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Deposition and spatial variation of thoron decay products in a thoron experimental house using the Direct Thoron Progeny Sensors

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Abstract
Experiments have been carried out using the deposition-based Direct Thoron Progeny Sensors (DTPS) in a thoron experimental house. The objective was to study the thoron decay product characteristics such as the deposition velocities, spatial variability and dependence on aerosol particle concentrations. Since the deposition velocity is an important characteristic in the calibration of the DTPS, it is very important to study its dependence on aerosol concentration in a controlled environment. At low aerosol concentration (1500 particles/cm^3) the mean effective deposition velocity was measured to be 0.159 ± 0.045 m h^{-1}; at high aerosol concentration (30 000 particles/cm^3) it decreased to 0.079 ± 0.009 m h^{-1}. The deposition velocity for the attached fraction of the thoron decay products did not change with increasing aerosol concentration, showing measurement results of 0.048 ± 0.005 m h^{-1} and 0.043 ± 0.014 m h^{-1}, respectively. At low particle concentration, the effective deposition velocity showed large scattering within the room at different distances from center. The attached fraction deposition velocity remained uniform at different distances from the wall. The measurements in the thoron

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experimental house can be used as a sensitivity test of the DTPS in an indoor environment with changing aerosol concentration. The uniform spatial distribution of thoron decay products was confirmed within the experimental house. This indicates that direct measurement of thoron decay product concentration should be carried out instead of inferring it from thoron gas concentration, which is very inhomogeneous within the experimental house.

Keywords: ^{220}Rn decay products, thoron measurement, deposition velocity, spatial homogeneity

Introduction

Study of the behaviour of ^{222}Rn (radon) and ^{220}Rn (thoron) decay products in indoor air is important for assessing the natural background radiation exposures received by general populations through the inhalation route. The decay products of ^{222}Rn and ^{220}Rn are basically the radioisotopes of polonium, bismuth and lead. These are initially formed as positively charged atoms due to sustained recoil, followed by a cauterization process with trace gases and vapour present in the air to form an unattached fraction (0.5–5 nm). Further, these unattached decay product atoms undergo random motion and stick to available aerosol particles, thus forming an attached fraction of decay product concentration. The exposure and resulting inhalation dose are dependent on the airborne concentrations of these two fractions which have different dose coefficients (Bi *et al* 2010, Brudecki *et al* 2014).

The time dependence of airborne decay product concentrations, both unattached and attached, within an indoor environment is governed by the balance between their generation and removal processes (Jacobi 1972). So, the unattached fraction indoors can either attach to an aerosol, deposit on any available surface, or be carried outdoors by ventilation, or undergo radioactive decay. Similarly, the attached fraction can also detach from the host aerosol, undergo surface deposition, or may be transported outdoors, or undergo radioactive decay (Porstendörfer *et al* 2000). These are the basic processes influencing the indoor activity balance of decay product atoms (Meisenberg and Tschiersch 2011). Among the several aspects of decay product behaviour, deposition on surfaces is of special significance because of its prominent role in activity removal and the consequent occurrence of progeny disequilibrium with the parent gases. Deposition and ventilation are two major mechanisms of removal of the decay products from indoor air. Unlike the removal process by ventilation, which depends essentially on the air exchange rate in a given room, removal by deposition depends on the activity size distribution and the structure of turbulence at the air–surface interface. The activity size distribution, brought about by the attachment process, is governed by the concentration and distribution of indoor aerosols. The application of a standard Jacobi model (Jacobi 1972, Meisenberg and Tschiersch 2010) to indoor progeny dynamics shows that the proportion of fine fraction, which contributes to deposition to a significant extent, depends upon the ventilation rate itself. Besides the significance for protection against airborne radionuclides themselves, radon and thoron decay products provide excellent surrogates for understanding nano-particle deposition rates because of their easy measurability. In this regard, deposition-based progeny sensing techniques are being increasingly explored as dosimetric alternatives in indoor air, and deposition velocity is a crucial parameter in its calibration (Meisenberg *et al* 2017, Omori *et al* 2017). For these reasons it is important to arrive at an understanding of progeny deposition rates.

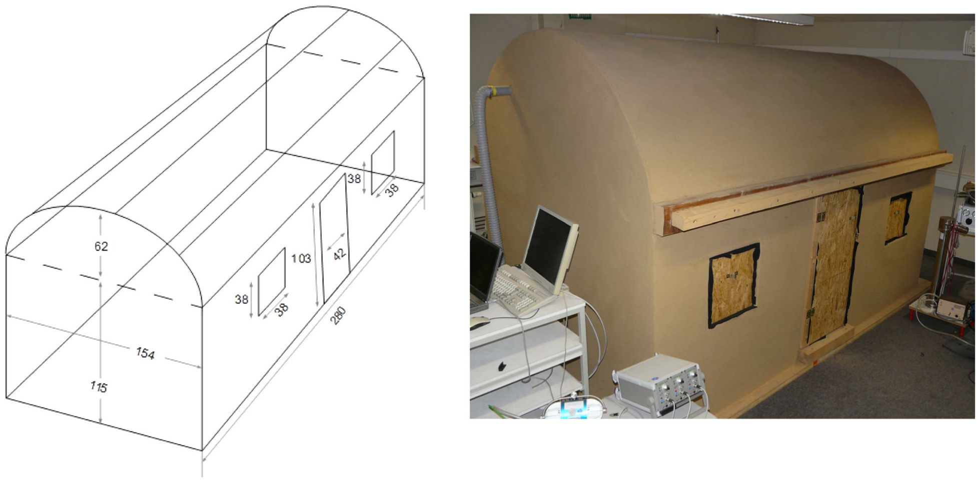

Figure 1. Sketch with dimensions (in cm, left) and photo (right) of the HMGU thoron experimental house inside which the experiments were performed.

In the present work, experiments have been carried out using the deposition-based Direct Thoron Progeny Sensors (DTPS) (Mishra and Mayya 2008, Mishra *et al* 2010) in a thoron experimental house at Helmholtz Zentrum München (HMGU) (Tschiersch and Meisenberg 2010). The objective was to study the thoron progeny characteristics such as the deposition velocities, spatial variability and dependence on aerosol particle concentrations. Since the deposition velocity is an important characteristic in the calibration of the DTPS, it is very important to study its dependence on aerosol concentration in a controlled environment. Details of the experimental house and the methodology followed are discussed in the following sections.

Material and methods

HMGU thoron experimental house

The experiments were performed inside the HMGU thoron experimental house. It has a volume of 7.1 m^3 (length 2.8 m, width 1.54 m, height 1.77 m), wherein the inner walls are coated with granite powder-enriched clay plaster which serves as a thoron exhaling surface with an area of 21 m^2. An external view and a schematic diagram are shown in figure 1. More details are given in Tschiersch and Meisenberg (2010).

The thoron activity in the room without air exchange was reported as 1720 $\pm$ 250 Bq at maximal exhalation, leading to a maximal potential alpha-energy concentration of the thoron decay products of 18.4 $\pm$ 2.7 μJ m^{-3}, equivalent to 243 Bq m^{-3}, assuming no air exchange and deposition. At an air exchange rate of 0.4 h^{-1}, the potential alpha-energy concentration of the decay products has been reported to be about 1.5 μJ m^{-3}, which is equivalent to 19.8 Bq m^{-3}. The ambient gamma dose rate inside the experimental room is 0.13 μSv h^{-1}.

Instruments deployed in the thoron experimental house

Measurements of thoron decay products inside the thoron experimental house were carried out using the DTPS (Mishra and Mayya 2008, Mishra *et al* 2010). These are passive sensors, basically LR115 type solid state nuclear track detectors, wherein the alpha tracks are created due to decay of deposited ^{212}Bi and ^{212}Pb progeny atoms. The selective registration of alpha particles from the thoron progeny is provided by mounting an additional energy degrader foil onto the detector, thereby selecting only the 8.78 MeV alpha particles emitted from ^{212}Po as the decay product of the deposited ^{212}Bi and ^{212}Pb atoms.

The response of the DTPS is directly proportional to the total deposited progeny atom flux on the detector surface, which can yield information only on the effective deposition velocities corresponding to the total progeny atom concentrations present in the atmosphere. Accordingly, the effective deposition velocity (V_e) for thoron progeny is defined by the formula

$$V_e = \frac{\text{Total atom deposition flux (atoms cm}^{-2}\text{ s}^{-1}\text{) of progeny species on the sensor}}{\text{Total atom concentration (atoms cm}^{-3}\text{) in the atmosphere}} \quad (1)$$

The effective deposition velocities combine the contributions from the fine (unattached) and the coarse (attached) fractions of the individual progeny species. In the case of ^{220}Rn progeny, the flux of ^{212}Pb + ^{212}Bi atoms on a given deposition surface may be given by

$$J_T = \{p_2 V_f + (1 - p_2)V_c\}n_2 + \{p_3 V_f + (1 - p_3)V_c\}n_3 \quad (2)$$

where n_2 and n_3 denote the atom concentrations, and p_2 and p_3 denote the fine fractions, of ^{212}Pb and ^{212}Bi in the atmosphere, respectively. V_f and V_c are the deposition velocities of the fine and coarse fractions.

From the definition in equation (1), the effective deposition velocity is then given by

$$V_e\,(\text{thoron progeny}) = \frac{\{p_2 V_f + (1 - p_2)V_c\}n_2 + \{p_3 V_f + (1 - p_3)V_c\}n_3}{n_2 + n_3} \quad (3)$$

The numerator of equation (3), which represents the total deposition atom flux, is obtained from the alpha tracks registered on the DTPS and the corresponding track registration efficiency (Mishra and Mayya 2008). The denominator represents the total thoron progeny atom concentrations in the indoor environment. It can be obtained by using a DTPS-mounted integrated sampler, wherein sampling is carried out by capturing the decay products on a filter at a certain flow rate for a pre-determined time. The DTPS faces the filter paper and registers the alpha particles emitted from the captured total thoron decay products (Mishra *et al* 2010).

Similarly, the attached fraction deposition velocity is given as the ratio of attached fraction deposition flux and the attached fraction concentration in the room. The attached fraction deposition flux was obtained using the DTPS capped with a suitable wire mesh (Mayya *et al* 2010). The attached fraction of thoron progeny concentration was measured using an integrated sampler mounted with a wire mesh and filter paper combination, whereas the DTPS was facing the filter paper. The tracks registered in this DTPS correspond to the attached fraction progeny concentration.

For thoron gas, passive time-integrated measurements were carried out with CR-39 based detectors commercially available as RADUET detectors (Tokonami *et al* 2005; Radosys Ltd, Budapest, Hungary).

Experimental methodology

Time-integrated deposition velocity of the thoron progeny at different locations in the house. Eighteen DTPS in bare mode and in wire-mesh capped mode were deployed from the centre towards the left and right side of the walls in the horizontal direction as well as towards the

Table 1. Deposition velocity ($V_{effective}$) and aerosol-attached deposition velocity ($V_{attached}$) measured at different points in the experimental house using the DTPS.

Position of detectors	Dosimeter code	Distance from centre (cm)	Deposited flux (10^{-3} atoms cm^{-2} s^{-1})	Atom conc. (atoms cm^{-3})	$V_{effective}$ (m h^{-1})	Attached Deposited flux (10^{-4} atoms cm^{-2} s^{-1})	Attached Atom conc. (atoms cm^{-3})	$V_{attached}$ (m h^{-1})
Centre	1	0	3.01	0.38	0.285	4.60	0.31	0.053
	2		2.31		0.219	3.80		0.044
	3		1.18		0.112	4.20		0.049
Towards left wall from centre	4	40	1.27		0.106	4.70		0.055
	5	90	2.96	0.43	0.248	4.40	0.31	0.051
	6	110	1.58		0.132	3.30		0.038
	7	120	1.30	0.4	0.117	3.80	0.30	0.046
	8	130	1.27		0.114	4.40		0.053
Towards right wall from centre	9	40	2.11	0.48	0.158	4.20	0.26	0.058
	10	90	2.06		0.155	3.80		0.053
	11	110	1.72		0.129	3.10		0.043
	12	120	1.94		0.146	3.30		0.046
	13	130	2.76		0.207	3.10		0.043
Towards the roof from centre	14	40	1.44	0.34	0.152	4.20	0.27	0.056
	15	90	0.99		0.105	3.80		0.051
	16	120	1.49		0.158	3.20		0.043
	17	140	1.75		0.185	3.30		0.044
	18	160	1.30		0.138	3.10		0.041

Table 1. (Continued.)

Position of detectors	Dosimeter code	Distance from centre (cm)	Deposited flux (10^{-3} atoms cm^{-2} s^{-1})	Atom conc. (atoms cm^{-3})	$V_{effective}$ (m h^{-1})	Attached Deposited flux (10^{-4} atoms cm^{-2} s^{-1})	Attached Atom conc. (atoms cm^{-3})	$V_{attached}$ (m h^{-1})
In the room corners	19		2.66	0.57	0.168	3.10	0.24	0.047
	20		2.59		0.164	2.80		0.042
	21		2.39		0.151	2.90		0.044
	22		2.31		0.146	3.20		0.048
Mean deposition velocity m h^{-1}					0.159 ± 0.045			0.048 ± 0.005

roof of the experimental house in the vertical direction. The details are given in table 1. Apart from these, four DTPS were deployed at the four corners of the house. The exposure was carried out for eight days. For measurement of thoron progeny atom concentration using a DTPS-loaded integrated sampler, the sampling was carried out for 1 h at $2\,\mathrm{l\,min^{-1}}$ at the selected positions, as shown in table 1. The exposure was carried out in test house conditions (at close to zero ventilation rate). For later exposures, additional thorium nitrate coated mantles were used to supplement the thorium source. During the experiments low aerosol concentration ~1500 per $\mathrm{cm^3}$ was maintained which was continuously monitored using a condensation particle counter (CPC 3022A, TSI Inc., Aachen, Germany).

Thoron progeny deposition velocity under high aerosol conditions. Since the deposition velocity is an environment-dependent parameter, its dependence on the aerosol concentration was studied in the experimental house. A high aerosol concentration of ~30 000 per $\mathrm{cm^3}$ was maintained. To increase the thoron source strength, additional thorium nitrate coated mantles were used in the experimental house. Ten DTPS in bare mode and 10 in wire-mesh capped mode were suspended in the center of the room for a period of four days. During this time, active samplings using DTPS-loaded integrated samplers were also carried out to estimate the progeny airborne atom concentration. The unattached fraction was also measured using the integrated sampler.

Spatial distribution of thoron gas and thoron progeny in the experimental house without any additional sources of thoron. In this case, the source of thoron gas and hence the decay products were only the walls of the test-house. The detectors for measuring thoron gas and the thoron progeny were suspended at different distances from the left and right walls. For thoron gas, passive time-integrated measurements were carried out with RADUET detectors (Radosys Ltd, Budapest, Hungary), whereas for thoron progeny measurements the DTPS (Mishra *et al* 2014) were used. The exposure was carried out for five days.

Results and discussion

The following results were obtained in the experiments carried out in the thoron experimental house, along with the methodologies described above.

Time-integrated deposition velocity of the thoron progeny at different locations in the house

In the case of DTPS, which are basically deposition-based progeny sensors, the calibration factor is dependent on the deposition velocity of thoron progeny atoms in the environment. Deposition velocity is an environment-dependent parameter that needs to be evaluated under different conditions. The experimental data available show large variations in the deposition velocity estimates of the progeny species (Jacobi 1972, Porstendörfer *et al* 1978, Knutson *et al* 1983, Toohey *et al* 1984, Bigu 1985, Morawska and Jamriska 1996, Zhuo *et al* 2000).

As shown in table 1, the average effective deposition velocity for thoron progeny is $0.159 \pm 0.045\,\mathrm{m\,h^{-1}}$, whereas that for attached fraction thoron progeny is $0.048 \pm 0.005\,\mathrm{m\,h^{-1}}$. Using the thoron progeny deposition model of Mishra *et al* (2009), the effective deposition velocity at 1500 particles/$\mathrm{cm^3}$ was calculated to be about $0.25\,\mathrm{m\,h^{-1}}$ in the experimental house. As discussed in Mayya *et al* (2010), the attached fraction deposition velocity is less dependent on the change in environmental parameters and it has been reported that the value varied between 0.02 and $0.05\,\mathrm{m\,h^{-1}}$ for thoron progeny in the experimental

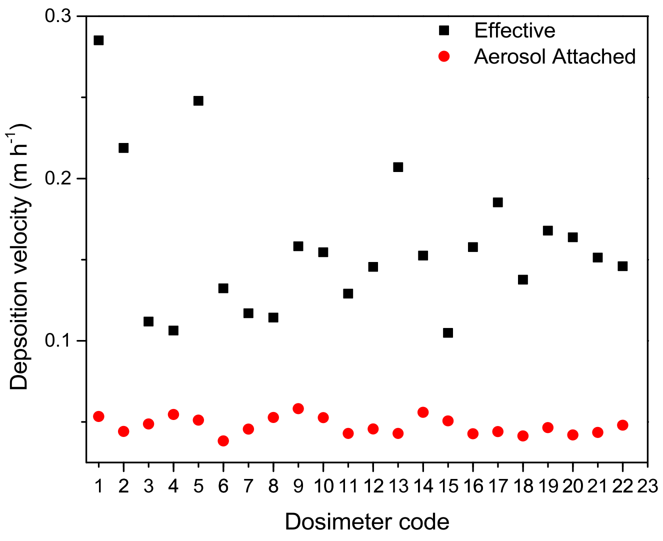

Figure 2. Effective and aerosol-attached deposition velocity at different points in the experimental house.

house as well as indoors. The ^{212}Pb attached fraction deposition velocity in the same experimental house has been reported as 0.01 m h^{-1} by Meisenberg and Tschiersch (2011).

Figure 2 shows the effective as well as the aerosol-attached deposition velocity versus the dosimeter code. It shows that, although the effective deposition velocities show a considerable variation within the experimental house, the standard deviation of the deposition velocities is less in the case of the attached fraction of thoron progeny. As discussed in Mayya *et al* (2010), the attached fraction of thoron progeny atoms is less dependent on environmental factors.

Thoron progeny deposition velocity under high aerosol conditions

The deposition velocity was studied in the experimental house in terms of dependence on the aerosol concentration. A high aerosol concentration of about 30 000 particles/cm^3 was maintained. The average effective deposition velocity was measured as 0.079 ± 0.009 m h^{-1}, whereas that for the attached fraction deposition velocity was 0.043 ± 0.014 m h^{-1}.

The effective deposition velocity under high aerosol conditions is closer to the thoron progeny deposition velocity generally measured indoors at such high aerosol concentrations (Mishra and Mayya 2008). Though the effective deposition velocity decreased with the increase in aerosol concentration, which is essentially attributed to a decrease in the unattached fraction, the attached fraction deposition velocity remained almost constant, nearly equal to that obtained when the particle concentration in the test house was only 1500 particles/cm^3.

These results point towards the fact that the wire-mesh capped sensors are more robust instruments for measurement of progeny concentration, as predicted by Mayya *et al* (2010). The calibration of the wire-mesh capped DTPS is based on the attached fraction progeny deposition velocity, which has proved to be least affected by any change in aerosol concentration. Thus, the test measurements in the thoron experimental house are also a useful sensitivity study of the wire-mesh capped DTPS.

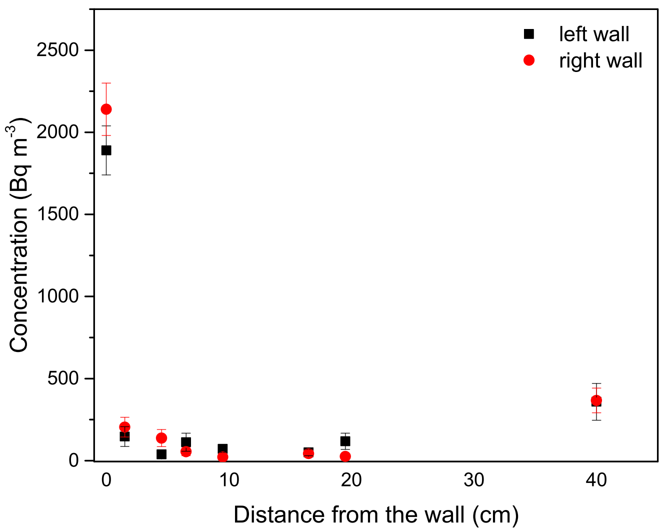

Figure 3. Thoron concentration measured at different distances from the wall.

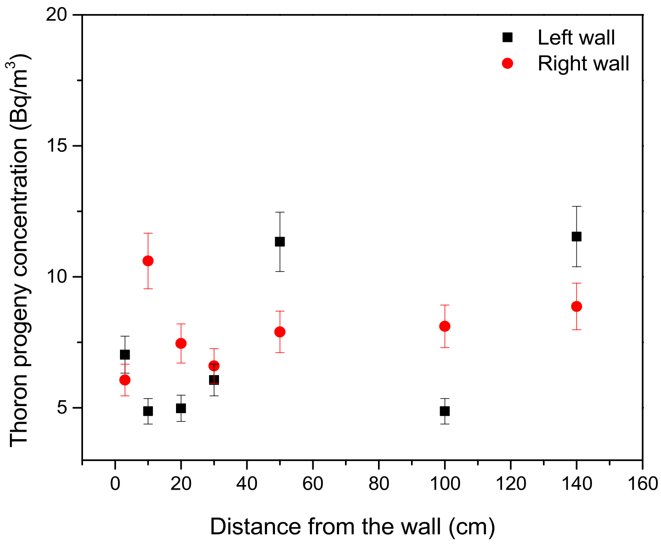

Figure 4. Thoron progeny concentration measured at different distances from the wall.

Spatial distribution of thoron gas and thoron progeny in the experimental house without any additional sources of thoron

The results depicted in figure 3 show that the thoron gas concentration varied from about 2000 Bq m^{-3} near the walls to about 72 Bq m^{-3} at 10 cm from left wall and 22 Bq m^{-3} from the right wall. On the other hand, as shown in figure 4, thoron progeny concentration was uniformly distributed throughout the room from both left and right walls, with the average concentration being 7.6 ± 2.2 Bq m^{-3}. This clearly indicates that thoron gas concentration as an input for estimating the thoron progeny concentration could lead to large uncertainties and hence a direct measurement of progeny concentration is more prudent.

Conclusions

Thoron progeny behavior under controlled conditions in the HMGU experimental house was studied, under which experiments were carried out to study the thoron progeny deposition velocities at different aerosol concentrations. The spatial distribution of thoron progeny in the experimental room was also studied using time-integrated measurements. The following observations were noted.

(a) At a particle concentration of 1500 particles/cm^3, the effective deposition velocity showed large scattering within the room at different distances from the center. The attached fraction deposition velocity remained uniform at different distances from the wall. Since the particle concentration was less than that observed indoors, the effective deposition velocity was also higher than that observed indoors. As expected, the attached fraction deposition velocity remained the same despite the low particle concentration. This further confirms that the wire-mesh capped DTPS is a more robust way to measure the thoron progeny concentration, because it will be less dependent on changes in environmental parameters like aerosol concentration.
(b) At a particle concentration of 30 000 particles/cm^3, the effective thoron progeny deposition velocity was obtained to be similar to that under typical indoor conditions (ventilation rate 1 h^{-1}). The attached fraction deposition velocity was in the same range as obtained for low aerosol concentration.
(c) The measurements in the thoron experimental house prove the constancy of the attached fraction progeny deposition velocity, thus proving that the wire-mesh capped sensor is more robust against change in environmental parameters.
(d) The uniform spatial distribution of thoron progeny was confirmed within the experimental house. This indicates that direct measurement of thoron progeny concentration should be carried out instead of inferring it from thoron gas concentration, which does not show a uniform spatial distribution within the room.

Acknowledgments

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