



## Remarks to history of radon activity concentration metrology

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**Abstract.** The radon issue has been known worldwide for dozens of years. Many scientific (ICRP Publication No. 137), technical (ICRU Report No. 88), and legislative (Council Directive 2013/59/EURATOM (EU-BSS)) documents have been published in the last decade. More and more attention is being paid to precise quantification to determine the concentration and consequent effects of various pollutants on human health worldwide. The quality of measurement and the variety of measurement techniques increase the need to unify measurement procedures and metrology continuity. Countries around the world are beginning to unify metrological procedures for determining different quantities based on international recommendations and standards. Not only for these reasons, it became more actual a need for more accurate radon activity concentration measurement and radon metrology unification. This paper summarizes the main remarks and technical aspects to the historical development of radon metrology.

**Keywords:** Intercomparison • Metrology • Radon • Radon chamber

### Introduction

The radon metrology is a complex technical discipline. For laboratory experiments and metrological procedures, the Ra-226 with the known activity concentration in the solid or liquid form as a source of gaseous Rn-222 is used. The individual metrological laboratories perform the internal calibration of their radon activity concentration measurement devices based on these types of sources [1, 2]. The commercial companies measuring radon in the field mostly follow the producers' calibration. Only some of them are using radon chambers for verification of their results – it is an adequate way of metrological order.

### History of radon measurement

The radon issue has been known worldwide for dozens of years. The first mention of the harmful effects of mine atmosphere (radon) was recorded by Georgius Agricola by observing miners of silver mines in the Schneeberg in the 16th century [3].

In 1901, Rutherford and Brooks experimented to determine if the emanation from radium was a vapour of the substance, a radioactive gas, or particles of matter that contained a large number of molecules [4]. In 1903, Rutherford and Soddy measured the half-life of the radium emanation as 3.7 days (now known as 3.823 days). In 1932, Robley Evans demonstrated, in his 1.5 L ionization

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**Table 1.** Calibration factors obtained by the participants given as arithmetic mean, weighted mean, and comparison reference value

Activity concentration [kBq/m <sup>3</sup> ]	Calibration factor		
	Arithmetic mean with standard uncertainty	Weighted mean with standard uncertainty	Comparison reference value with coverage interval at 95%
1	1.004 ± 0.028	0.998 ± 0.011	0.999 ± 0.057
3	1.004 ± 0.028	1.000 ± 0.009	0.979 ± 0.075
10	1.007 ± 0.035	1.011 ± 0.009	0.987 ± 0.081

chamber, that the lower level of detection of radon gas was at a background statistical variation value of 0.074 Bq. The device he developed for this determination combined a double ion chamber with a string electrometer. In 1957, the “Lucas cell” was developed using the alpha-scintillation properties of silver-activated zinc sulfide [5].

### Intercomparisons

In the 1980s, the rising concern about radon-induced lung cancer triggered the start of global comparison programmes based on a common radon atmosphere in which multiple radon and radon progeny detection systems (active as well as passive ones) were exposed. The Organization for Economic Co-operation and Development (OECD)/Nuclear Energy Agency and the Commission of the European Communities (CEC) ran the “Programme on radon and thoron dosimetry”, starting 1983 (OECD, 1985). The responsibility for managing the programme was shared by the former Australian Radiation Laboratory (ARL) for the Pacific region, the US Department of Energy (DOE), the former Environmental Measurements Laboratory (EML), and the US Bureau of Mines for North America, as well as the former National Radiological Protection Board (NRPB) for Europe [6].

The first intercomparisons were carried out using scintillation cells or sample flasks, which were filled in chosen radon test chambers. The range of the radon concentrations during these intercomparisons was from 1000 Bq·m<sup>-3</sup> to almost 20 000 Bq·m<sup>-3</sup> [7, 8]. During these comparative measurements, some drawbacks of air sample transport in scintillation chambers or sampling vessels have been described. One conclusion of the comparison of laboratories from Australia, the USA, and the UK was that as of mid-1985, the primary methods used by the four

laboratories to measure <sup>222</sup>Rn mutually agree to the point that the coefficient of variation of the pooled results was about 7%.

With the development of measuring techniques in the area of radon concentration measurement, a comparative measurement based on the transport of the comparison equipment between individual laboratories was started at the turn of the millennium. One of these intercomparisons for radon activity concentration has been performed within the scope European Metrology Education and Training (EuroMET) in project 657. Twelve laboratories from nine European countries were involved in this project. The intercomparison was performed in different radon atmospheres at 1, 3, and 10 kBq/m<sup>3</sup> via a transfer standard. Some results from this intercomparison are presented in Table 1 [9].

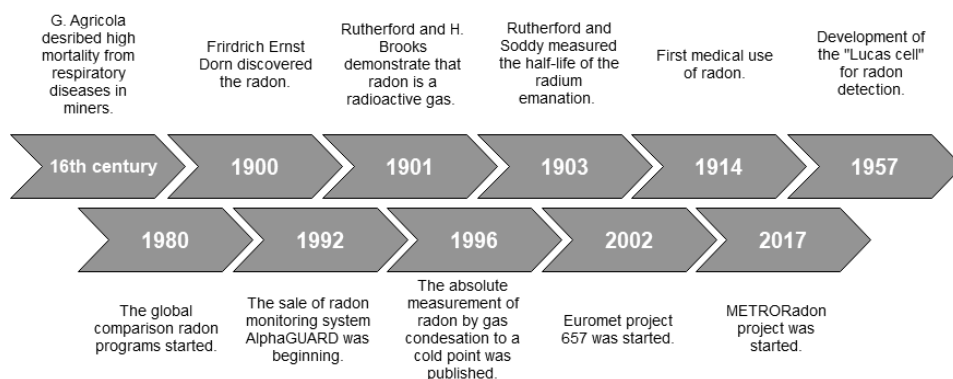
Owing to the need to develop reliable technics and methodologies to enable SI traceable radon activity concentration measurements and calibration at low radon concentrations, the Metrology for Radon Monitoring (MetroRADON) project 16ENV10 was started in 2017. One of the main goal of this project was to develop novel procedures for the traceable calibration of radon measurement instruments at a range of activity concentration from 100 Bq·m<sup>-3</sup> to 300 Bq·m<sup>-3</sup> with relative uncertainties ≤5% (*k* = 1).

The main historical milestones of radon activity concentration measurements and metrology are presented in Fig. 1.

### Material and methods

#### Absolute measurement of <sup>222</sup>Rn activity

By 1996, J. L. Picolo published a paper where presented absolute measurement of radon-222 activity consisted of applying the standard defined geometry



**Fig. 1.** Timeline of the main milestones of radon history. EuroMET (European Metrology Education and Training); MetroRADON (Metrology for Radon Monitoring).

measurement technique, used for the solid source, to a gas condensed on a cold point [10]. This measurement was adopted by several laboratories from China [11], Germany [12], Switzerland [13], and Korea [14]. The original measuring system allowed measuring with the relative uncertainty of 0.5% ( $1\sigma$ ). This system was upgraded by the Laboratoire National Henri Becquerel (LNE-LNHB) and now allows the measurement of radon sources from 100 Bq to 4 MBq with a relative standard uncertainty of 0.3% [15].

### On-site comparison

During the prehistory of the radon metrology, one of the first over world on-site intercomparisons were realized in the underground spaces (Badgastein) in 1991 [16]. In these, underground spaces were not possible to ensure the time stable and homogeneous radon atmosphere.

The technical devices that are frequently used for the on-site comparison of measuring instruments or grab sampling methods for radon activity concentration determination are the radon chambers. One of the oldest chambers was constructed from plywood in the form of a rectangular box in the Denver Research Center during the 1970s [17].

The effective volume of these chambers varies from 0.01 m<sup>3</sup> to 78 m<sup>3</sup>. They are built in various size and structure such as closed pipe system, cylindrical or cubic. The chambers are equipped by sensors of temperature, relative humidity, and atmospheric or inner pressure. The radon atmosphere is generated in different levels of radon activity concentration from hundreds of Bq·m<sup>-3</sup> to hundreds of kBq·m<sup>-3</sup> [18].

The special types of radon chambers are radon-aerosol chambers, which have sufficient inner volume (mostly more than 10 m<sup>3</sup>) and appropriate ratio of the volume and inner surface for creating of the time-stable aerosol atmosphere. The radon-aerosol chambers are equipped by monitoring system of inner climatic conditions and by different technical devices changing the concentration and size distribution of aerosol particles, temperature, or humidity [19–21].

The radon atmosphere is generated mainly from the solid radium sources, but it is also possible to use the liquid radium solution. The radon source is possibly placed directly in the chamber and used in the diffusion regime or operate outside the chamber in the flow regime.

The method of on-site comparison is suitable mainly for comparison of solid-state nuclear track detectors (SSNTDs) for integral determination of radon activity concentration.

### Radon in the air sample exchange

Several technics could prepare the reference radon activity concentration in the transportable package. The first intercomparisons were between the samples prepared from the atmospheres of the radon



Fig. 2. The glass bubble for the transport of radon activity.

test chambers. The air was field to scintillation or ionization cells directly or to other sample flasks (an example of a glass bubble for reference radon activity transport presents Fig. 2). Some problems were identified during the transport between laboratories. Owing to a long time in transit, perhaps aggravated by repeated handling and by the reduced pressure in aircraft cabins, some cells appeared to develop leaks.

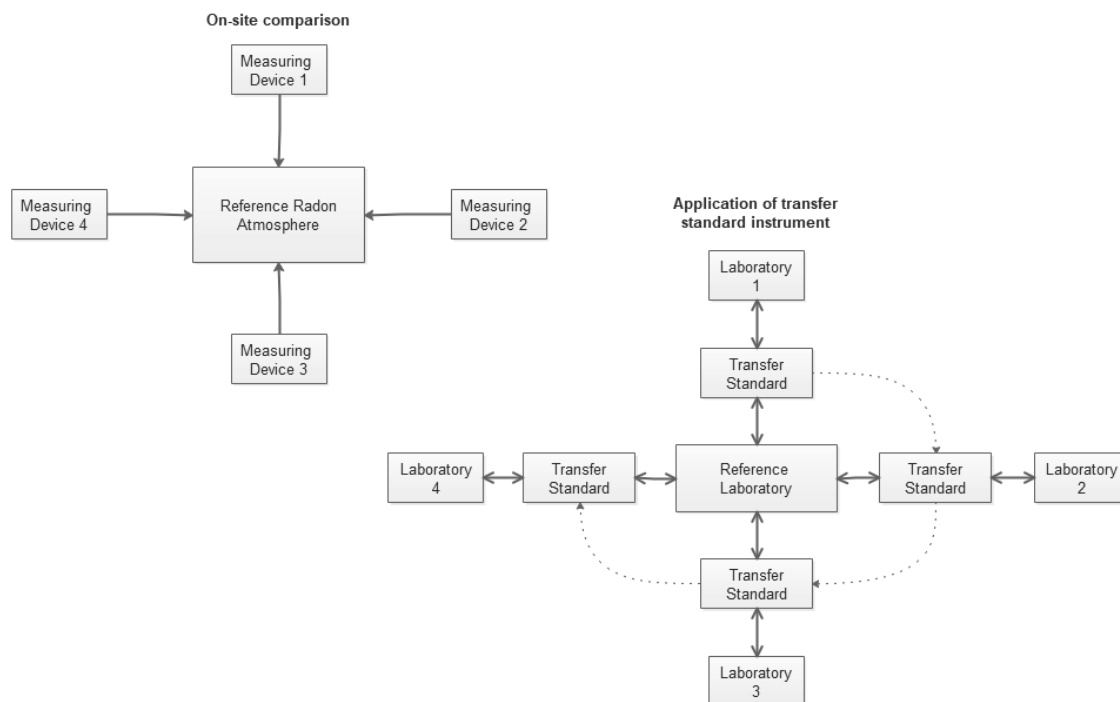
### Application of transfer standard instrument

As a transfer standard are defined the devices allowing determination of the comparison reference value (CRV) [4]. For determination of radon activity concentration using CRV, continuous radon monitors (AlphaGUARD PQ 2000 or AlphaGUARD DF 2000 are most frequently used) are used. The transfer standard instrument is applied to individual radon chambers of participants. After the individual measurement, the transfer standard (TS) instrument is send back to the main laboratory. It is checked there and is send to the other laboratory. This method is particularly suitable for comparing the determination of radon activity concentration in individual laboratories. The application of transfer standard instrument has been successfully used in the EuroMET project 657 [9].

Figure 3 presents the main differences between on-site comparison (left) and application of transfer standard instrument (right). On-site comparison is suitable mainly for grab sampling methods and SSNTDs; it is less time consuming and less logistically demanding. The application of transfer standard instrument is less costly after good logistics planning.

### Conclusions

At present, no one has doubt regarding the harmful effects of radon and especially its decay products on the human body. Radon measurements had a scientific and cognitive character in the early 20th century. During the radon history, hundreds of experts were dedicated to radon metrology and thousands of people performed the field measurements. Therefore, many significant technician steps in determining radon activity concentration have been realized over the years. Now, attention needs to be focused on the unification of measurement procedures



**Fig. 3.** Metrology of radon activity concentration performed by on-site comparison and application of transfer standard instrument.

and the metrological linking of individual regions over the world. Presently, it would be the best to apply on-site comparison or application of transfer standard instrument for radon activity concentration metrology between different laboratories.

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