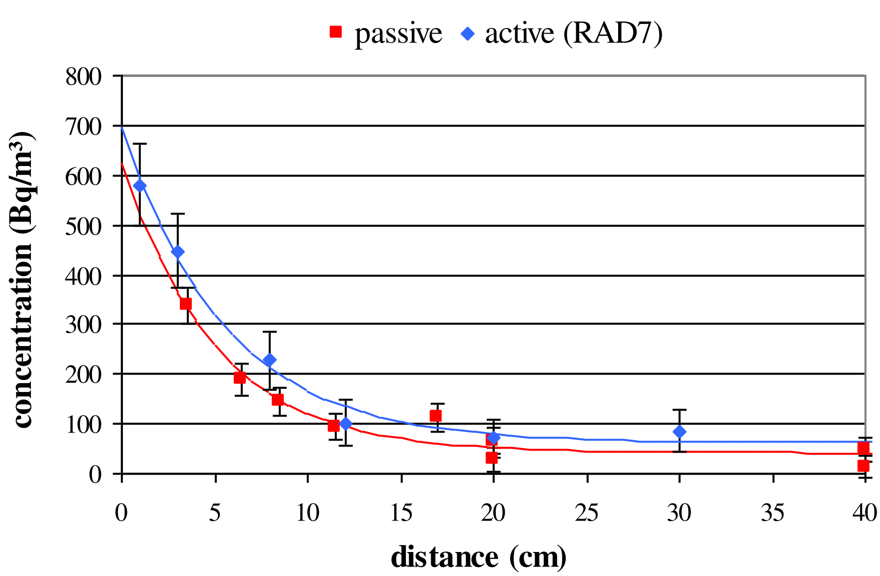

Fig. 2. Comparison of thoron concentrations measured with passive devices from HMGU and an actively sampling and measuring device RAD7. The measurements were performed in front of the same position of clay wall 1-a in room 1 of house A but at different times in the experimental period.

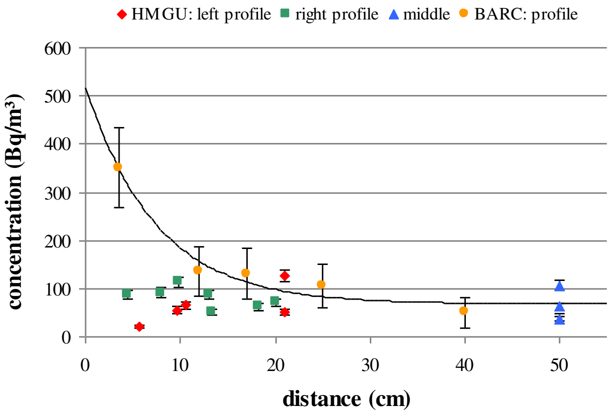

Fig. 3. Thoron concentration profiles in the guestroom of house B in front of three sections of one wall: Two profiles were measured with passive dosimeters by HMGU, measurement results at 50 cm can be attributed to both profiles. In addition, one profile was measured with pin-hole dosimeters by BARC; for this profile an exponential fit is also shown.

as the rooms 1 and 2 but with no earthen material visible at the wall surfaces, exhaled thoron at rates between 1.0 and 4.0 Bq/(m^2 s). Probably older clay had been covered with other material as a superficial render. At the walls in room 5, which is situated in the renovated part of the upper floor, exhalation rates below the detection limit of 0.5 Bq/(m^2 s) were determined.

4. Conclusions

Significant contributions of radon and thoron to the radiation dose were determined for both investigated exemplary houses of earthen architecture although the specific activities of ^{226}Ra and ^{232}Th in the building material were not particularly high. For thoron, whose most important source is the building material, this confirms the prominence of earthen building material as a source of exposure.

In Table 1 the independently calculated inhalation dose is presented for each room of house A and for each applied measurement technique. In Table 2 the same is given for house B. The same assumptions about equilibrium factor, unattached fraction, and other dose affecting parameters were made where necessary. This allows an independent comparison of the different applied methods and techniques referring only to the resulting dose of the dwellers.

In house A direct progeny measurements with actively sampling devices, which are supposed to yield the most reliable results, result in annual dose contributions of 1 to 2 mSv from radon progeny in the upper floor and 3 to 6 mSv in the ground floor and additional 1 to 2 mSv from thoron progeny with slightly smaller values in the adjacent rooms without thoron-exhaling building material. In house B about 1 to 5 mSv from radon progeny and 1 to 2 mSv from thoron progeny were calculated for the living room under usual living conditions and 12 and 9 mSv, respectively, for the guestroom under the condition of low ventilation.

In general, the different applied measurement devices of HMGU and BARC yielded consistent results for the respective measured quantity. However, two exceptions were observed. First, there was a discrepancy of the measured thoron progeny concentration in the guestroom of house B: There the actively measuring working level monitor of HMGU yielded 15.8 ± 6.5 Bq/m^3 (annual dose of 7.8 mSv), whereas several passive DTPS of BARC showed only 3.6 Bq/m^3 (1. 8mSv) on average with a double standard deviation of 1.5 Bq/m^3. In contrast, the radon progeny results of the two methods agree well. A possible reason for this discrepancy can be a smaller deposition velocity of the thoron progeny than implemented in the calibration of the passive device. The influence of environmental factors on the deposition was discussed in Li et al. (2012). Thus a calibration that is specific for the respective exposure situation should be performed. Second, the inhalation doses calculated from exhalation rate measurements are strongly overestimated in the rooms with thoron exhalation. This indicates a significant transport of thoron progeny from the rooms with thoron sources to the other rooms in the house. In the other rooms the sensitivity of this method is too small to reliably assess the inhalation dose.

From thoron gas measurements, even from tracing the thoron profile in front of a wall with several measurements at different positions, no reliable statement could be made about the progeny concentration in several rooms. Besides a variant equilibrium factor, the following reasons can be supposed:

- The thoron profiles at different positions within one room might vary by their characteristic distance because of variable (turbulent) mixing of the indoor air.
- The concentration directly at the wall might vary because of locally different exhalation rates of the building material.
- The concentration of indoor thoron progeny might be influenced not only by thoron exhalation in the respective room but also by thoron exhalation in other rooms of the building (as in house A, rooms 5 and 6) and subsequent advection of the decay products.

This ambiguity cannot be resolved by application of a locally defined equilibrium factor, i.e. a ratio of the progeny concentration to the local thoron concentration at a certain position in the room. Such local equilibrium factors vary even stronger than the globally defined equilibrium factor applied in this study (Chen et al., 2012; Prasad et al., 2016; Singh et al., 2016).

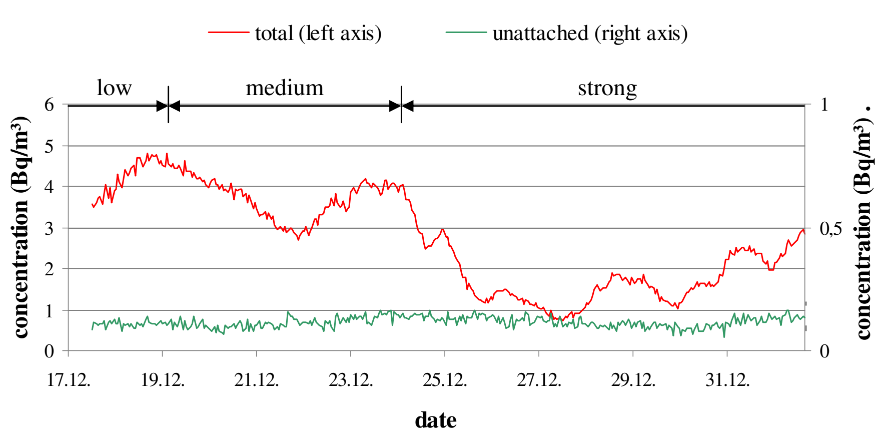

Fig. 4. Temporal variation of total and unattached thoron progeny concentration (EEC) in the living room of house B. The three periods of different ventilation rates are marked.

Table 1
Calculation of annual inhalation doses for house A from exposure to radon, thoron and their progeny by application of the respective measurement technique and assumptions about equilibrium factor and unattached fraction where necessary.

House A Room no.	Measurement technique	Evaluation protocol	Radon dose (mSv/a)	Thoron dose (mSv/a)
1	Active progeny measurement (HMGU)	Total, full time	1.56 ± 0.68	1.19 ± 0.24
		Total, working time	1.11 ± 0.52	1.13 ± 0.46
		Total, free time	1.31 ± 0.60	1.22 ± 0.50
	Active progeny sampling, passive measurement (BARC)	Total	–	1.58 ± 0.24
	Passive progeny sampling and measurement (BARC)	Unattached/attached progeny separately	2.54 ± 0.42	1.36 ± 0.30
		Total	3.15 ± 0.78	1.35 ± 0.10
	Active gas measurement (HMGU)	For thoron: complete profile	<2.2	1.9 ± 1.2
	Passive gas measurement (HMGU)	For thoron: complete profile	2.16 ± 0.88	1.25 ± 0.92
	Exhalation rate (BARC)		–	12.9 ± 7.6
2	Active progeny sampling, passive measurement (BARC)	Total	–	1.43 ± 0.12
	Passive progeny sampling and measurement (BARC)	Unattached/attached progeny separately	2.76 ± 0.62	1.41 ± 0.26
		Total	3.39 ± 0.82	1.42 ± 0.20
	Passive gas measurement (HMGU)	For thoron: complete profile	2.6 ± 1.1	6.2 ± 3.3
	Exhalation rate (BARC)		–	11.5 ± 6.9
3	Active progeny sampling, passive measurement (BARC)	Total	–	1.50 ± 0.66
	Passive gas measurement (HMGU)	For thoron: complete profile	4.5 ± 1.7	<1.5
	Exhalation rate (BARC)		–	<5.1
4	Passive gas measurement (HMGU)	For thoron: complete profile	5.8 ± 1.1	<1.5
5	Active progeny measurement (HMGU)	Unattached/attached progeny separately, full time	2.17 ± 0.68	0.76 ± 0.28
		Total, full time	2.6 ± 1.2	0.79 ± 0.32
	Passive progeny sampling and measurement (BARC)	Unattached/attached progeny separately	1.99 ± 0.60	1.21 ± 0.30
		Total	2.55 ± 0.90	1.13 ± 0.14
	Passive gas measurement (HMGU)	For thoron: complete profile	3.7 ± 1.5	<0.62
	Exhalation rate (BARC)		–	<1.4
6	Active progeny measurement (HMGU)	Unattached/attached progeny separately, full time	3.1 ± 1.0 (door open)	1.34 ± 0.50 (door open)
			4.5 ± 1.5 (door ajar)	0.82 ± 0.15 (door ajar)
		Working time	4.7 ± 1.6 (door ajar)	0.80 ± 0.30 (door ajar)
		Free time	4.4 ± 1.5 (door ajar)	0.83 ± 0.30 (door ajar)
		Total, full time	4.0 ± 1.8 (door open)	1.43 ± 0.58 (door open)
			5.6 ± 2.3 (door ajar)	1.04 ± 0.42 (door ajar)
	Passive gas measurement (HMGU)	For thoron: complete profile	4.4 ± 1.7	<0.38

Thus, measurement of thoron, at different positions (but even worse at only one position in a room), has proven to be an unreliable surrogate for a direct measurement of thoron progeny.

Time-resolved measurement of radon and thoron progeny, was found to be not necessary in the investigated houses as long as the indoor conditions such as ventilation are not dependent on the occupancy. Determining the unattached fraction of radon decay products and even more of thoron decay products causes only a slightly better precision; this confirms the relative unimportance of the unattached thoron decay products due to their low concentration.

Therefore, active/time-resolved but also passive/integrating measurements of the total concentration of thoron progeny seem to be a both precise and efficient method for determining the exposure and inhalation dose from thoron and its progeny. These methods should be applied for the dose assessment in existing houses and thus also for the related justification and planning of mitigation measures.

Exhalation rate measurements might be a useful method for a rough dose estimate only if the exhalation rate is homogeneous throughout the house. However, *in-vitro* exhalation rate measurements on the building material can yield information about the range of exposure to be expected before the construction of a building. They are therefore an important method for civil engineers for the selection of building materials and the planning of necessary mitigation measures that must be installed during the erection of the building.

The mitigating influence of ventilation both on the thoron and on the radon exposure was demonstrated. Ventilation is advised to reduce a variety of airborne indoor pollutants. Therefore it is not only an effective but also an appropriate measure for mitigation. Thus, buildings with

Table 2
Calculation of annual inhalation doses for house B from exposure to radon, thoron and their progeny by application of the respective measurement technique and assumptions about equilibrium factor and unattached fraction where necessary.

House B Room use	Measurement technique	Evaluation protocol	Radon dose (mSv/a)	Thoron dose (mSv/a)
Living room	Active progeny measurement (HMGU)	Unattached/attached progeny separately, full time	3.89 ± 0.65 (vent. low)	1.99 ± 0.72 (vent. low)
			3.2 ± 1.1 (medium)	1.86 ± 0.66 (medium)
			1.64 ± 0.52 (strong)	1.05 ± 0.37 (strong)
		Total, full time	4.7 ± 1.9 (vent. low)	2.02 ± 0.82 (vent. low)
			3.7 ± 1.7 (medium)	1.86 ± 0.76 (medium)
			1.58 ± 0.74 (strong)	0.92 ± 0.38 (strong)
Guest room	Active progeny measurement (HMGU)	Unattached/attached progeny separately, full time	12.2 ± 1.9	8.8 ± 2.8
		Working time	11.8 ± 1.9	8.9 ± 3.0
		Free time	12.4 ± 3.8	8.7 ± 2.8
		Total, full time	12.3 ± 3.7	7.8 ± 3.2
	Passive progeny sampling and measurement (BARC)	Unattached/attached progeny separately	–	2.4 ± 1.5
		Total	18.9 ± 7.5	1.76 ± 0.76
	Passive gas measurement (HMGU)	For thoron: complete profile	5.8 ± 2.3	Not possible
	Passive gas measurement (BARC)	For thoron: complete profile	6.5 ± 2.6	2.0 ± 1.5

earthen architecture that have a reduced ventilation e.g. for energy conservation should be monitored for thoron exposure in particular whereas in such buildings with strong ventilation, which are common in many parts of the world, lower exposures can be expected.

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