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Metrological aspects of international intercomparison of passive radon detectors under field conditions in Marie Curie's tunnel in Lurisia

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Abstract. In 2014, an intercomparison exercise of passive radon detectors under field conditions in the Marie Curie's tunnel belonging to the Lurisia spas complex (Lurisia, Piedmont, Italy) has been held. Radon activity concentration in the tunnel was measured with six radon active monitors, previously calibrated at ENEA-INMRI facilities. In the present paper, a synthesis of the metrological aspects of the intercomparison is given. Indeed particular attention was paid to metrological characterization of radon monitors and their response upon ambient conditions. Correction factors have been defined to be applied when measurements are performed in severe environmental conditions. In particular, it has been found that monitors are particularly sensitive to the effect of air density: the AlphaGUARD (AG-SAPHYMO, GmbH) efficiency decreases with the air density, while for the MR1 PLUS (Tesys, Italy), the opposite applies. When the reference monitors were placed into the Marie Curie's tunnel, to the recorded average radon concentrations correction factors were applied. After the correction the difference between data coming from AG and MR1 PLUS is within the 1.7%.

Key words: active monitors • calibration • intercomparison • radon

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Introduction

In 2014, the international intercomparison of passive radon detectors under field conditions in the Marie Curie's tunnel (Lurisia, Piedmont, Italy) has been held to give radon measurements services and laboratories the possibility to test their passive systems under field conditions, which are less controlled and much more challenging. Radon in the tunnel was monitored by six radon active monitors, previously calibrated at the ENEA-INMRI facilities. The monitors were three MR1 PLUS (Tesys, Italy) based on a scintillation cell, and three AlphaGUARD (AG-SAPHYMO, GmbH), based on a ionization chamber.

In the present paper, a synthesis of the metrological aspects of the intercomparison is given, with particular attention to Radon Reference Measuring System (RRMS) operating at ENEA-INMRI; quality control system of RRMS; dependence of monitors response upon ambient conditions; calibration procedure of monitors used in the intercomparison.

The primary radon reference measurement system operating at ENEA-INMRI

The ENEA-INMRI RRMS is shown in Fig. 1. It is used to calibrate all the reference monitors of the ENEA-INMRI. It consists of an aluminium flask whose volume is 112 litres, one radon monitor and

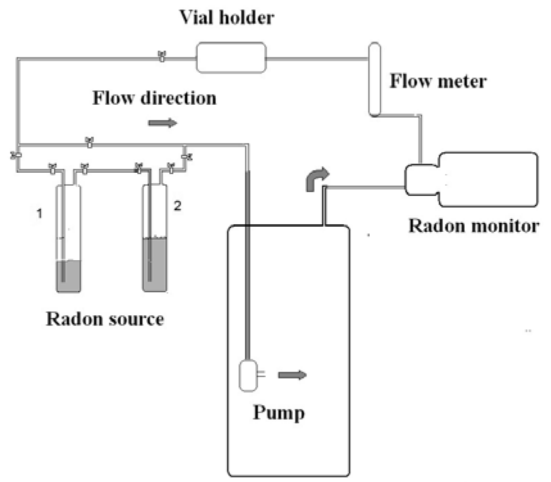

Fig. 1. Primary Radon Reference Measurement System (RRMS) operating at ENEA-INMRI.

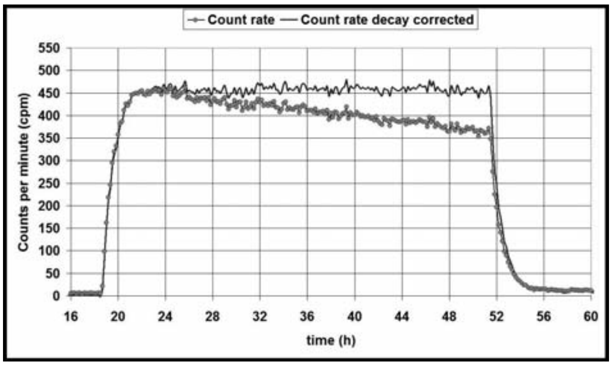

Fig. 2. Example of measurement for calibration of MR1 PLUS radon monitor in RRMS: the actual counting rate and after the correction for the radon decay.

a group of radon sources. All the elements of this system are connected in a closed circuit in which the air can circulate by the aid of a micro-pump placed in the aluminium flask. In the circuit, there are also a flowmeter, a manometer, a vial-holder and an hygrometer.

The system is calibrated using two bubblers (BBLs) containing a ^{226}Ra reference solution of certified activity (about 1500 Bq), referred to an NIST standard (National Institute of Standards and Technology, USA) [1]. The radon activity inside the sources is in equilibrium with the activity of the radium solution. The BBL containing the radium solution is put in position no. 2 (see Fig. 1), while the BBL in the position no. 1 humidifies the air and avoids drying of the radium source. The air circulation through the bubblers extracts radon from the radium source and forces it into the circuit, giving rise to a radon reference atmosphere. In fact, because the radon gas is in equilibrium with radium solution and the volume of the circuit is carefully measured, it is possible to know precisely the radon activity concentration. The efficiency of the monitor is thus simply evaluated by comparing the count rate of the monitor with the value of the radon activity concentration. Moreover, when the circuit is calibrated with standard sources, it can be used to measure the activity of other radon sources of unknown activity. This can be achieved by simply comparing the count rate given by the reference bubbler with the one produced by the radon from the unknown source. This procedure can also be used to validate ENEA-INMRI bubblers by comparison with radon activity sources certified by other international institution. More details are given in the following paragraph.

The RRMS calibration procedure consists in three steps. First, background measurements are collected for some hours (from 5 to 20 h), in order to record eventual residual radon in the circuit and the instrumental background. Second, radon is extracted from the radium source and transferred to the circuit by air circulation through the BBLs for 40 min. Finally, BBLs are bypassed and the measurement goes on for more than 24 h to achieve a good statistic. During this time, radon decays into the circuit.

Result of a typical measurement for the calibration of MR1 PLUS monitor is showed in Fig. 2: the actual counting are corrected for radon decay at the time in which BBL were closed after radon extraction. If no radon leakage occurred, an horizontal line is obtained.

Two ENEA-INMRI monitors, one AG and one MR1 PLUS, have been calibrated independently in the ENEA-INMRI RRMS. The monitors are connected to the circuit by two specific adapters made in ENEA: the adapters are cylinders of about 40 cm^3 and one base of the cylinder is hermetically connected to the input filter of the monitor. Radon goes from the adapter to the monitor by diffusion, as it occurs in the typical use of the monitor. The adapter for AG is made in Teflon to avoid perturbation of the electric field in the ionization chamber of the instrument.

After calibration, the performances of two monitors were compared in more than 20 experiments executed in the 1 m^3 ENEA-INMRI radon chamber and their responses were found, on an average, perfectly overlapped. The results are summarized in Table 1: on an average, the results had a low standard deviation, about 1.4% as coefficient of variation. This value represents the reproducibility limit of the radon measurements performed with these instruments.

The quality control of the primary radon reference measurement system

The RRMS is a relative measurement system, that is, calibration factors are based on the count rate produced by the radon extracted from a reference radium solution. The most critical part of the calibration system is thus the actual activity of the reference radium sources and the difficulty of extracting the whole radon gas from the bubblers. In order to control these aspects, three methods are followed:

1) the 'absolute method' using radon sealed in glass vial [2];
2) comparison with reference radon sources of the Czech Metrological Institute (CMI);
3) participation to international intercomparison organized by Consultative Committee for Ionizing Radiation (CCRI) – BIPM.

Table 1. Comparison of the results achieved with the two ENEA-INMRI reference monitors in the 1-m^3 radon chamber

Test no.	AG [Bq/m^3]	MR1 PLUS [Bq/m^3]	AG/MR1 PLUS
508	4 126	4 081	1.011
509	4 075	4 115	0.990
512	8 996	9 170	0.981
513	1 457	1 424	1.023
515	21 372	21 227	1.007
516	20 324	20 415	0.996
518	4 176	4 151	1.006
519	16 844	17 303	0.973
521	44 220	44 900	0.985
522	8 997	9 074	0.992
528	3 985	3 948	1.010
531	9 134	9 008	1.014
532	21 047	21 180	0.994
534	4 156	4 083	1.018
535	76 677	76 254	1.006
536	105 377	105 629	0.998
537	1 435	1 410	1.018
539	9 036	8 984	1.006
550	30 030	30 364	0.989
557	42 983	43 505	0.988
560	4 085	4 000	1.021
		Average	1.001
		St. dev.	0.014
		CV [%]	1.4

The sources used for 'absolute method' are two glass vial, flame sealed, containing about 5 kBq of radon: their activity was determined by γ spectrometry measurements (in 4π geometry), with a NaI well detector. The efficiency of the γ spectrometer was computed with GEANT3.21 Monte Carlo Code. After γ measurements, the vials were placed on vial holder of the RRMS (see Fig. 1). Vials were broken and the activity of radon was measured. Results are reported in Table 2: the very good agreement between results obtained with the 'absolute method' and those related to the measure with reference bubblers confirms the validity of RRMS.

At ENEA-INMRI, two flow-throw type radon sources produced and certified by Czech Metrological Institute are also available. The activity of these sources was measured with RRMS and the results were compared with the nominal activity certified by the Czech Metrological Institute: the results are in very good agreement as shown in Table 3.

Moreover, in December 2014, ENEA-INMRI participated to the CCRI-BIPM radon intercomparison, in which INMRI received a sealed glass vial containing an unknown activity of radon gas from BIPM. The radon activity in the vial was measured with the RRMS with the same procedure described above, and it was found to be 8200 kBq at the reference date (December 16, 2014; h 11:40 UTC), with an uncertainty of 1% at $k = 1$. The ENEA-INMRI result was positive, in particular the BIPM affirmed that: "Result is on line within the uncertainty with most of the other results obtained for this radionuclide at $k = 2$".

Effects of air density on the radon monitors performance

Generally, the calibrations of the monitors are carried out at 22°C and 1000 mbar, while in Marie Curie's tunnel, the temperature was 11°C and pressure was about 930 mbar. The parameter with the greater influence on the response of AG and MR1 PLUS monitors is the density of the air, that is, the P/T ratio, where P is the atmospheric pressure and T is the absolute temperature.

So, a number of tests under different air pressure and temperatures were performed in ENEA-INMRI laboratory in order to calculate the correction factors when the monitors work in severe environmental conditions. The results of one test are plotted in Fig. 3: in this test, AG and MR1 PLUS monitors were put in a 141 litres radon chamber; a certain amount of radon gas was introduced and the pressure was slightly lowered to 890 mbar at constant temperature: radon activity concentration was monitored for 24 h. Later, the pressure in the chamber was raised to 998 mbar blowing in 'aged air' (without radon) and radon measurements were performed for 24 h more.

It can be observed in Fig. 3 that the AG efficiency decreases as the air pressure decreases, while for

Table 2. Comparison of results obtained with the 'absolute method' and relative method with ENEA-INMRI bubbles (BBLs)

Test no.	Activity measured by γ spectrometry [Bq]	Activity measured by comparison with ENEA-INMRI BBLs [Bq]	Δ [%]
616	5300 ± 60	5310 ± 53	0.19
618	5912 ± 65	5975 ± 60	1.06

Table 3. Comparison between the radon activity certified by the Czech Metrological Institute (CMI) and the one measured by ENEA-INMRI with the relative method with BBLs

^{226}Ra source (Serial number)	Activity certified by CMI [kBq]	Activity measured by comparison with ENEA-INMRI BBLs [kBq]	Δ [%]
RF200 s.n. 524005	187.64 ± 2.80	187.45 ± 1.80	0.1
RF20 s.n. 524003	17.74 ± 0.27	18.00 ± 0.18	1.4

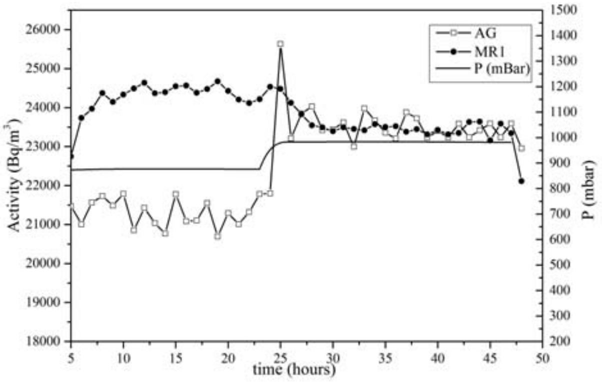

Fig. 3. Measurements of the effect of air density on radon monitors sensibility.

the MR1 PLUS, the opposite occurs. These facts can be easily explained by considering that AG is based on an ionization chamber where the ionization is proportional to the quantity of the atoms in a volume (the density); conversely, for MR1 PLUS, based on a scintillation detector, the decrease in air density allows more α particles to reach the wall of the detector.

A series of tests was carried out with different pressure variations. In Fig. 4, the dependence of efficiency of active monitors is plotted as function of the decreasing 'equivalent pressure' (at 295 K), P_{eq}, as defined in the following equation:

$$P_{eq} = 295\, P/T$$

where T [K] is the absolute temperature at which the measurement is carried out.

Indeed, the relevant parameter is air density, proportional to P/T ratio, so the use of the 'equivalent pressure' allows to apply the proper correction according to the actual environmental conditions.

In the Madame Curie's tunnel (pressure = 931 mbar; temperature = 11°C), the AG monitors underestimated the actual radon concentration of about 2.5%, while the MR1 PLUS monitors overestimated them to about 1.7%. For these reasons, the experimental results achieved during the exposure in Madame Curie's tunnel were corrected in order to take into account for these effects. Average difference between AG and MR1 PLUS in the tunnel was about 5% and was reduced to less than 1% after correction.

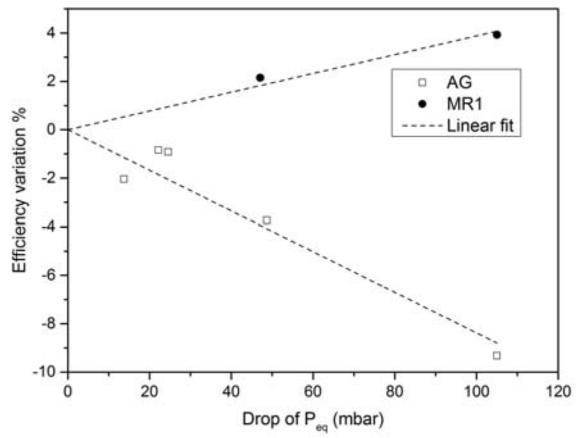

Fig. 4. Dependence of efficiency of active monitors as function of the drop of the equivalent pressure.

Calibration of the monitors used during the Lurisia intercomparison

During the intercomparison, the reference radon concentrations in the Marie Curie's tunnel were measured using six different monitors: three AG and three MR1 PLUS. Two of these monitors were the ENEA-INMRI reference monitors calibrated using RRMS. The other four monitors have been calibrated by comparison with reference monitors at ENEA-INMRI laboratory before the intercomparison. For this scope, 13 calibration tests have been performed. Three tests have been held in the stainless steel 1 m^3 radon chamber, at different radon constant concentrations (1500, 9000 and 25 000 Bq/m^3, respectively): each test lasted about 100 h.

Radon is provided by flow-through type radon sources provided by Pylon or Czech Metrological Institute. Radon sources are connected with the chamber by a closed-loop circuit; a constant air flow of 0.1 l/min transfers radon gas from the source to the chamber, making constant the indoor radon concentration. The activity concentration achieved by each source is reproducible within 1.5–2.0%. The test results are given in Table 4.

Other tests have been done in the ENEA-INMRI 'walk-in' radon chamber (volume 18 m^3), with variable radon concentrations in the range 1500–4000 Bq/m^3. Indeed, in the 'walk-in' radon chamber, a slight indoor depressurization enhance the natural radon emanation from the underneath soil, so the indoor radon activity concentration achieved strictly depends on the meteorological conditions, which are not constant in time and not reproducible. Calibration tests results in the 'walk-in' radon chamber, each one performed for more than 100 h with measurement conditions closer to an in-field situation, showed not spatially uniform radon levels with variation up to 4% in a distance of 1.5 m. In Fig. 5, monitors lectures are shown.

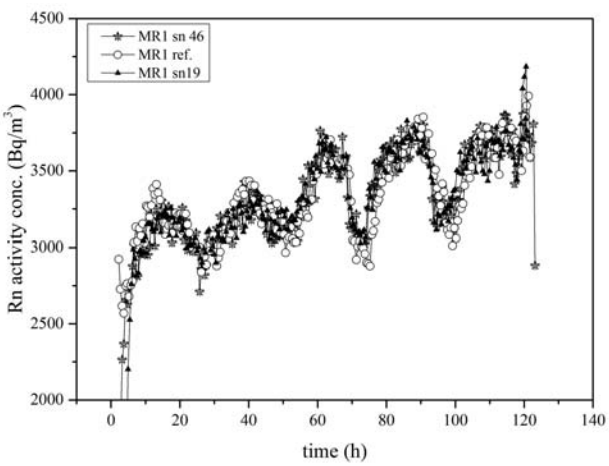

Fig. 5. Measurements for calibration of the monitors in the 'walk-in' radon chamber. Results of one measurement test.

Table 4. Results of calibration tests of active monitors at ENEA-INMRI facilities

Radon concentrations [Bq/m³]	Equivalent pressure P_{eq} [mbar]	AG s.n. 0933	AG s.n. 1312	MR1 PLUS s.n. 19	MR1 PLUS s.n. 46
		Calibration factor			
1 500	991	–	1.027	–	31.580
4 400	989	1.006	–	–	–
3 350	1003	1.000	0.966	30.71	32.710
4 200	992	0.990	0.998	29.35	31.324
3 600–5 600	1003	1.040	1.033	29.266	31.600
4 200	1004	0.970	–	29.06	31.590
9 000	1007	–	0.995	–	–
25 000	998	–	0.996	–	–
2 000–4 000	995	1.040	–	30.087	33.46
	Calibration factor	1.001	1.0025	29.695	31.761
	St. dev.	0.026	0.024	0.687	0.543
	CV [%]	2.6	2.4	2.3	1.7

Table 5. Average concentration of radon in Marie Curie's tunnel during the exposure at high radon level: data before and after correction for the effect of air density

	MR1 PLUS without correction for P_{eq} [kBq/m³]	AG without correction for P_{eq} [kBq/m³]	Δ% without correction for P_{eq}	Δ% with correction for P_{eq}
Site A	23.17	22.17	4.4	0.26
Site D	21.79	20.71	5.2	0.9
Site H	16.15	15.23	6	1.7

The calibration factor of each monitor was computed considering at least five tests; the reproducibility of the calibration factor, in terms of coefficient of variation (CV), ranged from 1.7% to 2.6%, and the overall uncertainty of the calibration factor of monitors was thus estimated in the order of 3.3%. More details about calibration test results are given in Table 4.

Discussion and conclusions

A deep metrological characterization has been done in order to properly calibrate radon monitors to be used in the international intercomparison of passive radon detectors under field conditions carried out in the Marie Curie's tunnel in Lurisia (Piedmont, Italy). The metrological characterization has been carried out at the ENEA-INMRI facilities: especially the effect of air density on radon monitors has been evaluated in order to define correction factors to apply when measurements are done in severe environmental conditions. In particular, it has been found that the AG efficiency decreases with the air density, while for the MR1 PLUS, the opposite occurs.

Indeed, the average radon concentrations, recorded by the reference monitors (one AG and one MR1 PLUS) placed in three different positions of the tunnel during the intercomparison, were very different before correction for the effect of air density; they differed on an average of about 5%. As shown in Table 5, after the application of the correction factor, the difference between data coming from AG and MR1 PLUS was within 1.7%.

References

1. De Felice, P., & Myteberi, X. (1996). The ^{222}Rn reference measurement system developed at ENEA. *Nucl. Instrum. Methods Phys. Res. Sect. A-Accel. Spectrom. Dect. Assoc. Equip.*, *369*(2/3), 445–451.
2. Picolo, J. L. (1996). Absolute measurement of radon 222 activity. *Nucl. Instrum. Methods Phys. Res. Sect. A-Accel. Spectrom. Dect. Assoc. Equip.*, *369*(2/3), 452–457.