The thoron issue: exhalation rate and interference with radon measurements performed with passive radon dosemeters

Mauro Magnoni[*1], Enrico Chiaberto[1], Massimo Faure Ragani[2]

[1] Technological and Physical Risks Department, ARPA Piemonte, Via Jervis, 30 – 10015 Ivrea (TO), Italy
[2] Environmental Radioactivity Unit, ARPA Valle d'Aosta, Loc. La Maladière, 48 - 11020 Saint-Christophe (Aosta) Italy,
*Corresponding author: Mauro Magnoni
m.magnoni@arpa.piemonte.it

Introduction

Although very seldom thoron (^{220}Rn; $t_{1/2}$=55.6 seconds) can be considered as a significant source of exposure to population and workers, there are some particular cases, for example when bricks containing high ^{232}Th levels are used as main material in building constructions, in which the thoron dose is not negligible. Moreover, the thoron contribution can't be simply ignored even in standard situations, i.e. when the concentrations of the thorium series radioisotopes in soil and building materials are similar to those of the uranium series ones. In fact, it can be shown that the thoron exhalation rate from the walls of a room can interfere with the radon concentration measurements performed by means of the most widely used passive radon detectors, those equipped with nuclear track etched detectors such as CR-39, for example. Actually, as the thoron concentrations close to the walls of a room are usually much higher than those in the center, the radon dosemeters, that are normally installed very close to walls, may be exposed to quite high thoron concentrations. In the following Figure 1 the radon and thoron activity concentration profiles are shown for an hypothetical room in which the radon and thoron sources are only due to the exhalation from building materials containing the same massic activity concentration of the parents of the two natural series, namely ^{238}U and ^{232}Th respectively. In this simulation a diffusion-dispersion mechanism is assumed and the radon-thoron source strength is set in such a way to produce close to the wall an asymptotic activity concentration of about 800 Bq/m^3.

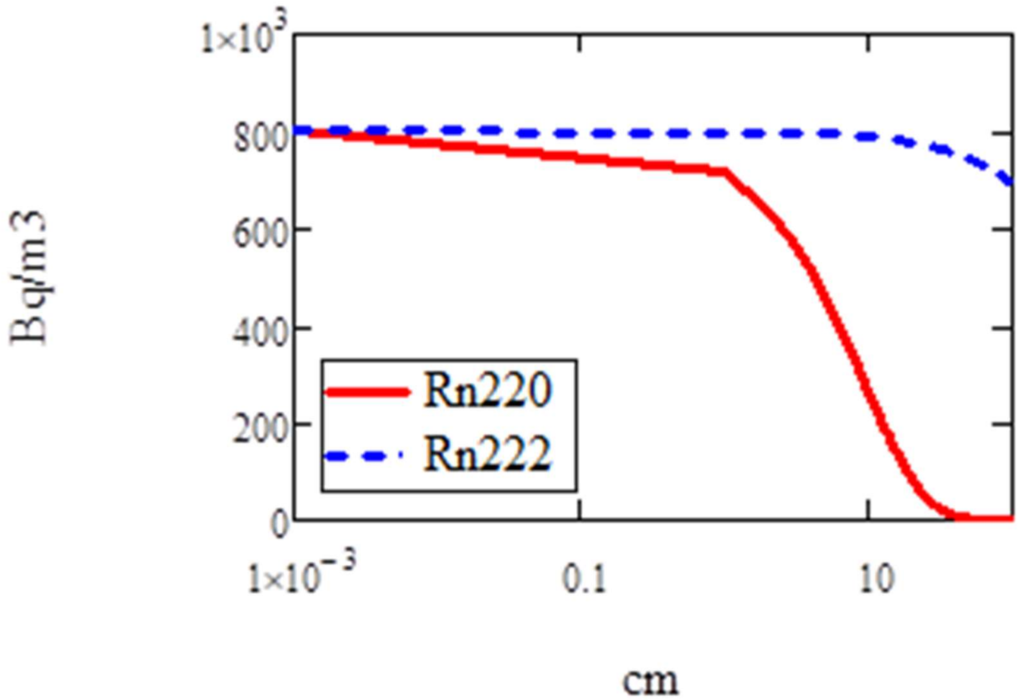

Figure 1 – Theoretical simulation of ^{222}Rn and ^{220}Rn profiles in a standard room. In abscissa the distance from the wall. The thoron activity concentration remains high at short distances (a few centimeters)

In figure 1 it can be easily seen that while the ^{222}Rn activity concentration remains almost constant at great distances (> 10 cm), the ^{220}Rn drops dramatically, reaching a negligible value in the center of the room. However, at short distances (a few centimeters, the typical range of the dosemeters place), the thoron levels are still very high (at least 600 Bq/m^3) thus able to interfere with the radon measurements.
Therefore, in order to tackle properly this problem it is important: 1) to assess the response to thoron of the radon measurements passive devices; 2) to evaluate the thoron exhalation rates of the different building materials.

Materials and methods

In order to study the sensitivity to thoron of a typical passive radon detector, two different devices were studied:

- The ARPA Piemonte radon dosemeter, equipped with two CR-39 detectors and with a polyethylene bag as a radon permeable barrier preventing radon daughter to enter inside the dosemeter (Figure 2)
- The Radout™ , from Miam Company, an half sphere shaped dosemeter, equipped with just one CR-39 detector, in which the acts as barrier for the radon daughters the thin circular slit existing between the bottom stopper and the semisphere (Figure 3)

Figure 2 – The ARPA Piemonte dosemeter: radon enters into the dosemeter passing through the radon permeable polyethylene bag

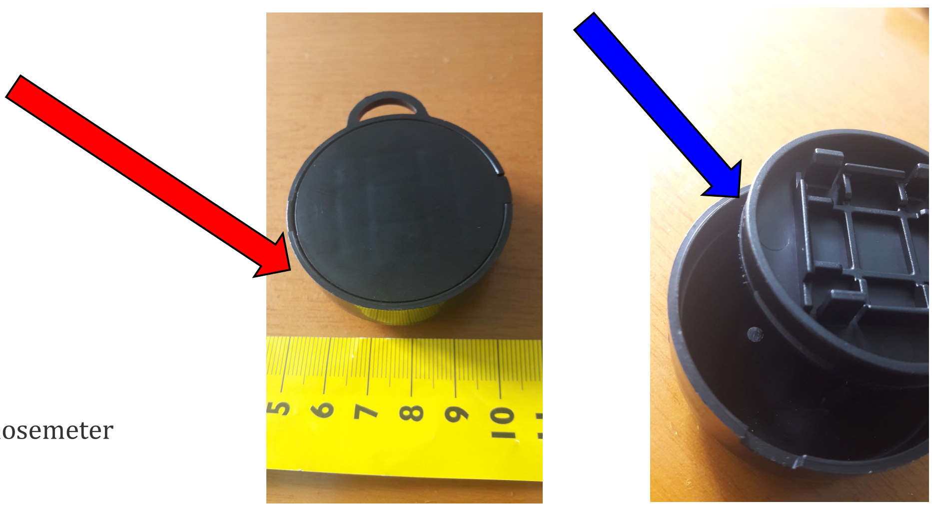

Figure 3 – The Radout™ dosemeter

The two dosemeters were put in a thoron chamber (18 liters) and exposed for 121 hours. Inside the chamber, the Thoron Scout, a monitor from Sarad Company, equipped with a silicon surface detector (alpha spectrometer) was able to register the thoron levels every hour. The experimental apparatus is showed in Figure 4: a pump, with a flow of 0.5 liter per minute, forced the thoron produced by a ^{232}Th source to enter into the chamber (Figure 4). An external computer collected the data. Due to the forced air circulation the thoron atmosphere inside the chamber is supposed to be homogeneous.

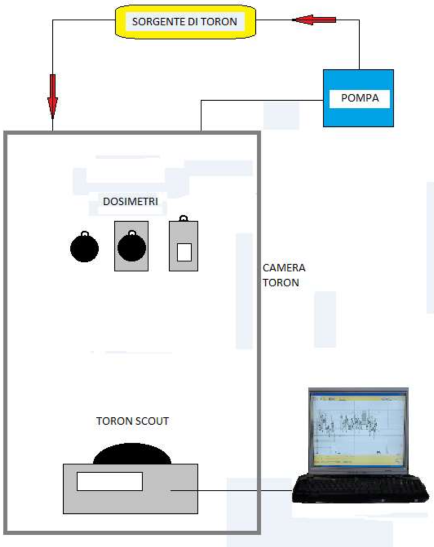

Figure 4 – The thoron chamber and the experimental apparatus

Before discussing the experimental results some considerations have to be made regarding the different mechanisms of the radon and thoron diffusion into the two devices.
For the ARPA Piemonte dosemeter the polyethylene barrier prevents entering the radon daughters existing in the ambient air and slows down the diffusive flow of the radioactive noble gases accordingly with the following simple expression, coming from the solution of diffusion equation for a radioactive substance:

$$C(d) = C_0 e^{-\sqrt{\frac{\lambda}{D_m}}\cdot d} \quad (1)$$

where C_0[1] is the activity concentration in the polyethylene material on the outside of the bag, $C(d)$ is the activity concentration inside, D_m is the corresponding radon-thoron diffusion coefficient, d is the thickness of the polyethylene foil (about 60 μm) and λ is the decay constant. Putting in (1) the proper numerical values we have: $C(d)_{Rn222}$ =0.975·C_0 and $C(d)_{Tn=}$0.145·C_0. While, as expected, the inside radon concentration remains almost equal, the thoron concentration is reduced but not at a completely negligible level: a significant quantity of thoron still survives, reaching the inner part of the detector.
The Radout™ diffusion mechanism is different and difficult to be modelled using the diffusion equation. A simplified approach is thus proposed, linearizing the diffusion equation as follows:

$$\frac{dC}{dt} + \lambda C = -\frac{D \cdot (C - C_0)}{\Delta x} \cdot \frac{S}{V} \quad (2)$$

in which D is the radon diffusion coefficient in air, S and Δx are respectively the area and the thickness of the slit (see Figure 2) and V is the volume of the semispheric Radout™ chamber. Taking the asymptotically value of the solution of equation (2), the following expression can be written:

$$\frac{C}{C_0} = \frac{1}{1 + \frac{\lambda \Delta x V}{DS}} \quad (3)$$

[1] It is important to point out that C_0 is not the radon activity concentration in air: it is the equilibrium value of the radon (or thoron) activity concentration in the polyethylene depending on the radon solubility in the material

It gives the fraction of radon and thoron able of enter into the device. Putting in equation (3) the proper data we obtain a value of almost 100% for radon and up to 15-16% for thoron, depending of the choice of the values of the parameters of the model. A calculation of the capability of thoron to reach the inner part of the dosemeter and thus contributing to the alpha irradiation of the CR -39 detector was also made using a Monte Carlo technique, developed using the open source package R (see Figure 5). Both approaches, the analytical and the Monte Carlo, demonstrated that a small, but not negligible quantity of thoron can enter into the dosemeter.

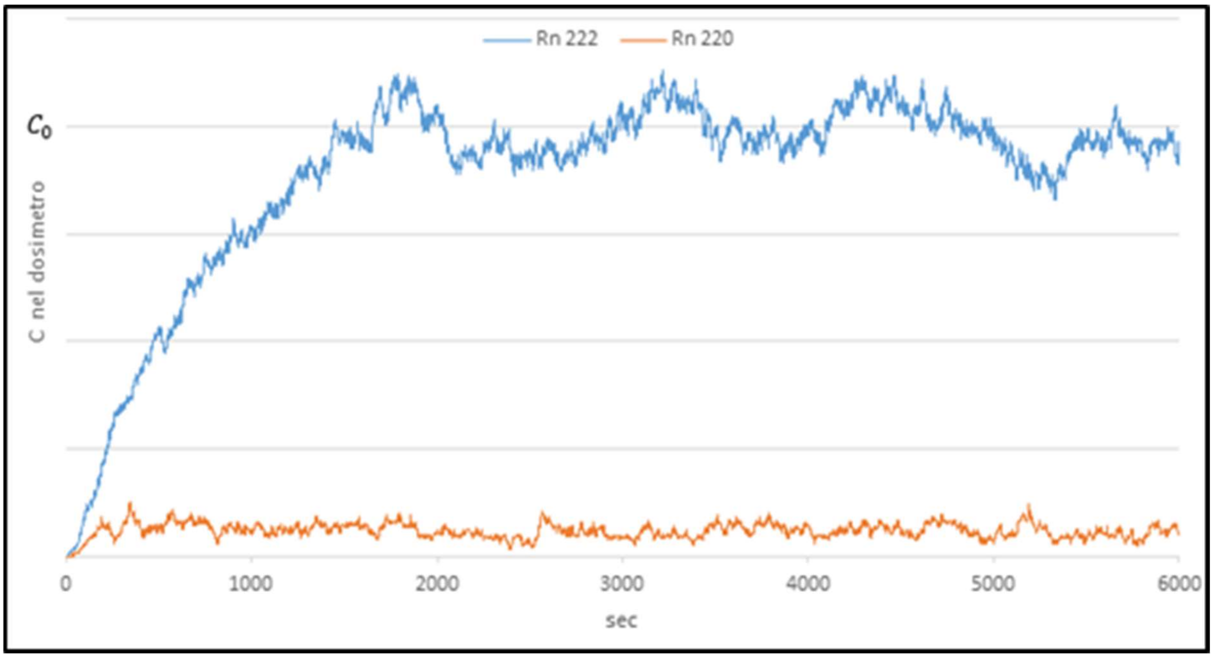

Figure 5 – Result of the Monte Carlo simulation for radon and thoron. A small but not negligible quantity of thoron enters into the dosemeter

Results and discussion

In the following Table 1 the results of the experiment (exposure for 121 hours) are shown, expressed as the number of the alpha tracks detected per square centimeters. Four sets of 5 CR-39 detectors were analyzed: one set of unexposed detectors, in order to check the blank of the CR-39 lot utilized in the experiment, one set of CR-39 coming from the ARPA dosemeters, one set of CR-39 coming from the Radout™ dosemeters and the last one set of Cr-39 coming from a Radout™, inserted in the same a polyethylene bag used by the ARPA dosemeter.

Table 1 – Experimental results. Tracks/cm^2 detected

	Background CR39	ARPA Dosemeter	Radout™ Dosemeter	Radout™ + polyethylene
1	23	175	1230	117
2	11	146	821	83
3	18	173	323	144
4	13	157	526	114
5	15	278	1564	85
Average	16±5	186±53	893±507	109±25

All the the sets of exposed CR-39 show values well above the background, confirming the fact that also thoron can be detected by standard radon dosemeters. These experimental results allows the calculation of a "Thoron Calibration Factor" (CF_{Rn220}) for each type of dosemeter, simply dividing the thoron exposure value calculated by means of the Radon Scout measurements by the track densities (T):

$$CF_{Rn220} = \frac{\int_0^{\tau} C(t)dt}{T} \quad (4)$$

where $C(t)$ is the thoron concentration inside the chamber during the time of exposure which lasted τ=121 hours. The results of these calculations are summarized in Table 2, together with the experimental evaluation of the fraction of thoron able to enter in the different types of devices. The thoron exposure was evaluated in 2468 kBqhm^{-3}.

Table 2 – Thoron Calibration Coefficients and thoron sensitivity of the radon detectors

	ARPA Dosemeter	Radout™ Dosemeter	Radout™+ polyethylene
Thoron Net Tracks Tracks/cm^2	170±53	877±507	93±25
Calibration Coefficient kBq·h·m^{-3}/tracks·cm^{-2}	14.53	2.81	26.65
C/C_0 experimental %	2.2±0.7	10.1±5.9	1.0±0.3
C/C_0 theoretical analytical %	-	4.3	-
C/C_0 theoretical Monte Carlo %	-	4.5	-

Conclusions

The experimental data have shown a significant permeability to thoron of all the tested dosemeters. Moreover, the obtained results permitted the calculation of a specific thoron Calibration Coefficient for all the radon dosemeters. This achievement allows a quite accurate evaluation of the thoron contribution and eventually could also permit the subtraction of the interfering thoron signal in those environments where the occurrence of the shorter lived isotope of radon isn't negligible. The Radout™ dosemeter resulted the most sensitive to thoron: accordingly with the experimental data (see Table 2) about 10% of the thoron present in the ambient air seems able to enter in the dosemeter chamber, a value even greater with respect with the theoretical predictions (4.3 – 4.5 %). These values are not negligible, considering the fact that the exposure to thoron of a dosemeter installed close to the wall could be relevant also for an apparent thoron free environment (see the profiles in Figure 1).
Much lower values (1-2%) were found for the ARPA dosemeter and for the Radout™ dosemeter put inside a polyethylene bag as for the ARPA Piemonte dosemeter: the thin polyethylene foil proved to be quite effective

to slow down the diffusion of the thoron atoms, allowing an almost complete decay before reaching the core of the dosemeters.

References

1. Antonopoulos-Domis et al., Diffusion model of radon exhalation rates, Health Physics, 75 (5) : 574 – 580.
2. Bigu J., A method for measuring thoron and radon gas concentrations using solid-state alpha-particle detectors, International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes, Vol. 37, Issue 7,1986, pp. 567-573.
3. Carrera G., Garavaglia M., Magnoni S., Valli G., Vecchi R., Natural Radioactivity and Radon Exhalation in Stony Materials, Journal of Environmental Radioactivity, Vol. 34, N°2, pp. 149-159, 1997.
4. Cosma C., Cozar O., Jurcut T., Baciu C., Pop I., Proceedings of the Conference: The Natural Radiation Environment VII: Seventh International Symposium on the Natural radiation Environment, Rhodes, Greece, 22-24 May 2002, (2005).
5. Cothern C. R. and Smith J. E. edited by, Environmental Radon, Plenum Press, New York and London, 1987.
6. ISTISAN Rapporto 17/36, Natural radioactivity in building materials in the European Union: a database of activity concentrations, radon emanations and radon exhalation rates, ISS-Ambiente e Salute, 2014.
7. Jonassen N., The determination of radon exhalation rates, Health Physics, Vol. 45, N°2, pp. 369-376, 1986.
8. Magnoni M., Chiaberto E., Prandstatter A., Serena E e Tripodi R, Thoron exhalation rate in stony materials: a simplified approach, Construction and Building Materials, 173, (2018), 520-524.
9. D. Nikezić,N. Stevanović, Radon progeny behavior in diffusion chamber, Nuclear Instruments and Methods in Physics Research Section B 239 (2005) 399-406
10. Nuccetelli C., Bochicchio F., The Thoron Issue: Monitoring Activities, Measuring Techniques and Dose Conversion Factors, Radiat Prot Dosimetry (1998) 78 (1): pp. 59-64.
11. Pressyanov D, Georgiev S., Dimitrova I., Mitev K., Boshkova T., Determination of the diffusion coefficient and solubility of radon in plastics, Radiation Protection Dosimetry, Volume 145, Issue 2-3, May 2011, pp.123-126.
12. Sarad® GmbH, Thoron Scout Radon & Thoron measurements, https://www.sarad.de/product-detail.php?lang=en_US&catID=&p_ID=61.
13. Silverman, M.P. and Mudvari, A. Brownian Motion of Radioactive Particles: Derivation and Monte Carlo Test of Spatial and Temporal Distributions, World Journal of Nuclear Science and Technology, 2018, 8, 86-119.
14. L. Tommasino, J. Chen, R. Falcomer, M. Janik, R. Kanda , F. DeFelice, F. Cardellini, R. Trevisi, F. Leonardi, M. Magnoni, E. Chiaberto, G. Agnesod, M. Faure Ragani, G. Espinosa, J. Golzarri, K. Kozak and J. Mazur, An international cooperation by using an all-encompassing passive radon monitor, Radiation Protection Dosimetry, Volume 177, Issue 1-2, November 2017, pp. 12-15.