

## **ORIGINAL RESEARCH**

# Testing of thoron cross-interference of continuous radon measuring instruments

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## Abstract

Thoron (220Rn) may interfere with radon (222Rn) measurements, if present. We measured the thoron cross-interference (CI) signal of nine types of electronic radon instruments in constant thoron concentration without the presence of radon. The CI signal increases for the first 3 days of the exposure. Also, the initial interference signal may vary between instruments. Therefore, we propose a new test method for quantifying the thoron CI in radon measuring instruments. This includes exposure of the instrument in constant thoron concentration for a minimum of 3 days and fitting the acquired data in a simplified function, which will provide two parameters: initial and final CI.

Keywords: radon measuring instrument; thoron; Rn-220, cross-interference; test method

adon (<sup>222</sup>Rn) measuring instruments may be sensitive to thoron (<sup>220</sup>Rn) in varying degrees. It is important that a radon measuring instrument is not too sensitive to thoron, or the sensitivity is known because, in some cases, thoron interference may lead into a false conclusion, such as:

- Assessment of radon exposure levels in surveys and epidemiological studies. Thoron interference may be pronounced especially in the lower end of radon concentrations and may lead to too high estimates of radon concentration (1).
- Assessment of effective doses at workplace. According
  to European Basic Safety Standards, if a workplace
  cannot reduce radon concentration below the reference
  level, assessment of effective dose is carried out. If the
  dose exceeds 6 mSv per year, the exposure must be regarded as planned exposure, and below it, exposures
  are kept under review. Thoron interference during the
  measurement may lead into wrong classification and
  incorrect dose assessment because the dose conversion
  factors of radon and thoron are different (2).
- Decision on radon mitigation. The measured radon concentration may be recorded falsely as above reference level if thoron was present in the vicinity of the detector. Radon mitigation may be time-consuming

and expensive and, if imposed by the competitive authority by false basis, may lead into inequality among workplaces. In addition, different mitigation measures are used for radon and thoron (3).

Thoron entry into the sensitive volume of the detection unit is generally controlled by a diffusion barrier, which can be a filter (e.g. AlphaE by Bertin Instruments) or a narrow gap (e.g. Corentium Pro by Airthings AS). Separation of radon and thoron is based on the different half-lives of these isotopes. The diffusion barrier should also prevent radon and thoron progeny from entering the sensitive volume of a radon or thoron measuring instrument (4).

The diffusion barrier, however, must not be too effective because this will slow down radon entry into the sensitive volume of the detection unit. Subsequently, response time of the unit will be longer, and rapid changes in radon concentration cannot be measured accurately. Rapid changes need to be measured, for example, at workplaces where mechanical ventilation is operated according to working hours. When the ventilation is operated at low or zero power, radon concentration indoors may increase significantly. If the instrument reacts too slowly, the measured radon concentration during the first working hours in the morning may be recorded as too high and lead into a false conclusion.

The scope of this work was to investigate cross-interference (CI) of thoron for electronically operated, continuous radon measuring instruments in pure thoron atmosphere. Usability of CI test method described in standard IEC 61577-2 will be discussed, and a new test method is proposed.

### **Theory**

Radon instruments employ either active or passive sampling (5). In active sampling, air is pumped into the sensitive volume of the detection unit through a filter. In most cases, thoron gas enters the detection volume along with radon regardless of its short half-life of 55.6 sec unless transfer volume lag (which would allow the decay of thoron before it enters the detection volume) is applied. In passive sampling, air is transported into the detector by diffusion. In order to prevent thoron (and radon and thoron progeny) from entering the detection volume, a diffusion barrier is normally applied either in form of a filter or a narrow gap. Depending on the diffusion coefficient of the barrier, various amounts of thoron may enter the detection volume.

After thoron has reached the detection volume of the instrument, the first thoron progeny <sup>216</sup>Po reaches equilibrium inside the detection volume within a couple of seconds and, thus, doubles the alpha activity inside the sensitive volume. The next progeny is <sup>212</sup>Pb, whose half-life is 10.64 h. Its in-growth takes about 70 h to reach 99% of the equilibrium activity. The next decay product <sup>212</sup>Bi has a half-life of 60.55 min, and it follows the in-growth of <sup>212</sup>Pb with only a short lag. Here, the decay chain branches and the last two progeny <sup>212</sup>Po and <sup>208</sup>Tl follow closely the ingrowth of <sup>212</sup>Bi (Fig. 1). The

maximum activity inside the detection volume is reached in about 3 days if thoron concentration remains stable at the site of measurement.

Some continuous radon measuring instruments employ silicon alpha detectors and apply spectroscopy, that is, they separate alpha counts from different isotopes based on their energies. The most advanced of these types of instruments can calculate both radon and thoron concentrations from the recorded data (Fig. 2). There are also instruments that can be operated in 'fast mode' and 'normal mode'. In the fast mode, only <sup>218</sup>Po (and <sup>212</sup>Bi) counts are included in order to shorten the response time of the instrument in changing radon concentration. In the normal mode, counts from <sup>214</sup>Po are also included. The manufacturers generally do not inform consumers what range of alpha particle energies is included in the calculation. and hence, <sup>212</sup>Po and <sup>216</sup>Po counts may or may not cause CI. Moreover, there are instruments that do not apply spectroscopy and calculate results from all recorded counts. In monitors that apply electrostatic collection of progenies onto the detector, most recorded counts are due to radon and thoron progeny; alpha emissions from <sup>222</sup>Rn and <sup>220</sup>Rn contribute only little to the total counts (6). Ionizing chambers detect both alpha and beta emissions. Some instruments have predictive algorithms for making the response time shorter. Some monitors apply averaging for smoothing the data.

It is obvious that the CI signal from thoron may evolve in different ways considering the range of detection principals. This has been reported previously by, for example, Michielsen and Bondiguel (7). Two common features, however, can be distinguished. There is a fast response for

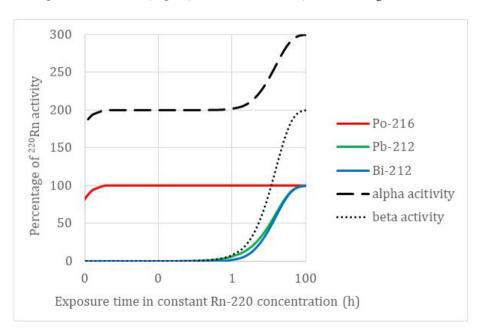


Fig. 1. Ingrowth of thoron progeny inside the sensitive volume of the detection unit of a radon measuring instrument in a constant thoron concentration.

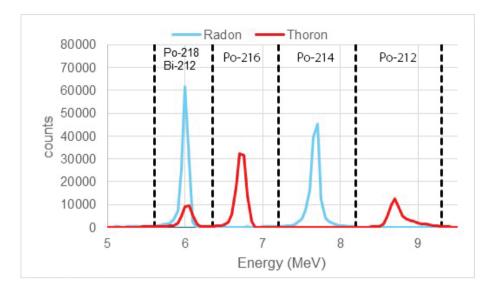


Fig. 2. Radon (with progeny) and thoron (with progeny) spectra recorded using the alpha spectrometer RAD 7 (Durridge).

<sup>220</sup>Rn, or it may be lacking. This CI signal comes from thoron and 216Po if their alpha particles are counted and not rejected by the instrument. This signal represents a case where there is a short-term, pulse-like thoron concentration around the instrument. We call this signal as initial cross-interference (ICI). If the exposure continues at a constant thoron concentration, the CI signal increases for the first 3 days and is due to the <sup>212</sup>Pb and its progeny. After this transient period, an equilibrium is attained. We call this signal as final cross-interference (FCI).

The relative intensity (branching ratio) of <sup>212</sup>Bi alpha decay is 36%. The maximum ICI-to-FCI ratio is obtained when the <sup>216</sup>Po and <sup>212</sup>Bi alpha particles are counted as radon progeny, but all high-energy alpha particles of <sup>212</sup>Po are rejected. In this case, the ratio would be 100%:136% = 0.74.

We can write a simplified function to which we can fit the acquired CI signal,  $\mu(t)$ :

$$\mu(t) = \mu_i + \mu_s \left(1 - e^{-\lambda_{Pb-212}t}\right)$$

The ICI is then calculated as

$$ICI = \frac{\mu_i}{C_{Tn}} \times 100\%$$

The FCI is calculated as

$$FCI = \frac{\mu_i + \mu_s}{C_{T_n}} \times 100\%$$

in which

 $\mu_i$  = the ICI signal from the instrument after the start of thoron exposure

 $\mu_s$  = the difference between the final signal (t > 5 days) and the µ,

 $^{\lambda}_{Pb-212}$  = the decay constant of  $^{212}$ Pb

t =time from the start of thoron exposure

 $C_{T_n}$  = thoron concentration used in the exposure According to Standard IEC 61577-2, the CI of thoron for an instrument made for radon measurement should be less than 20%. The test method described in the standard requires minimum 4-h exposure in constant thoron concentration and subsequent data acquisition for at least 1 h. As can be seen in Fig. 1, the required minimum exposure time is too short for sufficiently quantifying the true CI signal. In this article, we shall demonstrate that the test should be carried out for at least 70 h in order to reliably estimate the FCI signal in constant thoron exposure.

## Materials and methods

We selected nine different models of radon monitors for testing the CI (Table 1). The CI tests were carried out at laboratories of STUK - Radiation and Nuclear Safety Authority, Finland and Faculty of Physics, Sofia University (SUBG), Bulgaria. The set-ups and the equipment were similar and varied mostly in the size of calibration container. Two monitors were tested at both STUK and SUBG for validating the test procedures.

At STUK, the thoron atmosphere was created in a 101.1 L Emanation Calibration Container (Saphymo GmbH). Air exchange through the container was created using the Qdos60 peristaltic pump (Watson Marlow) and monitored from the air outlet using the Thermo GFM Pro flow meter. The inlet air was from the laboratory and first desiccated with Laboratory Drying Unit (Durridge) filled with freshly regenerated Drierite. Second, radon in the air was removed using a 1-litre activated carbon unit (Saphymo). Next, the air was

Table 1. Instruments selected for cross-interference testing

Instrument	Detector	Serial number	Manufacturer	Tested at
AlphaE	Silicon diode	000260 000542 000499	Saphymo GmbH	SUBG and STUK
AlphaGuard PQ2000 Pro	Ionization chamber	EF1641	Genitron GmbH	STUK
AlphaGuard PQ2000	Ionization chamber	EF0408	Genitron GmbH	STUK
Corentium Pro	Silicon diode	2700007355 2700007357	Airthings AS	STUK
Airthings wave	Silicon diode	2900151289	Airthings AS	STUK
Airthings wave plus	Silicon diode	2930	Airthings AS	STUK
Radon Eye +²	Ionization chamber	PE21812110009 PE21904100016	FTLAB Co., Ltd	SUBG and STUK
TSR 3	Silicon diode	16014	Tesla (CZ)	SUBG
TSR 4M	Silicon diode	19015	Tesla (CZ)	SUBG

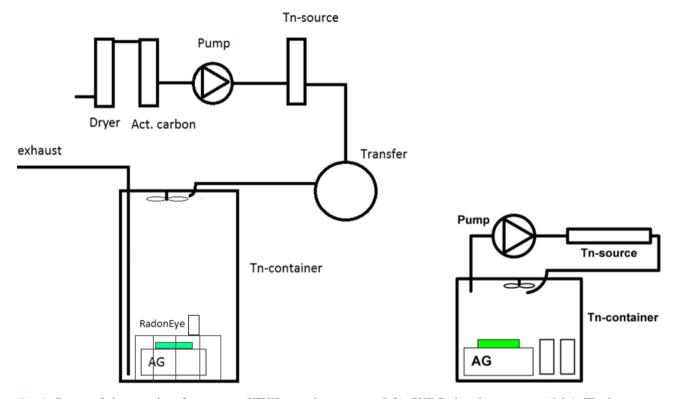


Fig. 3. Set-up of the cross-interference test: STUK open-loop system (left); SUBG close-loop system (right). The instrument under testing (in this example RadonEye) was placed on a grid above the reference instrument (AG = AlphaGuard PQ2000 Pro RnTn) at STUK and around the reference instrument at SUBG.

directed into a flow-through thoron source TH-1025 from Pylon Electronics Inc. The thoron-rich air was then directed (after a 175 mL transfer volume) in front of a small fan situated under the lid of the container (Fig. 3, left). The tubing used in the set-up was the clear PVC metric tubing (Thermo Scientific/Nalgene). PVC has lower radon leakage rates than, for example, silicon rubber, norprene, or tygon (8). At SUBG, the thoron atmosphere was created in a 50.4 L Emanation

Calibration Container (Saphymo GmbH). The air exchange through the system was created using the Masterflex<sup>TM</sup> peristaltic pump (Cole-Parmer). A close-loop system was chosen, so there was no need of desiccant. The thoron in the system was supplied by a flow-through thoron source TH-1025 from Pylon Electronics Inc., and the thoron-rich air from the source was directed in front of a small fan attached to the lid of the container (Fig. 3, right).

Thoron concentration was regulated by adjusting the flow rate of the peristaltic pump in both set-ups. Thoron concentration used in the CI tests was kept generally high (>20 kBq/m<sup>3</sup>) to allow good counting statistics and easy fitting of the data. When testing the RadonEve RD200 Plus2, the thoron concentration in the tests was adjusted to a lower value because the maximum detectable radon concentration of the instrument is only 9,400 Bg/m<sup>3</sup> and the thoron CI is high.

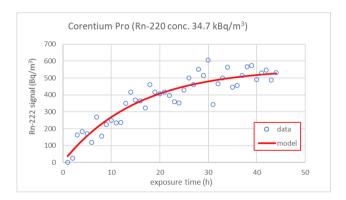
In both systems (STUK and SUBG), the homogeneity of thoron gas inside the container was validated with aerogel samplers (9). The variation of thoron concentration inside the container was assessed as  $\pm 2\%$ . Both laboratories STUK and SUBG used the same type of reference instruments for thoron concentration measurement (AlphaGuard PQ2000 RnTn, Saphymo GmbH), which had been calibrated against the primary thoron standard at IRSN (10).

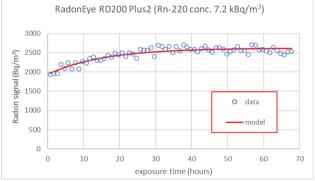
For the STUK system, the flow rate of the AlphaPump was checked before and after exposures using the GFM 17 mass flow meter (Aalborg). Humidity and temperature were measured using the HygroClip HC2A-S-probe (Rotronic AG). Air velocity measurements were carried out using the Swema 3000MD and SWA31 hot-wire anemometer. The flow rate at the grid level, in the middle, was recorded as  $0.24 \pm 0.03$  m/s, and it was normal to the grid plane. In most experiments at STUK, the humidity was not regulated, and therefore, it decreased when dry

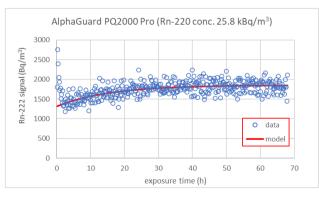
thoronous air was pumped into the container. In three tests, humidity in air was regulated with a bottle of supersaturated MgCl<sub>2</sub> aqueous solution placed inside the container. At the SUBG system, the humidity remained constant due to the closed-loop exposure. The temperature and air pressure during the exposures were not regulated in either laboratory.

#### **Results**

Evolution of different types of CI signals in constant thoron concentration can be seen in Fig. 4. Some thoron can enter the sensitive volume of the detection unit of Corentium Pro as the signal increases over time. It is, however, obvious that most alpha emissions from <sup>216</sup>Po are rejected from the calculation of radon concentration as the ICI is very small. RadonEye RD200 Plus2 is sensitive to thoron, and the ICI signal is already significant. The increase of the CI signal fits well into the simplified function  $\mu(t)$ . AlphaGuard PQ2000 Pro (operated in 10-min diffusion mode) exhibits a peculiar response. The CI signal decreases rapidly during the first hour and then follows the simplified function. This is probably due to internal calculation that takes the physical response time into consideration and aims at predicting the true radon concentration and, hence, making the apparent response time shorter. Thoron exposure stops at 92.5 h for AlphaE. When thoron exposure stops, the counts due to <sup>216</sup>Po decrease immediately, and the remaining signal follows







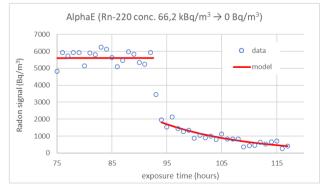


Fig. 4. Evolution of cross-interference signal in three different radon measuring instruments.

Table 2. Cross-interference signals for all cross-interference tests carried out at STUK and SUBG

Instrument	Tested at	s/n	Test dates	ICI (initial)	FCI (final)
AlphaE	STUK	000260	5–10 Jun 2019	6.5% <sup>p</sup>	9.3% <sup>p</sup>
			31 Jul-5 Aug 2019	8.9% <sup>p</sup>	12% <sup>p</sup>
	STUK	000542	11–17 Jun 2019	5.6% <sup>p</sup>	9.2% <sup>p</sup>
	STUK	000499	31 Jul–5 Aug 2019 5–9 Dec 2019	8.7% <sup>n</sup> 5.7% <sup>n</sup>	13% <sup>n</sup> 8.6% <sup>n</sup>
	31010	000177	5 7 Dec 2017	3.770	0.070
AlphaE	SUBG	000499	14-17 Oct 2019	4.5%	13.7%
			22–28 Oct 2019	11.5%	17.0%
			16-20 Sep 2019 26-30 Sep 2019	10.0% 12.7%	17.1% 18.7%
			4–8 Nov 2019	15.5%	20.2%
AlabaCuand DO2000 Bas	CTLIV	EELCAL			7.29/
AlphaGuard PQ2000 Pro (10-min diffusion mode)	STUK	EF1641	6–9 Aug 2019	5.1–11%	7.2%
•	CTI III	FF0.400	22 27 1 2222	4.4.0.00%	4.007
AlphaGuard PQ2000 (10-min diffusion mode)	STUK	EF0408	22–27 Jan 2020	4.6–9.2%	6.0%
,					
AlphaGuard 2000 RnTn Pro	IRSN	EF2283	18–22 May 2018		1.1%
			22-24 May 2018		0.6%
RadonEye +2	STUK	PE21812110009	20-23 August 2019	28%	42%
	STUK	PE21904100016	28 Nov-I Dec 2019	27%	37%
RadonEye +²	SUBG	PE21904100016	14-17 Oct 2019	32.6%	52.7%
	20RG	PE21904100016			
			22–28 Oct 2019	38.7%	54.7%
			4–8 Nov 2019	18.7%	42.3%
Corentium Home	STUK	2403008304	20-24 Jun 2019		1.8%
			24-29 Jul 2019		2.5%
Airthings Wave	STUK	2900151289	28 Jun-2 Jul 2019		1.3%
			6–9 Aug 2019		2.3%
Airthings Wave Plus	CTLIV	2020	· ·		
	STUK	2930	24–28 Jun 2019		2.7%
			24–29 Jul 2019		3.6%
Corentium Pro	STUK	2700007355	3-5 Sep 2018	0.2%	1.2%
		2700007357	3–5 Sep 2018	0.0%	1.6%
TSR3 – Fast mode	SUBG	16014	16-20 Sep 2019	1.0%	7.7%
			26-30 Sep 2019	1.2%	12.3%
TCD2 CI I	CLIDG	14014	·		
TSR3 – Slow mode	SUBG	16014	22–28 Oct 2019	2.7%	15.3%
TSR4M – Fast mode	SUBG	19015	22–28 Oct 2019	6.2%	125%
			16-20 Sep 2019	-	127%
			26-30 Sep 2019	-	186%
			4-8 Nov 2019	11.2%	114%
TSR4M – Slow mode	SUBG	19015	22-28 Oct 2019	15.9%	85.8%
	<b>-</b>		16-20 Sep 2019	-	69.5%
			26–30 Sep 2019	7.9%	115%
			4–8 Nov 2019	18.7%	76.4%
DoseMan Fast Mode	IRSN	DM357	18–22 May 2018	11%	36%
			22-24 May 2018	18%	39%
DoseMan Slow Mode	IRSN	DM357	18-22 May 2018	14%	41%
			22-24 May 2018	26%	48%

Three tests performed at IRSN during the same project are also included (11). IRSN exposure facility differs from that of STUK and SUBG, and the results are presented only for reference.

<sup>&</sup>lt;sup>n</sup>Air flow normal to the plane of the air inlet.



 $<sup>{}^{\</sup>mathrm{p}}\mathrm{Air}$  flow direction parallel to the plane of the air inlet.

the decay of <sup>212</sup>Pb. Cross-contamination signal remains for 3 days even if there is no thoron present.

The CI results in Table 2 show that despite its short half-life, some thoron always diffuses into the sensitive volume of the detection unit of radon measuring instruments. All tested radon instruments except for RadonEye, TSR4M, and DoseMan comply with the standard IEC 61577-2 requirement of CI being <20%.

Another observation is that the repeatability of the test is not perfect. First, there may be differences between instruments of the same model, which explains some of the variation. These could be due to internal calibration and settings of region of interest, small differences in size of diffusion gap, or cleanness of the diffusion barrier filter. Second, fitting data that are scattered due to counting statistics to a mathematical function always lead to uncertainties and variations in the results. The fitting of the results into the simplified function  $\mu(t)$  was, however, in many cases, straightforward and accurate because of low counting uncertainties resulting from the rather high thoron concentrations used in the exposures (see Fig. 1). Hence, counting statistics and imperfect data fitting do not explain the variation observed in replicated tests carried out on a single instrument.

We suspected that the direction and velocity of the air flow inside the calibration container may affect the results especially for instruments that have an air inlet at one side of the unit. Therefore, we tested some of the instruments, so that the air inlet was either parallel or normal to the direction of the air flow (see AlphaE test results in Table 2). The number of tests and instruments were not enough to obtain any conclusive evidence supporting this.

AlphaE (s/n 00499) was one of the two instruments used for comparing STUK and SUBG test chambers. SUBG tested this instrument several times, and the measured *ICI* was 4.5–15.5%. The test at STUK resulted in 5.7%. However, the *FCI* measured at STUK (8.6%) was clearly lower than those measured at SUBG (13.7–20.2%). The other comparison instrument (RadonEye RD200 +², s/n PE21904100016) exhibited similar trend; the *ICI* obtained at STUK was in the range of *ICI* obtained at SUBG, but the *FCI* obtained at STUK was somewhat lower.

The chamber at SUBG is half the size of that of STUK but use the same type of fan. Therefore, the air velocity at the point of testing is probably higher at SUBG chamber than that at STUK (0.24 m/s). The flow velocities were not recorded at SUBG, so we cannot conclude if air velocity has an influence on the results. Anyway, the reason for variation of results should be examined in more detail.

## **Discussion**

The FCI varied from less than 2% to over 100%. In most cases, the amount of CI is acceptable and below the <20%

requirement given in standard IEC 61577-2. Only three instruments did not comply with this.

The variation of the results clearly indicates the different design choices made by manufacturers. As explained before, an efficient diffusion barrier reduces not only the CI but also the responsiveness of the instrument in detecting changes in radon concentration. This has been evident in Corentium Pro, which has little CI but a long response time. The manufacturer has acknowledged this feature and has released a new software in autumn 2020. Using the new software, radon concentration can be recorded from <sup>218</sup>Po counts only. The instrument gains a faster response time at the cost of counting statistic. RadonEye, which is one of the instruments sensitive to thoron, has been marketed as one of the most responsive instruments on the market. The instrument is indeed responsive, but at the cost of sensitivity to thoron.

If there is little thoron in the air, and rapid changes in radon concentration need to be measured, short response time is a desired feature when choosing the right instrument for the task. Similarly, when measuring long-term average concentration of radon in an underground facility or a building with elevated levels of thorium in the construction materials, the significant feature is the thoron CI. Under these circumstances, all manufacturers should include thoron CI and response time in the specifications of their instruments. With information on response time and thoron CI, the end-user would be able to select the right instrument for different environments and purposes of measurement. Unfortunately, this information is seldom available in the specifications of radon measuring instruments.

Due to the short half-life of thoron (55.6 sec), a thoron atmosphere needs constant mixing of air in order to obtain evenly distributed thoron gas across the volume. The larger the volume, the more difficult this is, and more powerful fans must be applied. If the instrument tested for CI is sensitive to air flows, the air flow velocity should be low, just enough to obtain homogeneity. There is always a trade-off between maintaining homogeneous thoron concentration, which requires high airflow in the exposure chamber and the risk of introducing additional thoron CI in air-flow sensitive instruments. Based on our observations, we suspect that some of the variation in the results may be due to different air velocities and directions during the tests. Other reasons for the observed variation between instruments of the same model may be the differences in internal sensitivity and calibration, differences in the diffusion barrier, differences in other environmental conditions during the tests, and the precision of fitting the data into the simplified function,  $\mu(t)$ . More studies are necessary to investigate the possible influence of the airflow in the test chamber on the measured CI values.

The main finding of this study is that the test protocol described in standard IEC 61577-2 is not adequate for quantifying the CI signal from thoron. The minimum duration of the test is too short for assessing true thoron CI in constant or repeated thoron exposures. Furthermore, the outcome of the test described in the standard depends on the duration of the exposure of the test. Our suggestion for the test protocol is

- · Recording of air flow velocity inside the test chamber
- Exposure of the instrument under testing for minimum 70 h in constant thoron concentration
- Calculating the initial and FCI signal during the test

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