



HAL
open science

International comparison of activity measurements of radon 222

Sylvie Pierre, Philippe Cassette, Benoit Sabot, Carole Fréchou, A. Antohe, C. Barna, P. Blahusiak, Francesco Cardellini, R. Dersch, A. Honig, et al.

► **To cite this version:**

Sylvie Pierre, Philippe Cassette, Benoit Sabot, Carole Fréchou, A. Antohe, et al. International comparison of activity measurements of radon 222: EURAMET Project n°1475 – EURAMET.RI(II)-S8.Rn-222. *Metrologia*, 2021, 58 (1A), <https://doi.org/10.1088/0026-1394/58/1A/06015>. 10.1088/0026-1394/58/1A/06015 . cea-03326177

HAL Id: cea-03326177

<https://cea.hal.science/cea-03326177>

Submitted on 25 Aug 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

International comparison of activity measurements of radon 222
EURAMET Project n°1475 – EURAMET.RI(II)-S8.Rn-222

S. Pierre¹, P. Cassette¹, B. Sabot¹, C. Fréchet¹, A. Antohe², C. Barna², P. Blahušiak³, F. Cardellini⁴, R. Dersch⁵, A. Honig⁵, F. Juget⁶, M. Krivošík³, A. Luca², F. J. Maringer⁷, F. Mertes⁵, S. Röttger⁵, M. Sahagia², J. Slučiak³, M. Stietka⁷, L. Szűcs⁸, C. Teodorescu².

¹CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), 91191 Gif-sur-Yvette, France,

²Horia Hulubei National Institute for Research and Development in Physics and Nuclear Engineering - IFIN-HH, Bucharest-Magurele, Romania,

³Slovenský metrologický ústav, Bratislava, Slovakia,

⁴Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti, Roma, Italy,

⁵Physikalisch-Technische Bundesanstalt, Braunschweig, Germany,

⁶Institut Universitaire de Radiophysique Appliquée, Lausanne, Switzerland,

⁷Bundesamt für Eich- und Vermessungswesen, Wien, Austria,

⁸Budapest, Főváros Kormányhivatala, Budapest, Hungary.

Abstract

An international comparison of activity per unit volume of ²²²Rn (EURAMET project N° 1475, registered in KCDB as supplementary comparison EURAMET.RI(II)-S8.Rn-222) was organised in 2019 within the framework of the European project MetroRADON. The LNE-LNHB is the pilot of the comparison. Samples of ²²²Rn gas provided by the LNE-LNHB were sent to the participants and were measured using various techniques. The reference value is the power moderated mean (PMM) (Pommé, 2013) of the seven submitted results.

1. Introduction

²²²Rn is a radioactive noble gas decaying through alpha transition to short half-life progenies and is one of the main sources of natural radioactivity. It can be monitored with commercial instruments to evaluate radon activity concentrations in individual rooms, water or soil. National standards of ²²²Rn are available from National Metrology Institutes (NMIs) and comparisons of these standards are necessary to ensure the traceability of this radionuclide and to support the CMC's of the NMIs and Designated Institutes (DIs).

This comparison of activity per unit volume of ²²²Rn was organised within the framework of the MetroRADON project under the auspices of EURAMET. Samples were made available to the participants by the LNE-LNHB.

The reference measurement method is Defined Solid Angle alpha counting (DSA) with a frozen disc source of ²²²Rn which is a primary measurement method. This method was used by two laboratories. Other methods such as comparison by a scintillation cell with ²²²Rn extracted from standard ²²⁶Ra solution or comparison by a scintillation cell with ²²²Rn in glass ampoules previously measured with an NaI detector, were used by other participants. Another laboratory used a LSC method (Cassette, 2006), (Sahagia, 2010), (Sahagia, 2011).

2. Relevant information about the comparison

The list of the participants is given in Table 1. IRA (Switzerland) received a sample too. Unfortunately, when IRA started the measurement, a problem happened with the vacuum pump, so the measurement could not be completed.

Table 1: List of participants

BEV	Bundesamt für Eich- und Vermessungswesen, Austria
BFKH	Budapest Főváros Kormányhivatala, Budapest, Hungary
ENEA	Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti (INMRI), Italy
IFIN-HH	Horia Hulubei National Institute for Research and Development in Physics and Nuclear Engineering, Romania
LNE-LNHB	Laboratoire National de métrologie et d'Essais - Laboratoire National Henri Becquerel, France
PTB	Physikalisch-Technische Bundesanstalt, Germany
SMU	Slovenský metrologický ústav, Slovakia

Metal cells containing ^{222}Rn at low pressure (below 10^{-3} hPa) were prepared by the LNE-LNHB and dispatched to the participants. A container (105 cm³) and its dimensions can be seen in

Figure 1 and

Figure 2. Each volume is measured by comparison with a certified volume, as described in Section 4.2 of (Sabot, 2020).



Figure 1: Picture of a container

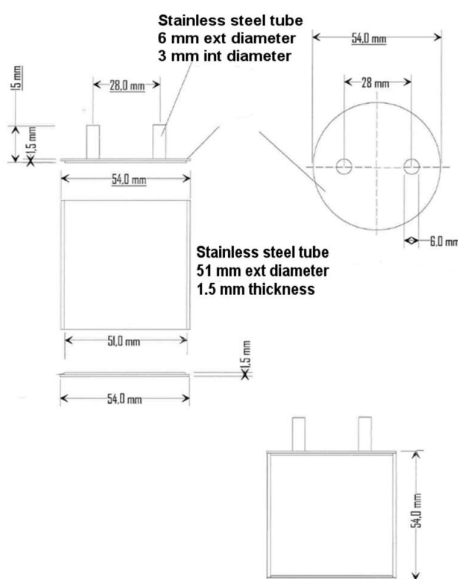


Figure 2: Dimensions of the containers

Table 2 gives information about the volume of the containers.

The relative standard uncertainty on each volume measurement is less than 0.4%, and the relative standard deviation of the volume of all containers is 0.4%. This uncertainty of 0.4% was quadratically added to the uncertainty of the activity given by the participants to calculate the uncertainty on the activity concentration.

Table 2: Container volumes

Laboratory	Container number	Volume (cm ³)	Absolute standard uncertainty (cm ³)	Relative standard uncertainty (%)
PTB	GAZ 1	105.84	0.31	0.30
BFKH	GAZ 2	104.37	0.44	0.42
BEV	GAZ 3	104.93	0.44	0.42
IRA	GAZ 6	104.70	0.46	0.44
IFIN-HH	GAZ 7	104.87	0.41	0.39
ENEA-INMRI	GAZ 8	105.29	0.41	0.39
SMU	GAZ 9	105.03	0.42	0.40
LNE-LNHB	GAZ10	104.76	0.42	0.40
Mean		104.97		
Standard deviation (cm ³)		0.44		
Relative standard deviation		0.41%		

LNE-LNHB divides the activity given by each laboratory by the appropriate volume to calculate the activity per unit volume. The homogeneity of the filling of the containers was verified by gamma-ray spectrometry using an HPGe detector, to measure the activity of the radon daughters. The relative standard deviation of the results from these measurements of the daughter products was 0.3%, so the containers are considered homogeneous.

The half-life value $T_{1/2} = 3.8232$ (8) d (Bé, 2008) was used and the results were evaluated on the reference date 15 April 2019 at 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 (BIPM-2).

Table 3: List of the methods used as reported by participants

Laboratory	Method Acronym	Description of the method
BEV	4P-IC-AP-00-00-00	Exhalation of the ²²² Rn activity into the air of a traceable volume calibration container and measurement of the ²²² Rn activity concentration in air by a traceable calibrated secondary standard ²²² Rn ionisation chamber.
BFKH	4P-IC-AP-00-00-00	The measurements were carried out with two AlphaGUARD radon measuring instruments. One of them was calibrated by the PTB, the other has a manufacturing calibration.
ENEA-INMRI	4P-SC-AP-00-00-HE calibrated by 4P-NA-GR-00-00-HE	Glass ampoule measured by Scintillation Cell previously calibrated for ²²² Rn by a 4πγ-integral counting method.
	Arithmetic mean of the values obtained by:	
IFIN-HH	4P-LS-MX-00-00-TD	Absolute measurement by liquid scintillation counting of an ampoule containing an amount of ²²² Rn transferred in a quantitative mode from the gas container (Cassette, 2006) (Sahagia, 2010).
	UA-GH-GR-00-00-00 4P-IC-MX-00-00-00, both calibrated by 4P-LS-MX-00-00-TD	Relative measurements of a gas vial, using the calibration figures for the HPGe and Ionisation chamber (Sahagia, 2011).
LNE-LNHB	SA-PS-AP-00-00-00	Alpha Counting in Defined Solid Angle.
PTB	SA-PS-AP-00-00-00	Alpha Counting in Defined Solid Angle.
SMU	4P-IC-AP-00-00-00	Continual monitoring of a volume activity of ²²² Rn in a radon chamber with a volume of ~ 1 m ³ with AlphaGUARD DF2000 in diffusion 60 min mode.

2.1. The Italian ^{222}Rn Reference Measurement System at ENEA INMRI

The INMRI radon reference measurement system (RRMS) for radon in air is shown in **Figure 3**.

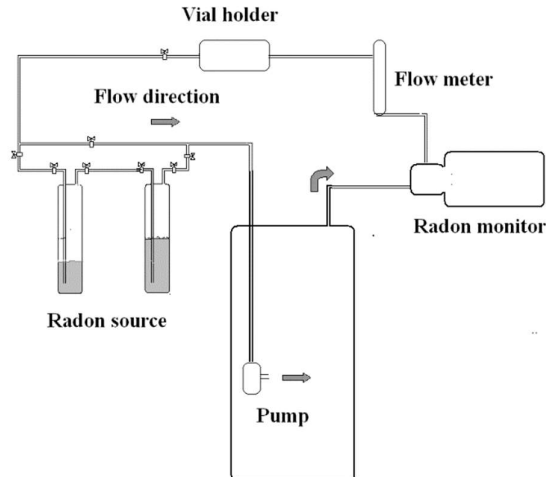


Figure 3 – Primary ^{222}Rn Reference Measurement System operating at INMRI-ENEA.

It is used to measure the activity of ^{222}Rn sources and to calibrate all the reference monitors of the INMRI (De Felice, 1996) (Cardellini, 2016). It consists of an aluminium flask whose volume is 111.77 L (the larger volume in the figure), one radon monitor based on a Lucas Scintillation Cell and a group of ^{222}Rn sources. All the elements of this system are connected in a closed circuit in which the air can circulate by means of a micro-pump placed in the aluminium flask. In the circuit are also a flow-meter, a manometer, a vial-holder and a hygrometer. The exact configuration of the radon source assembly (labelled as “radon source” in the figure) depends on the measurement that is carried out: in some experiments, the source to be measured is in the left bubbler, while the right bubbler is only a trap; in other measurements, the radon source is in the right bubbler and the left bubbler contains only water. The bubbler may be easily removed and replaced by any other suitable vessel containing radon.

The system is calibrated using bubblers (BBL) containing a ^{226}Ra reference solution of known activity, referred to a NIST standard (National Institute of Standards and Technology, USA) (De Felice, 1996). The ^{222}Rn activity inside the sources is in equilibrium with the activity of the ^{226}Ra solution. The BBL containing the ^{226}Ra solution is put on the right (see **Figure 3**), while the BBL on the left humidifies the air to ensure that the ^{226}Ra source does not dry out.

The circulation of the air through the BBL extracts the ^{222}Rn from the ^{226}Ra source and forces it into the circuit, giving rise to a ^{222}Rn reference atmosphere, *i.e.*, a volume where the ^{222}Rn concentration value is known. When the circuit is calibrated with the standard sources, it can be used to measure the activity of other ^{222}Rn sources of unknown activity. This can be achieved simply by comparing the count rate given by the reference bubbler with the one produced by the ^{222}Rn from the unknown source.

Alternatively the system may be calibrated by the use of glass vials containing a known activity of ^{222}Rn . The system is equipped with a proper vial-holder used to break the vials and introduce the radon into the system without leakage (shown in **Figure 4**).



Figure 4 – Vial holder used to introduce the radon contained in glass vials into the radon reference measurement system. The small vertical cylinder on the top of the vial holder is used to break the vial and transfer the radon into the circuit.

The activity of the ^{222}Rn gas inside the glass vials was measured by the $4\pi\text{-}\gamma$ integral counting method using the ENEA-INMRI well-type NaI(Tl) 5” x 5” scintillator detector. The total counting efficiency, with which the count rate was normalized, was computed by the GEANT4 Monte Carlo code (Caponi, 2012). The two methods (reference bubblers and

radon in glass vials) are in agreement within 0.5%. A set of experiments was carried out to evaluate the reproducibility of the RRMS and to calibrate a fixed Ionization Chamber (IC) for ^{222}Rn measurement. ^{222}Rn gas was collected and sealed in a glass vial and the vial was measured with the IC.

After some days the ^{222}Rn activity of the vial was measured in the ^{222}Rn reference circuit with the procedure described in the previous paragraph. The obtained activity value was corrected for decay at the date of measurement in the ionization chamber and related to the ionization current. A series of 8 measurements were carried out obtaining reproducibility within 0.5%.

The procedure and minor features of the ^{222}Rn measurement circuit were refined and a new set of 5 experiments was carried out. For these experiments the average calibration factor of the ionization chamber is $K = 15543 (26) \text{ nA/kBq}$.

2.2. The French ^{222}Rn Reference Measurement System at LNE-LNHB

This method is based on a Defined Solid Angle (DSA) measurement of solid alpha-emitting sources widely used for primary standardization of activity. In the case of ^{222}Rn , which is a noble gas, the use of a cold finger surface allows the deposition of ^{222}Rn to produce a pure ^{222}Rn disk source under vacuum (below 10^{-5} hPa). The cold finger is a 6 mm diameter nickel rod, laser-welded to a 6 cm diameter stainless steel disk of $100 \mu\text{m}$ thickness. The nickel rod, which allows good thermal conduction, is cooled down at 80 K, while the stainless steel disk of lower conductivity and its periphery are heated to 293.15 (7) K (Figure 5).

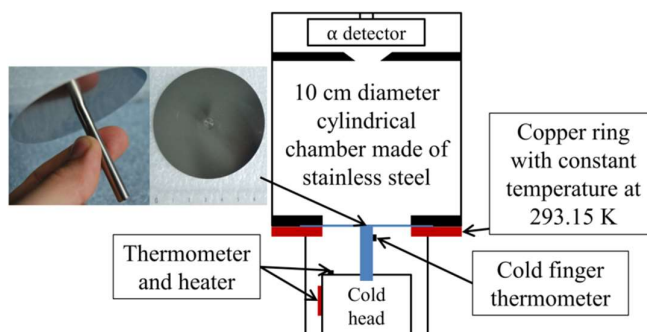


Figure 5 – Scheme of the DSA measurement chamber and picture of the cold finger used at LNE-LNHB.

The stainless steel disk is also used as a membrane to separate the volume containing the cold head and the measurement chamber. These two volumes have the same pressure (vacuum of $1 \cdot 10^{-5} \text{ hPa}$) to prevent mechanical deformation. At this pressure ^{222}Rn will be deposited on the cold surface as long as the temperature is lower than 100 K.

As the emission of alpha particles is isotropic, we assume that only the alpha particles passing through the collimator are counted (Fig. 4). Every alpha particle reaching the detector is counted by the silicon detector, and a spectrum is obtained in which radon and its daughters are well separated. The detection efficiency of the measuring system is simply equal to the geometry factor G defined by:

$$G = \frac{\Omega_{\text{eff}}}{4\pi},$$

where Ω_{eff} is the solid angle shown in Figure 6. In this device, $G = 2.019 \cdot 10^{-3}$ with a relative standard uncertainty of 0.28%.

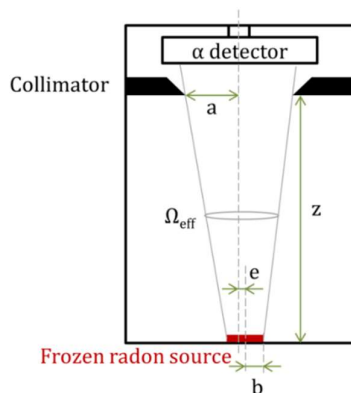


Figure 6 – Scheme of the DSA measurement system at LNE-LNHB: a, radius of the collimator; b, radius of the source; z, distance between source and diaphragm; e, eccentricity, if the source and collimator axes are different.

The geometry factor has been characterized by precise distance and diameter measurements by the French national metrology institute LNE. As a result, these quantities are directly traceable to national standards. The activity A of the measured source is then given by:

$$A = \frac{CR}{G} \quad (1)$$

With CR meaning “counting rate”.

The solid angle Ω_{eff} can be calculated using different kinds of software such as Anglesol from LNE-LNHB. As a result, with this kind of system, it is possible to obtain a small uncertainty as long as the counting statistical uncertainty is negligible and the dimensions of the apparatus are precisely known.

After the activity measurement, if needed, the ^{222}Rn is transferred by cryo-diffusion into a storage vial (**Figure 7**), which can be metal or glass. To perform this transfer, this storage vial is cooled with liquid nitrogen and the cold finger is heated to room temperature. A second measurement after the transfer, once the storage vial is closed, is performed in order to determine the background (e.g. ^{210}Po contamination, which varies over time).

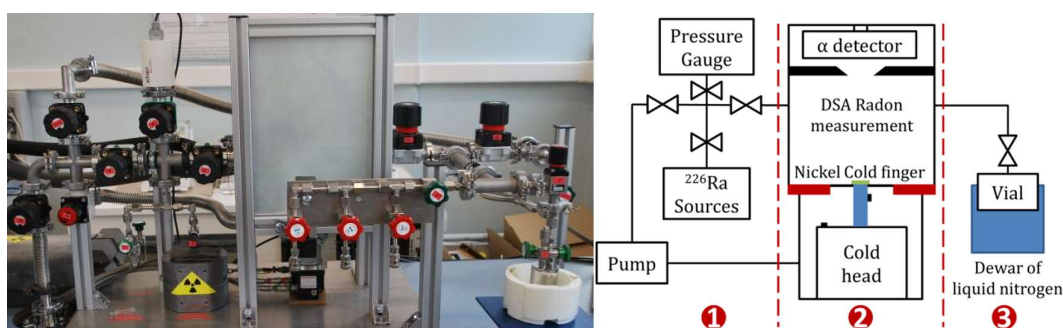


Figure 7 – Radon measurement system at LNE-LNHB (a) photograph and (b) simplified schematic representation.

Initially, a quantity of radon is selected by opening a radium source and waiting for the approximate activity required. The activity is measured and transferred. The aim is to ensure that all the radon on the cold finger is transferred, so enough time is allowed so that there is no residual radon in the chamber. Any radon left in the tubes is measured and the value deducted from the first measurement. If required, it is possible to transfer only part of the radon on the cold finger by closing the valve before all the radon is transferred. The remaining radon can then be condensed on the cold finger and deducted from the radon inside the container by calculating the difference between the two measurements.

The complete details of the measurement device are given in reference (Sabot, 2016), which is an upgraded version of an earlier system developed by LNE-LNHB in 1995 (Picolo, 1996).

2.3. The ^{222}Rn Measurement System at IFIN-HH

The method used by IFIN-HH is the 4π LSC-TDCR system for absolute activity measurement of an ampoule with transferred ^{222}Rn into a LS solution (Cassette, 2006). This measurement is completed by γ -ray spectrometry using an HPGe detector to measure net counting rates of the original receptacle GAZ 7 before and after the transfer of ^{222}Rn into the LS ampoule.

The original GAZ 7 receptacle from LNE-LNHB was measured successively over two days, at a distance of 45.5 cm from the surface of the HPGe detector, in vertical position, for both 352 keV and 609 keV Full Absorption Peaks (FAPs). These peaks are coming from the ^{222}Rn progenies (^{214}Pb and ^{214}Bi , respectively). Two mean net counting rates of N_{352} and N_{609} , counts per second (s^{-1}), were calculated (after background subtraction).

A quantity of ^{222}Rn was transferred to the LS type Ultimagold LLT ampoule N° 38, cooled with nitrogen vapour from liquid nitrogen (77 K) using the ^{222}Rn standard system at IFIN-HH (the system is described in reference (Sahagia, 2010)). The remaining quantity of ^{222}Rn was quantitatively recovered in the GAZ 7 receptacle, cooled as described above. The recovery was verified several times, as mentioned in section 4.1 of the reference (Sahagia, 2011). The method used, including the complete recovery of ^{222}Rn are also confirmed by the results of IFIN-HH within the K2 type comparison, CCRI(II)-K2.Rn-222, 2015 (BIPM-2015); this comparison was also piloted by the LNHB but the activity range was higher.

After the equilibrium was re-established in the GAZ 7 receptacle, it was again measured with the HPGe detector in the same initial geometry and new mean net counting rates, N'_{352} and N'_{609} , (s^{-1}), were registered. Two values of the counting

rate ratios: $R_{352} = N'_{352} / N_{352} = 0.926$ (41) and $R_{609} = N'_{609} / N_{609} = 0.894$ (42) were calculated. Their mean value is $R = 0.910$ (29), $u = 3.2\%$.

After the equilibrium was reached, the activity of LS ampoule N° 38, was repeatedly measured in the LSC TDCR system, obtaining the activity value $A_{38} = 913$ Bq, $u = 1.6\%$. The uncertainty of the activity of LS ampoule N° 38 has several components due to: counting statistics (measurement and background of D), detection efficiency and decay correction, respectively (see Table 5). More details about this type of measurements are presented in reference (Antohe, 2011).

The activity of the initial GAZ 7 receptacle at the reference date of the comparison, 15 April 2019, 12:00 UTC, was calculated as: $A_{GAZ7} = A_{38}/(1-R) = 10\,144$ Bq = 10.14 kBq. The uncertainty was calculated as the quadratic summation of the activity of A_{38} , (1-R), instability and recovery uncertainties. A value for the combined relative standard uncertainty of $u = 32.3\%$ was obtained.

A comparative measurement (estimation) by Monte Carlo simulation (GESPECOR software) for calculation of the HPGe efficiency for the two types of receptacles: GAZ 7 and LS ampoule, measured in vertical position, at 45.5 cm from the detector surface. The determined activity of (GAZ 7) before transfer is 10 696 Bq, the relative difference from the reported value (10 140 Bq), being about 5.5%.

Two problems were identified:

- Low transfer of the ^{222}Rn from the container into the ampoule (only about 10%): in vacuum (the pressure in the container was about 10% higher than in the radon transfer installation, approximately 3 Pa), with a transfer time of only about 5 min.
- Non-reproducible volume distribution of the ^{222}Rn daughters in the container (before and after transfer).

The arithmetic mean of the mean of the net counting rate ratios for ^{214}Pb and ^{214}Bi (two measurements before and two measurements after transfer) was used to compute the activity, which was 10 140 Bq at the reference time (15 April 2019, 12:00 UTC) as mentioned above. The results for the different sets of measurements (two before transfer and two after transfer) lead to four values of R , which were from 0.87 to 0.95; for (1-R), which is used to calculate the reported ^{222}Rn activity in GAZ 7 receptacle, result in a wide range, from 0.05 to 0.13.

The reported relative uncertainty of 32.3% resulted from quadratic summation of the following relative uncertainties: 1.6% (LS counting), 32.2% (relative uncertainty of the value of (1-R)) and 0.47% (supplementary due to recovery). It should be noted that the absolute standard uncertainty of (1-R) is equal to that of R , i.e. 0.029; so, the relative uncertainty of (1-R) is very high, $0.029/0.090=0.322$ (32.2%), which can explain the large relative difference between the IFIN-HH result and the reference value of this comparison.

In conclusion, the main reason identified for the rather large relative uncertainty in the result reported by IFIN-HH is the too low transfer of ^{222}Rn from the metallic recipient into the glass ampoule with LS solution. Consequently, the reported result had a very strong dependence of the relative measurements performed by gamma-ray spectrometry – measurements which might be affected by a non-reproducible volume distribution of the radon progenies (^{214}Pb , ^{214}Bi) before and after the radon transfer.

For the future, a better way to transfer the ^{222}Rn gas from the metallic container (100 cm³) into a glass vial with LS solution (20 cm³) should be identified, tested and implemented. The use of an air pump and a significantly longer time for the radon transfer might improve this type of radon activity measurement at IFIN-HH.

2.4. The Radon Measurement System at BEV

The measurement is done by complete exhalation of the ^{222}Rn from the small sample container into a big calibration container barrel with a net air volume V_c of 204.2 (3) L. The ^{222}Rn is completely exhaled from the small sample container into the absolute airtight calibration container barrel, homogenized and measured with the traceable calibrated Austrian national standard for ^{222}Rn activity concentration in air. The diffusion inlet of the ionization chamber has been positioned in the middle of the calibration container to ensure a highly representative measured ^{222}Rn activity concentration and reduce potential wall effects.

Before starting the exhalation and measurement of the ^{222}Rn activity concentration in the calibration container, the blank background rate r_0 (s⁻¹) of the used traceable calibrated ionization chamber (Genitron AlphaGuard PQ 2000 Rn/Tn, S/N 1338) is determined. For this purpose, the calibration container is flushed with outdoor air at low ^{222}Rn activity concentration < 5 Bq·m⁻³. The blank background count rate is determined over a measurement period of 3 600 s (1 h) in six 10 min sub-periods. The ionization chamber with an active measurement volume of 0.56 L is used in diffusion mode and operated in alpha particle impulse count mode.

After determination of the blank background count rate, the small sample container is placed inside the calibration container barrel (Genitron Typ EV, S/N 03211, D 56 cm, H 86 cm, 210.1 L gross empty nominal volume) and carefully opened. The net air volume of the container had been adjusted by subtracting the volumes of all entered solid parts (e.g.

ionization chamber, Rn-222 sample container). A small degassing hose – connected to an air pump (Genitron Alpha Pump) – is then put onto the small sample container. Then the calibration container is closed gastight and the air pump is switched on with a flow rate of $1 \text{ L} \cdot \text{min}^{-1}$ (Figure 8). The air circulates and degasses the ^{222}Rn from the small sample container into the barrel air volume. After about 20 min of air circulation, ^{222}Rn is almost completely exhaled from the sample container and distributed homogeneously by a fan into the air volume of the calibration container.

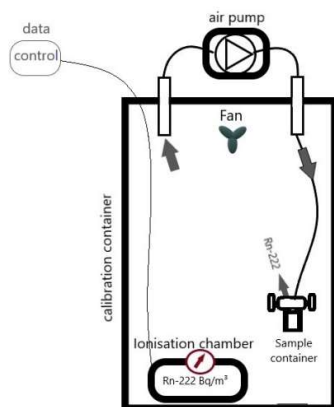


Figure 8 – Scheme of ^{222}Rn measurement inside the gastight radon calibration container at BEV.

Thirty minutes after the start of the ^{222}Rn emanation from the sample container into the air of the closed calibration container, the diffusion of ^{222}Rn into the ionization chamber is completed. Then the 3 600 s (1 h) measurement period starts. After finishing the data processing, the calibration container is opened and flushed again with low ^{222}Rn fresh outdoor air establishing the blank background situation in the calibration container.

The ^{222}Rn activity concentration in the container is calculated as follows:

$$c_A(T_m) = \frac{A(T_0)}{V_c} \cdot K_{\Delta t} = \frac{A(T_0)}{V_c} \cdot e^{-\ln(2) \cdot \frac{\Delta t}{T_{1/2}}}$$

- $c_A(T_m)$, ^{222}Rn activity concentration in the container at the time of measurement T_m .
- $A(T_0)$, ^{222}Rn activity of the radon sample at the reference time T_0 (Bq).
- V_c , air volume of the calibration container (m^3).
- $K_{\Delta t}$, correction factor for the radioactive decay of ^{222}Rn .
- Δt , time span between reference time and measurement ($T_m - T_0$) (d).
- $T_{1/2}$, half-life of ^{222}Rn : 3.8232 (8) d.

Details of the measured quantities (e.g. measured ionization current, calibration factor for ionization chamber) and detailed uncertainty calculation are given in (Maringer, 2019).

2.5. The German Radon Reference Measurement System at PTB

For the absolute determination of the activity of ^{222}Rn gas samples several methods are applied in the radioactivity laboratory of PTB. A primary method based on the Defined Solid Angle alpha spectrometry of condensed ^{222}Rn on a cold point (device Id.: M19) (Picolo, 1996). A gamma spectrometric method using an HPGe detector which is calibrated against PTB's Hönigschmid standard source (Schötzig, 1983) and a fast secondary standard system, applying an integral counting technique with dead time correction of an NaI(Tl) detector (device Id.: M07) together with selected, calibrated stainless steel containers.

During this EURAMET intercomparison the primary method and the fast secondary method were applied. The primary method was used to determine the activity of the ^{222}Rn source supplied by LNE-LNHB. The secondary method was used as a fall back, in case of vacuum problems and to determine the transfer losses of the vacuum system during the handling of the ^{222}Rn gas, in between the different measuring systems.

In the course of this activity determination, the ^{222}Rn activity supplied by LNE-LNHB was measured in the stainless steel container sent by LNE-LNHB. Then the activity was transferred to the PTB calibrated stainless steel container (Id. RN12) and measured again. Afterwards the activity was transferred to the primary measuring device M19, to determine the activity. In order to understand any transfer losses, the ^{222}Rn was retransferred to RN12 and measured again with the secondary standard M07 (a calibrated NaI(Tl) system).

The results from the secondary standard are in perfect agreement with the primary method results, which were reported as PTB results of this intercomparison. The secondary results from M07 of (12.90 ± 0.12) kBq ($k = 1$) have slightly higher uncertainty and prove that the transfer losses during this intercomparison can be neglected with the given frame of uncertainty. They also prove the usefulness of this stabilised secondary method, since the calibration factors, dating back to the year 2004, are still valid.

In Defined Solid Angle device, the detector is placed at the opposite side of the condensed ^{222}Rn source where only the alpha-particles passing through the diaphragm are detected. As the inner radius of the diaphragm is known and source-to-diaphragm distance as well as the source diameter, a defined geometry allowing the solid angle to be exactly calculated using standard elliptic integrals is achieved (Ruffle, 1967).

The cooling of the cold surface where the ^{222}Rn is condensed is created by a cryogenic system in which the cold head of the cryogenerator is connected to the cold surface with a flexible copper braid and a nickel rod of about 6 mm in diameter and 20 mm length. One end of the nickel rod serves as the cold surface and is welded to the centre of a thin circular stainless steel foil 75 μm thick. The construction from the cryogenic system to the cold point uses good thermal conductors of copper and nickel to ensure good thermal transfer.

At the cold head of the cryogenerator, there is a similar temperature regulating system allowing the temperature at the cold point to be adjusted. The coldest temperature available is 25 K.

The stainless steel foil with a thickness of 75 μm forms the bottom of the measurement chamber and is welded in the centre to the nickel rod and outside to the end of the UHV-flange.

Measurements of the system dimensions were carried out at the PTB Coordinate Metrology Section. The dimensional quantities have been determined as (7.997 ± 0.005) mm for the diaphragm radius and (168.40 ± 0.07) mm for the source-diaphragm distance. Specific investigations showed that the difference in the distance to the cold point does not change more than a few micrometres from room temperature to a temperature of less than 100 K.

The solid angle was evaluated first by calculation of the temperature distribution derived from the conduction of heat transfer described by Fourier's law, which show that radon cannot be condensed at a radius of more than 4 mm, considering the 3 mm radius of the nickel rod. An experiment using radiographic image was used to confirm the calculation results. It shows that the radius of the ^{222}Rn source is (4.9 ± 1.0) mm. Using all of these data, the geometry factor $\Omega/(4\pi)$ can be calculated to be $(5.626 \pm 0.010) \cdot 10^{-4}$ (Dersch, 2004).

With the DSA method, the relative standard uncertainty is reduced to less than 1%, where the most important components are: geometry factor = 0.18%, scattering of the alpha particles at the edge of the diaphragm = 0.3%, and peak evaluation of the ^{222}Rn spectrum = 0.6%.

The secondary measurement system with a digital dead time unit of the non-extendable type (Schönfeld, 1994) and an NaI(Tl) detector is used with integral counting of pulses (Dersch, 1998) for the confirmation of the absolute measurements. It was developed as an independent system for the activity determination of gaseous ^{222}Rn sources. To control the stability of the system, a ^{226}Ra point source is measured at regular intervals. From these measurements also a possible decrease of the detector efficiency can be observed and corrected for, guaranteeing the reliability of the radon activity determination.

2.6. The ^{222}Rn Measurement System at SMU

For the determination of the reference value of the ^{222}Rn gas sample No. GAZ 9 obtained from LNE-LNHB, a new SMU air radon secondary standard (AiRneSt) was used (**Figure 9** and **Figure 10**). This system will be used for calibration and verification of customers' devices as well as SMU standards in the future.

The standard consists of two main parts, a radon chamber and a measuring device. The chamber is a horizontal cylinder made from stainless steel with a diameter of 0.78818 (44) m, length of 2.00130 (10) m, thickness of the stainless sheet is 5.0 mm and inner volume of 0.9757 (23) m^3 (**Figure 9**). All dimensions (radius, diameter, length), inner volume and inner surface of the chamber were determined at SMU and are traceable to the Slovak National Standard of Length No. 002/97. The measuring device AlphaGUARD DF2000 was used as a reference device and the measurement system using Lucas cells as a parallel control device. Both systems are traceable to Czech National Standard via calibration in SÚJCHBO in Czech Republic.

Before starting measurements, the container with ^{222}Rn gas was connected to the chamber and ^{222}Rn gas was transferred through a HEPA filter and daughter products filter into the chamber by evacuating. The filling air (inner atmosphere of laboratory (air with background volume activity of radon less than 20 $\text{Bq}\cdot\text{m}^{-3}$) comes through the zero filter (a box with a volume of 30 l filled with activated charcoal), a dryer vessel with silica, a HEPA filter (acrylic copolymer on a non-woven support) to the container with ^{222}Rn (flushing of container with ^{222}Rn) and then is finally transferred to the radon chamber. Inside the chamber ^{222}Rn gas was mixed and homogenized with the inner atmosphere by a set of fans. Thermodynamic parameters like temperature (T), pressure (p) and humidity (h) were measured during measurement via calibrated monitors.

Then a long-term measurement was performed with a slight vacuum against ambient atmospheric pressure (about 1.5 kPa lower) to ensure the airtightness of the chamber.

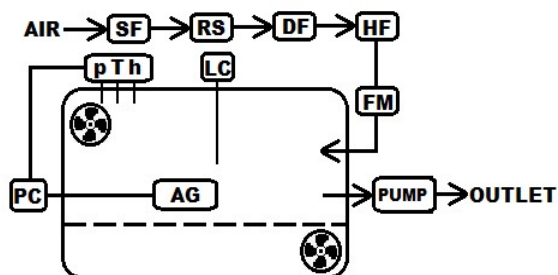


Figure 9 – Simplified block diagram of the radon chamber at SMU, where SF – silica gel filter, RS - source of ^{222}Rn , DF – daughters products filter, HF – HEPA filter, FM – flow meter, AG – AlphaGUARD, LC – Lucas cell, pTh - monitoring system for thermodynamic parameters, PC – personal computer.



Figure 10 – SMU radon chamber, AiRneSt

After transfer of radon to chamber is started, it is necessary to wait about 2 h to stabilize conditions inside the chamber to stabilize the AlphaGUARD response. Stable conditions are considered when the variance of measured values falls below 2%. Reference value of ^{222}Rn volume activity (RAC) was determined as a weighted mean of 21 specifically selected measured values in the chosen range. Selected measured data comes from stable AlphaGUARD response. The weight of each value was determined from the uncertainty of each value obtained directly from the radon monitor (AlphaGUARD). Each value was corrected for pressure (k_p) and temperature (k_T) measured directly in the chamber during measurement. Even though AlphaGUARD can measure p and T , these climatic data were not taken into account. Efficiency correction, decay correction and recalculation to the reference date were applied.

$$\text{RAC} = \text{RAC}_m \cdot k_p \cdot k_T$$

$$k_p = \left(1 + \left(A \cdot \left(\frac{p-1013}{1013} \right) \cdot \left(\frac{p}{p-620} \right) \right) \right)$$

$$k_T = \left(1 + \left(B \cdot \left(\frac{T-20}{293} \right) \cdot \left(\frac{130}{130-T} \right) \right) \right)$$

The radon monitor is connected to the PC via the KF system in order to collect measured data. There are two possibilities of a placement of radon monitor, either inside the chamber, mostly in diffusion mode, or outside the chamber in flow mode. Comparison measurements of volume activity of radon were performed in the radon atmosphere of the chamber in diffusion mode. The data from AlphaGUARD, RAC_m – measured volume activity of radon (Bq m^{-3}), were subsequently corrected by pressure k_p and temperature factors k_T , using the previous formula according to recommendations from Bertin Technologies (Technologies, 2019). A and B coefficients are taken from Bertin Technologies and are dependent on the measurement range.

This work at SMU was supported by the Slovak Research and Development Agency under the contract No. APVV-15-0017.

2.7. Measurement system from BFKH

The equipment consists of an airtight stainless steel chamber, a ^{222}Rn source (type RF 500 from Czech Metrological Institute), a leakproof gas pump (Alpha Pump, rate flows 0.3, 0.5 and $1 \text{ L} \cdot \text{min}^{-1}$), a nitrogen gas-cylinder, a controller unit with computer and an environmental parameters (temperature, air pressure and relative humidity) measuring unit. The volume of the chamber is 840 L. The activity of the radium source is 630.4 kBq with a very high emanation factor (99%). The homogeneity of the radon gas in the chamber is ensured by two continuously running fans. The radon chamber's distinctive features are the two gas circuits which have a common branch. The common branch consists of the radon generator and the pump. **Figure 11** shows the scheme of the measuring system. In the figure, the blue lines indicate the gas paths and the red one the electrical control lines.

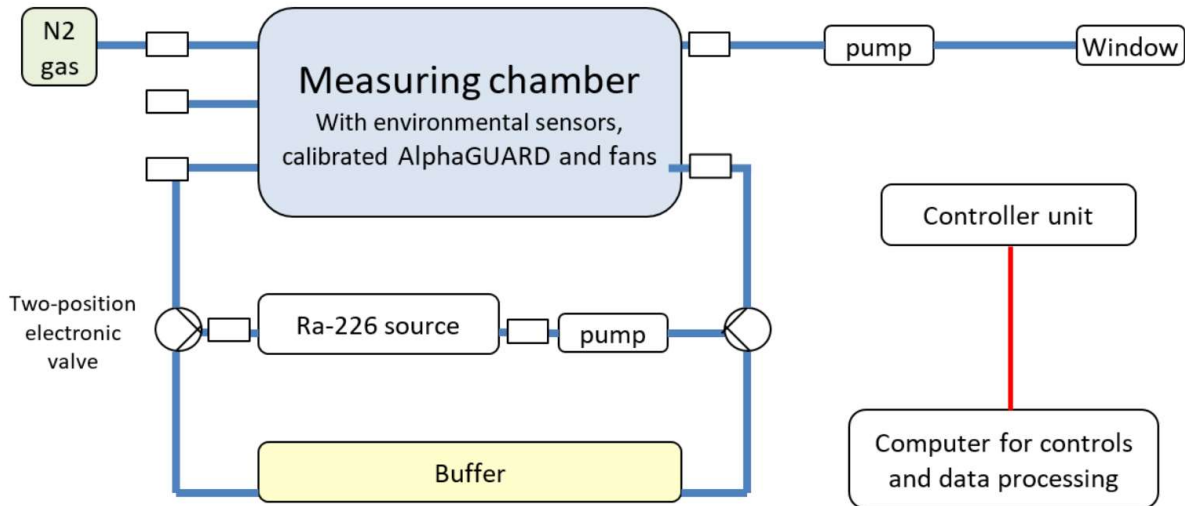


Figure 11 – Scheme of the measuring system.

The radon measuring system was flushed with nitrogen gas after being sealed. The radon gas container after its arrival at BFKH was connected to one of the inlet valves of the chamber. The radon was then transferred (flow through) to the measuring volume. Two internal fans assisted in homogenization. Once the radon concentration values measured by a calibrated (reference) AlphaGUARD in the chamber was stabilized, data were collected every 10 minutes.

The reference measuring device is a GENITRON AlphaGUARD PQ 2000 (serial number is 1150), calibrated in PTB.

The number of measurement readings was 830. An exponential function was fitted to the data. Extrapolation of the fitted function to the loading time gave the initial activity concentration of the transferred gas. To determine the total activity of the radon gas this value was multiplied by the volume of the radon chamber to give the reported value.

A very important parameter is the volume of the radon chamber. As AlphaGUARD only measures the radon concentration the volume is required to obtain the total activity.

It is not easy to determine the volume, since the value calculated from the side edges may be different from the actual one (side swell, etc ...). The radon chamber volume was determined by CO concentration measurement. The advantage of this method is that the walls of the chamber do not deform under high or low pressure. A well-known amount of CO was introduced into the chamber and circulated through a CO concentration meter.

The volume can be determined using the universal gas law : $pV=nRT$

where p is the gas pressure, V the volume, n the amount of chemical substance (mol), R the universal gas constant, T the gas temperature.

The measured volume of the measuring chamber is $V_{\text{chamber}} = (844.9 \pm 5.9)$ L. Of course, the minimum (828.6 L) and the maximum (849.1 L) volume have been determined from the side edges as well.

As the volume determined by CO concentration measurement is within the minimum-maximum range, it has been accepted as the volume value.

3. Results and evaluation

Seven laboratories took part in this comparison providing results in kBq, as shown in Table 4, where the combined uncertainties are also given.

Table 4: Measurement results evaluated on 15 April 2019 12:00 UTC. Values as reported by participants

Laboratory	Activity (kBq)	Combined standard uncertainty (kBq)	Combined relative standard uncertainty (%)	Activity concentration (Bq·cm ⁻³)*	Total relative standard uncertainty (%)**
BEV 4P-IC-AP-00-00-00	13.0	0.4	3.1	123.9	3.1
BFKH 4P-IC-AP-00-00-00	15.11	0.6	3.8	145	3.8
ENEA-INMRI 4P-IC-GR-00-00-00	13.056	0.155	1.19	124.0	1.3
IFIN-HH 4P-LS-MX-00-00-TD UA-GH-GR-00-00-00	10.14 [§]	3.28	32.3	97	32
LNE-LNHB SA-PS-AP-00-00-00	12.710	0.054	0.42	121.3	0.6
PTB SA-PS-AP-00-00-00	12.90	0.10	0.8	121.9	0.9
SMU 4P-IC-AP-00-00-00	13.14	0.49	3.75	125.1	3.8

* The activity concentration is the result of the activity (measured by each laboratory) divided by the volume given in **Erreur ! Source du renvoi introuvable.** Table 2.

** Total uncertainty also includes the uncertainty on the volume, cf. § 2.

§ Mean of four values measured.

Table 5 presents the detailed uncertainty budget as provided on the reporting forms of the participants for all individual parameters included in the measurement process.

Table 5: Relative standard uncertainty components as reported by participants for their activity measurement, where $u(x)_i$ is the uncertainty of the i^{th} input quantity, x_i , and $u(y)_i$ is the corresponding contribution to the output quantity (activity) from input quantity i .

Laboratory Method	Uncertainty components, x_i	$u(x)_i$ (%)	$u(y)_i$ (%)	Evaluation type
BEV 4P-IC-AP-00-00-00	K decay	0.09		A
	Mean activity concentration at ref. date (Bq/m ³)	0.7		A
	Background (Bq)	41		A
	Calibration factor	3.0		B
	Air volume	0.10		B
BFKH 4P-IC-AP-00-00-00	Volume of radon chamber (m ³)	0.69		B
	Statistical uncertainty of measured values (kBq/m ³)	3.8		A
ENEA-INMRI 4P-SC-GR-00-00-HE	Activity of reference ²²⁶ Ra solution for calibration	0.60		A
	Radon emanation factor of reference radium solution	0.80		A
	Reproducibility of calibration	0.40		B
	Reproducibility of the measurement of ²²² Rn gas	0.50		B
IFIN-HH 4P-LS-MX-00-00-TD; UA-GH-GR-00-00-00	Statistical uncertainty of the LSC-TDCR measurement	0.25	0.25	A
	Statistical uncertainty of the background of D (LSC-TDCR)	1.1	1.1	A
	Detection efficiency (LSC-TDCR)	1.1	1.1	B
	Decay correction (LSC-TDCR)	0.11	0.11	B
	Statistical uncertainty of the calculation of R	3.2	32.2	A
	Recovery of radon in the receptacle GAZ 7 after the transfer in the LS ampoule	0.47	0.47	B
LNE-LNHB SA-PS-AP-00-00-00	counting statistics	0.20	0.21	B
	dead time	0.001	0.001	B
	background	4.1	0.02	B
	pile-up	0.001	0.001	B
	counting-time	0.0023	0.0023	B
	decay-scheme	0.016	0.016	B
	other effects (G)	0.32	0.32	B
	decay correction during measurement	0.042		B
	backscattering	0.0000001		B
	repeatability	0.01	0.01	B
	reproducibility	0.18	0.18	B
PTB SA-PS-AP-00-00-00	solid angle	0.68	0.68	A
	counting statistics	0.15	0.16	A
	dead time	0.46	0.47	A
	decay correction during measurement	0.000004	0.000004	A
	background	1.2	0.078	A
	frequency standard	0.02	0.02	B
	time to start of measurement	0.17	0.012	A
	nuclear data	0.014	0.014	B
	time to reference date	0.04	0.0073	A
	transfer losses	0.13	0.13	A
SMU 4P-IC-AP-00-00-00	temperature	0.082		A/B
	pressure	0.034		B
	calibration factor	3.7		B
	volume of radon chamber	0.27		B
	decay correction (half-life)	0.021		B
	weighted mean	0.0072		A

A Grubbs test (5725-2:1994) was performed on the results submitted by the participants, which showed no outliers were identified (Table 6).

Table 6: Grubbs test

Nb of results	7
Gmin	1.846
Gmax	1.590
Critical value at 5%	2.02
Critical value at 1%	2.14
Test	Correct

The final results submitted by the participants in $\text{Bq}\cdot\text{cm}^{-3}$ together with the arithmetic mean and weighted mean values are shown in **Figure 12**.

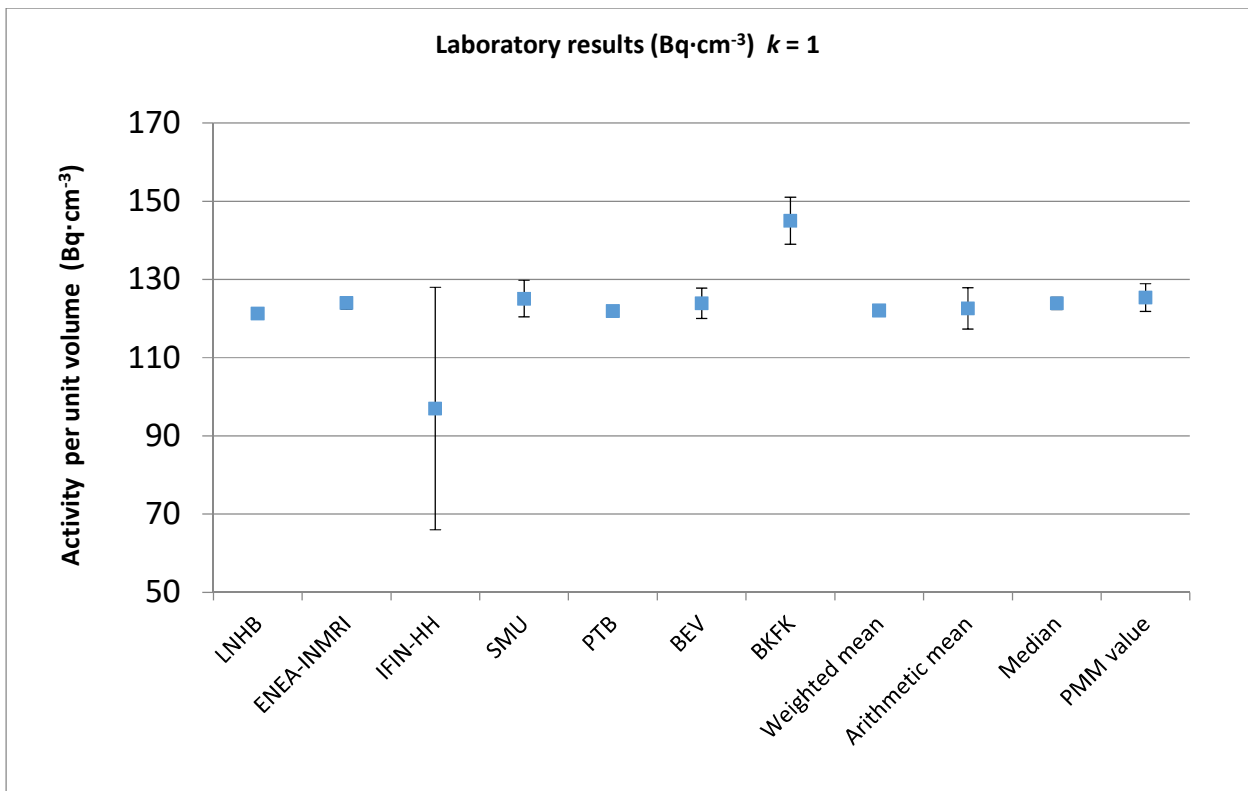


Figure 12- Individual results of the international comparison of activity per unit volume (7 individual results) in $\text{Bq}\cdot\text{cm}^{-3}$

The details of the weighted mean, arithmetic mean, median and PMM value are given in Table 7.

Table 7: PMM results, median, weighted mean, arithmetic mean for the activity concentration, C_A

	C_A ($\text{Bq}\cdot\text{cm}^{-3}$)	u_{CA} ($\text{Bq}\cdot\text{cm}^{-3}$)
Weighted mean	122.0	0.9 (external)
Arithmetic mean	122.6	5.3
Median	123.9	1.6
PMM value	125.4	3.5

Following the policy of the KCWG(II), the Power-Moderated Mean (PMM) (Pommé, 2013) was used to calculate the reference value of the comparison: $125.4 (35) \text{ Bq}\cdot\text{cm}^{-3}$ and details of PMM calculations are given in **Figure 13**. D corresponds to each value minus the mean and S50 corresponds to the standard deviation divided by the z-score for the middle 50% of normal distribution.

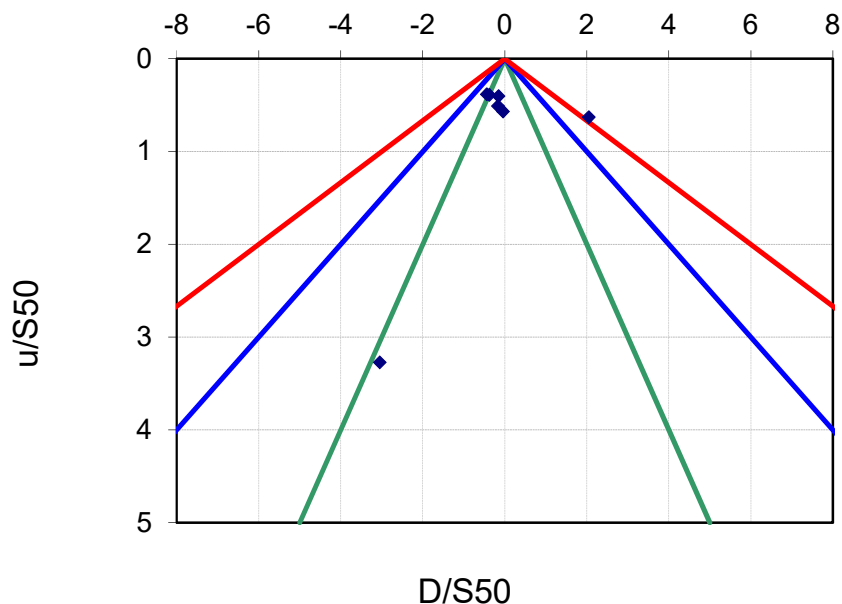


Figure 13 – PomPlot of the participants' results for the ^{222}Rn intercomparison.

4. Conclusion

All results are consistent, within the uncertainties. No outliers were identified by the Grubbs test (Table 6). Nevertheless, five results are in very good agreement whereas two others are quite discrepant (about 20% from the weighted mean). The relative standard deviation between the seven results is 11.4%.

5. References

- 5725-2:1994, ISO. ISO 5725-2:1994 .
- Antohe, A., Sahagia, M., Luca, A., Cassette, Ph., Ivan, C. 2011. Influence of detection efficiency on the measurement of radon by liquid scintillation counting. *RADIOCARBON, Special Volume (2011)*, pp. 189–197. *Proc. of the 2010 International Liquid Sc.* 2011, pp. 189-197.
- Bé, M.-M., et al. 2008. Table of Radionuclides (Vol. 4 - A = 133 to 252). Monographie BIPM-5, Vol. 4, Bureau International des Poids et Mesures (2008), Sèvres, France. 2008.
- BIPM. BIPM-1. <https://www.bipm.org/fr/cipm-mra>.
- BIPM-2. <https://www.bipm.org/utis/en/pdf/CIPM-D-01.pdf>.
- BIPM-2015. <https://www.bipm.org/kcdb/comparison?id=1383>.
- Caponi, M., et al. 2012. Comparison between two absolute methods used for ^{177}Lu activity measurements and its standardization. *ARI 70 (2012), Issue 9, 2075-2080*. 2012, pp. 2075-2080.
- Cardellini, F. et al. 2016. Metrological aspects of international intercomparison of passive radon detectors under field conditions in Marie Curie's tunnel in Lurisia. *NUKLEONIKA 2016, 61(3), 257-261*. 2016, pp. 257-261.
- Cassette, P., Sahagia, M., Grigorescu, L., Lépy, M.C., Picole, J.-L. 2006. Standardization of Rn-222 by LSC and comparison with alpha and gamma spectrometry. *ARI. 64 (2006), Issues 10-11, 1465-1470*. 2006, pp. 1465-1470.
- De Felice, P., et al. 1996. The ^{222}Rn Reference Measurement System Developed at ENEA., *NIM-A 369 (1996) 445-451*. 1996, pp. 445-451.
- Dersch, R.,. 2004. Primary and secondary measurements of ^{222}Rn . . *ARI 60 (2004), 387-390*. 2004, pp. 387-390.

- Dersch, R., Schötzig, U., 1998. Production and Measurement of ^{222}Rn Standards. . *ARI 49 (1998), Issues 9-11, 1171-1174*. 1998, pp. 1171-1174.
- Maringer, F.J., Wiedner, H., Cardellini, F. 2019. An innovative quick method for traceable measurement of radon 222 in drinking water. *ARI 155 (2019), 1-9*. 2019, pp. 1-9.
- Piccolo, J.-L.,. 1996. Absolute measurement of radon 222 activity. *NIM-A 369 (1996), 452-457*. 1996, pp. 452-457.
- Pommé, S. 2013. Determination of a reference value, associated standard uncertainty and degrees of equivalence for CCRI(II) key comparison, European Commission, Joint Research Centre, Institute for Reference Material, 2012. *Report EUR 25355 EN Errata notice published in CCRI(II) working document, 2013, CCRI(II)/13-18*. 2013.
- Ruffle, M.P., 1967. 1967. The geometric efficiency of a parallel-disc source and detector system. *NIM-A 52 (1967), 354-356*. 1967, pp. 354-356.
- Sabot, B., Pierre, S., Cassette, P., 2016. 2016. An absolute radon 222 activity measurement system at LNE-LNHB. *ARI 118 (2016), 167-174. ARI 118 (2016), 167-174*. 2016, pp. 167-174.
- Sabot, B., Rodrigues, M. Pierre, S. 2020. Experimental facility for the production of reference atmosphere of radioactive gases (Rn, Xe, Kr, and H isotopes), . *Applied Radiation and Isotopes, volume 155, 2020, 108934*. <https://doi.org/10.1016/j.apradiso.20.2020.108934>.
- Sahagia, M., et al. 2010. The Rn-222 standard system established at IFIN-HH, Romania. *ARI 68 (2010), 1503-1506*. 2010, pp. 1503-1506.
- Sahagia, M., et al. 2011. Establishment of the Rn-222 traceability chain with the Romanian Standard System. *NIM-A 631 (2011), 73-79*. 2011, pp. 73-79.
- Schönfeld, E., Janssen, H.,. 1994. Precise measurement of dead time. *NIM-A 339 (1994), 137-143*. 1994, pp. 137-1473.
- Schötzig, U., Debertin, K.,. 1983. Photon emission probabilities per decay of ^{226}Ra and ^{232}Th in equilibrium with their daughter products. *ARI 34 (1983), 533. ARI 34 (1983), 533*. 1983, p. 533.
- Technologies, Bertin. 2019. Bertin Technologies “Exchange with technical staff from Bertin Technologies from 02.09.2019”. 2019.

Index of Tables and Figures

Table 1: List of participants	1
Table 2: Container volumes	3
Table 3: List of the methods used as reported by participants	3
Table 4: Measurement results evaluated on 15 April 2019 12:00 UTC. Values as reported by participants	12
Table 5: Relative uncertainty components, $k = 1$, as reported by participants.....	13
Table 6: Grubbs test	14
Table 7: PMM results, median, weighted mean, arithmetic mean	14
Figure 1: Picture of a container	2
Figure 2: Dimensions of the containers	2
Figure 3 – Primary ^{222}Rn Reference Measurement System operating at INMRI-ENEA.....	4
Figure 4 – Vial holder used to introduce the radon contained in glass vials into the radon reference measurement system. The small vertical cylinder on the top of the vial holder is used to break the vial and transfer the radon into the circuit.....	4
Figure 5 – Scheme of the DSA measurement chamber and picture of the cold finger used at LNE-LNHB.	5
Figure 6 – Scheme of the DSA measurement system at LNE-LNHB:a, radius of the collimator; b, radius of the source; z, distance between source and diaphragm; e, eccentricity, if the source and collimator axes are different.	5
Figure 7 – Radon measurement system at LNE-LNHB (a) photograph and (b) simplified schematic representation.	6
Figure 8 – Scheme of ^{222}Rn measurement inside the gastight radon calibration container at BEV.	8
Figure 9 – Simplified block diagram of the radon chamber at SMU, where SF – silica gel filter, RS - source of ^{222}Rn , DF – daughters products filter, HF – HEPA filter, FM – flow meter, AG – AlphaGUARD, LC – Lucas cell, <i>pTh</i> - monitoring system for thermodynamic parameters, PC – personal computer.....	10
Figure 10 – SMU radon chamber, AiRneSt	10
Figure 11 – Scheme of the measuring system.....	11
Figure 12- Individual results of the international comparison of activity per unit volume (7 individual results) in $\text{Bq}\cdot\text{cm}^{-3}$	14
Figure 13 – PomPlot of the participants’ results for the ^{222}Rn intercomparison.	15