#### International comparison of activity measurements of radon 222 <u>CCRI(II)-K2.Rn-222</u>

By S. Pierre<sup>1</sup>, P. Cassette<sup>1</sup>, C. Fréchou<sup>1</sup>, A. Antohe<sup>2</sup>, F. Cardellini<sup>3</sup>, M. Capogni<sup>3</sup>, M.-R. Ioan<sup>2</sup>, F. Juget<sup>4</sup>, B.J. Kim<sup>5</sup>, J.M. Lee<sup>5</sup>, K.B. Lee<sup>5</sup>, A. Luca<sup>2</sup>, M. Sahagia<sup>2</sup>.

<sup>1</sup>Laboratoire National de métrologie et d'Essais - Laboratoire National Henri Becquerel, Gif-sur-Yvette, France <sup>2</sup>National Institute of Research and Development for Physics and Nuclear Engineering - "HoriaHulubei" - IFIN-HH, Bucharest, Romania <sup>3</sup>ENEA-Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti (INMRI) Roma, Italy <sup>4</sup>Institut de Radiophysique, Lausanne, Switzerland <sup>5</sup>Korea Research Institute of Standards and Science, Yuseong, Daejon, Republic of Korea

#### Abstract:

An international comparison of activity measurements of radon-222, CCRI(II)-K2.Rn-222, was organised in 2015 by the LNE-LNHB. Samples of this gas provided by the LNE-LNHB were measured using different techniques. The PMM (Power Moderated weighted Mean) formalism was applied [1].

## 1. Introduction

Radon-222 is a radioactive noble gas decaying through alpha transitions to short half-life progenies and is one of the main sources of natural radioactivity. It is monitored with commercial instruments to evaluate radon activity concentration in individual rooms, water or soil. National activity standards of radon-222 are available in several countries and comparison of these standards is necessary to ensure the international traceability of this radionuclide and to support the CMC's of the National Metrology Institutes (NMIs).

An international comparison of activity measurements of radon-222 was organised under the auspices of the Consultative Committee for Ionizing Radiation CCRI(II) in 2015: comparison CCRI(II)-K2.Rn-222. Samples were made available to the participants by the LNE-LNHB.

Activity measurements can be carried out using different primary methods, such as the defined solid angle method ([2, 3, 4]) and the  $4\pi\gamma$  integral counting method [5]. The first method was used by two laboratories. The latter was applied by one laboratory, by using a well-type NaI(Tl) 5"x5" detector to standardize a radon-222 source in glass ampoule which, in turn, was used to calibrate a Scintillation Cell (SC). This SC detector was also calibrated with radon-222 extracted from a standard Ra-226 solution available in the same laboratory. The SC detector was then used for the comparison. Another laboratory used LSC method described by P. Cassette in P. Cassette et al. [6] Sahagia et al [7, 8]. Five results were obtained.

#### 2. Relevant information about the comparison

The list of the participating institutions with information on the people who carried out the measurements is shown in Table 1.

ENEA	Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti (INMRI), Roma, Italy (Francesco Cardellini, Marco Capogni)				
IFIN-HH	National Institute of Research and Development for Physics and Nuclear Engineering – "HoriaHulubei" - IFIN-HH, Bucharest, Romania (Maria Sahagia, Aurelian Luca, Andrei Antohe, Mihail-Razvan Ioan))				
IRA	Institut de Radiophysique, Lausanne, Switzerland (F. Juget)				
KRISS	Korea Research Institute of Standards and Science, Yuseong, Daejon, Republic of Korea (B.J. Kim, J.M. Lee, K.B. Lee)				
LNE-LNHB	Laboratoire National de métrologie et d'Essais - Laboratoire National Henri Becquerel, sur-Yvette, France (P. Cassette, S. Pierre)				

#### Table 1 – List of participants

Metal containers with radon-222 at low pressure were prepared and dispatched to the participants by the LNE-LNHB. Table 2 gives information about the volume of the containers.

Laboratory	Container number	Volume (cm <sup>3</sup> )	absolute uncertainty (cm <sup>3</sup> , k=1)	relative uncertainty (%, k=1)
IFIN-HH	GAZ 3	104.9287	0.4	0.4 %
KRISS	GAZ 5	104.7086	0.4	0.4 %
<b>ENEA-INMRI</b>	GAZ 7	104.8654	0.4	0.4 %
IRA and LNE-LNHB	GAZ 10	104.7571	0.4	0.4 %
mean		104.81		
standard deviation		0.10		
relative standard deviation		0.1 %		

Table 2 – Containers volume

As far as the standard uncertainty on each measurement is 0.4%, whereas the relative standard deviation between all containers is 0.1 %, one can conclude that all the containers have the same volume, within the measured uncertainties. The used preparation protocol ensured the homogeneity of the filling of the containers (evaluated RSD of 0.1%). The half-life value  $T_{1/2} = 3.8232$  (0.0008) d [9] was used and the results were evaluated on the reference date 1<sup>st</sup> of July 2015 12:00 UTC.

Table 3 provides the list of the methods used, together with the laboratories who applied these methods. Acronyms of the methods are also given according to the CCRI(II) rules.

Method acronym Number of times used	Description of the method	Laboratories using this method
SA-PS-AP-00-00-00	Alpha Counting in Defined Solid Angle	LNE-LNHB, KRISS
4P-LS-MX-00-00-TD UA-GH-GR-00-00-00 4P-IC-MX-00-00-00, both calibrated by 4P-LS-MX-00-00-TD	Arithmetic mean of the value obtained by : - Absolute measurement by liquid scintillation counting of an ampoule containing an amount of Radon transferred in a quantitative mode from the gas container (see. P.Cassette et al., ARI 64 (2006) 1465-1470; Sahagia et al., ARI 68 (2010) 1505-1506) - relative measurements of a gas vial, using the calibration figures for the HPGe and Ionisation chamber (Sahagia et al. NIM A631(2011)73-79)	IFIN-HH
4P-IC-GR-00-00-00 calibrated by SA-PS-AP-00-00-00	Glass ampoule measured with an ionisation chamber which was calibrated with a standardized ampoule using the defined solid angle method	IRA
4P-SC-AP-00-00-HE calibrated by 4P-NA-GR-00-00-HE	Glass ampoule measured by Scintillation Cell previously calibrated for Rn-222 by $4\pi\gamma$ -integral counting method [5]	ENEA-INMRI
4P-SC-AP-00-00-HE calibrated by standard Ra-226 solution	Glass ampoule measured by Scintillation Cell previously calibrated for Rn-222 by standard Ra-226 solution	ENEA-INMRI

Table 3 – List of the methods used

IRA had a problem during their first measurement with container number 4. So a second measurement was performed using container number 10, previously measured at LNE-LNHB. As the method used by LNE-LNHB allowed several measurements, it was possible to measure the sample a second time.

IFIN-HH made three measurements with different methods and gave the mean as their result.

## 3. Results and evaluation

Five laboratories took part in this comparison providing 5 results in Bq, as shown in Table 4 that also presents the combined uncertainty. Table 4 - Results evaluated on 1<sup>st</sup> of July 2015 12 h UTC in Bq.

Laboratory	Activity (Bq)	Combined standard uncertainty (Bq)	Combined relative standard uncertainty (%, k=1)
ENEA-INMRI Scintillation Cell calibrated by 4πγ- integral counting method	74 152*	742	1.0
ENEA-INMRI Scintillation Cell calibrated by standard Ra-226 solution	74 196	816	1.1
IFIN-HH	73 680 <sup>§</sup>	1300	1.8
IRA 4P-IC-GR-00-00-00	65 990	303	0.46
KRISS SA-PS-AP-00-00-00	72 240	120	0.17
LNE-LNHB SA-PS-AP-00-00-00	71 930	350	0.50

\* selected as reference value for the comparison.

<sup>§</sup> mean of 3 values.

Table 5, Table 6, and Table 7 present the associated uncertainties as provided in the reporting forms of the participants for all individual parameters included in the measurement process.

Table 5 – Uncertainty components	in % of the	e activity concent	tration for $k = 1$
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Uncertainty components	Laboratory Method							
	Scintillat	ENEA-INMR ion cell calibrate Ra-226 solutio	tI ed by standard on	ENEA-INMRI Scintillation Cell calibrated by 4πγ-integral counting method				
	Relative value / (%)	Evaluation type	Sensitivity factor	Relative value / (%)	Evaluation type	Sensitivity factor		
counting statistics	0.0019	А		0.0017	А			
background	6	А		6	А			
Detection efficiency	1.1	В		1.1	В			
Decay correction during measurement	0.0016	В		0.0016	В			
Decay correction during background	0.00012	В		0.00012	В			
Repeatability	4	А		5	А			
Reproducibility	5	А		10	А			
Geometry factor	0.003	В		0.002	В			
Total uncertainty	1.1			1.0				

Uncertainty components	Laboratory Method					
	IFIN-HH 4π(LS)β TDCR			IRA 4P-IC-GR-00-00		
	Relative value (%)	Uncertainty type	Sensitivity factor	Relative value (%)	Uncertainty type	Sensitivity factor
counting statistics	0.8	А	1			
background	0.3	В	0.005	0.027	А	
Detection efficiency	0.2	В	1			
Decay correction During measurement	0.1	В	0.001	0.021	В	
Decay correction During background	0.1	В	0.001			
Repeatability	0.3	А	1	0.040	В	
Reproducibility						
Geometry factor						
Decay correction between ref date and measurement date	0.15	В	1	0.015	В	
Current of Rn ampoule				0.041	В	
Ionization chamber calibration factor				0.403	В	
Decay of Caesium reference source				0.035	В	
Current of Caesium source				0.055	В	
Correction for <sup>214</sup> Po decay	0.15	В	0.03			
Accumulation of <sup>210</sup> Pb during the measurement	0.5	В	1			
Transfer activity loss*	1.41	В	1	0.200	В	
Total uncertainty	1.8			0.460		

**Table 6** – Uncertainty components, in % of the activity concentration for k = 1

\*Combined uncertainty of two successive transfer operations

Uncertainty components	Laboratory Method							
	S.	KRISS SA-PS-AP-00-00-00			LNE-LNHB SA-PS-AP-00-00			
	Relative value / (%)	Uncertainty type	Sensitivity factor	Relative value / (%)	Uncertaint y type	Sensitivity factor		
counting statistics	0.058	А		0.07	А	1		
background	2.7	А		3	А	1.13 10-7		
Detection efficiency								
Decay correction during measurement	1.3 10-5	В		2.7 10-4	В			
Decay correction during background	2.6 10-9	В		0.04	В			
Repeatability	0.05	В			В			
Reproducibility	0.1	В		0.2	В	1		
Geometry factor	9.9 10-2	В		0.4	В	1		
Spectrum analysis	6.7 10-2	В			В			
Correction due to residual in cylinder container	7.6 10-4	В			В			
Total uncertainty	0.17			0.5				

**Table 7** – Uncertainty components, in % of the activity concentration for k = 1

A Grubb test was performed and showed no suspicious value (Table 8).

Table 8 –	Grubbs	test
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Nb of results	5
Gmin	1.714
Gmax	0.780
Critical value at 5%	1.715
Critical value at 1%	1.764
test	correct

The Figure 1 displays all final results in Bq.



Figure 1 – Individual results of the international comparison of activity (5 individual results) in Bq, arithmetic mean, median, weighted mean and PMM *value*.

The values of the arithmetic mean, median, weighted mean, and PMM value are in Table 9.

	A (Bq)	u (Bq)
Median	72240	1368
PMM value	71543	1474
Arithmetic mean	71598	1463
Weighted mean	71511	105

Table 9 – Median, PMM value, arithmetic mean and weighted mean.

Following a recommendation of the KCWG(II), the Power-Moderated weighted Mean formalism (PMM) [1] is used (**Figure 2**). The PMM value is 71 500 (1 500) Bq.

## 4. Conclusion

Four results are in a fair agreement, even if the uncertainties are quite different (by almost an order of magnitude). One value seems to be outside of the observed results but the Grubbs test does not allow to conclude that it can be considered as an outlier, even if it is difficult to use statistical tools on a so small sample of results.

#### 5. Acknowledgments

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