BEYOND THE STATE OF ART: NEW METROLOGY INFRASTRUCTURE FOR RADON MEASUREMENTS AT THE ENVIRONMENTAL LEVEL

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Abstract:

Radon gas is the largest source of public exposure to naturally occurring radioactivity. Radon activity concentration maps, based on atmospheric measurements, as well as radon flux maps can help Member States to comply with the EU Council Directive 2013/59/Euratom and, particularly, with the identification of Radon Priority Areas. Radon can also be used, as a tracer, to improve Atmospheric Transport Models and to indirectly estimate greenhouse gas (GHG) fluxes. This is important for supporting successful GHG mitigation strategies. One approach to estimate GHG fluxes on local to regional scale is the so-called Radon Tracer Method (RTM), which is based on the night-time correlation between atmospheric concentrations of radon and GHG measured at a given station together with information on the radon flux data within the station footprint. Thus, atmospheric monitoring networks are interested or are already measuring atmospheric radon activity concentrations using different techniques but a metrological chain to ensure the traceability of all these measurements was missing.

Since 2020 a large consortium engaged in the project traceRadon [1] to develop the missing traceability chains to improve the respective sensor networks http://traceradon-empir.eu/ . This paper presents results in the areas: Novel ^{226}Ra standard sources with continuous controlled ^{222}Rn emanation rate, radon chambers aimed to create a reference radon atmosphere and a reference field for radon flux monitoring. The achieved results are making new calibration services far beyond the state of art possible.

Keywords: radon calibration; radon flux calibration; traceRadon; radiation protection; climate observation

1. INTRODUCTION

Naturally occurring radon is the cause of most of the population's exposure to ionizing radiation. Radon is at the same time a highly efficient tracer for understanding atmospheric processes, evaluating the accuracy of chemical transport models, and providing integrated emission estimates of greenhouse gases. A metrological system for measuring atmospheric radon activity concentrations as well as the radon flux from the soil is therefore needed for atmospheric, climate, and radiation research. The particular challenge lies in the low activity concentrations of radon in outdoor air from 1 Bq·m^{-3} to 100 Bq·m^{-3}, where below 100 Bq·m^{-3} there is currently no metrological traceability at all. Thus, measured values of different instruments operated at different locations cannot be compared with respect to their results and exchange processes cannot be followed.

To remedy this situation, a suitable new metrological infrastructure was developed, including new radioactive sources based on ^{226}Ra, new calibration procedures in radon reference atmospheres [2, 3], and for the first time, the development of an infrastructure for radon-flux calibrations.

2. NEW ACTIVITY STANDARDS

The metrology of radon (^{222}Rn is considered here) with respect to the representation of the unit of activity concentration in Bq m^{-3} can be fundamentally divided into two different approaches. One way to quantify radon absolutely is the method of Picolo [4], in which gaseous radon from the decay of radium (^{226}Ra) is frozen out at a cold point in vacuum. The alpha particles emitted by the solid radon are measured at a defined solid angle so that the unit of activity can be traced back to the base units of seconds and meters. The radon is then returned to the gas phase and can subsequently be used to produce atmospheres of known activity concentration. Other methods are based on liquid scintillation counting [5] or $4\pi\gamma$ method [6]. The activity concentration of an atmosphere produced in this way decreases exponentially with a half-life of 3.8 days, limiting the counting statistics and hence the achievable uncertainty in the calibration of test samples. Thus, this approach is not suitable for representing reference atmospheres with activity concentrations comparable to those in outdoor air (< 100 Bq·m^{-3}).

Figure 1: New kind of activity sources for ^{222}Rn based on the principle of emanation from electro-deposition, implantation or vapor layered ^{226}Ra activity standard. From top to bottom: Electro-deposited ^{226}Ra activity standard: Deposition at 30 V < U < 200 V; Implanted ^{226}Ra activity standard: Implantation of ^{226}Ra into W / Al after mass separation; PIPS ^{226}Ra activity standard: 450 mm², 300 µm with 150 Bq ^{226}Ra layer.

An alternative to this method is the use of preparations containing radium and such that a portion of the resulting daughter nuclide radon is continuously released into a reference volume. This continuously replaces the radon decaying in the volume, resulting in a state of equilibrium, i.e., a constant activity concentration, after about five to ten half-lives [7].

The use of this activity concentration for calibration purposes requires that the released activity of radon can be quantified traceable to the SI system.

Since the mechanism that causes the release of radon consists of a combination of two processes, on the one hand the recoil of the nuclei as a result of alpha decay and on the other hand the diffusion processes, the amount of radon released usually correlates with environmental parameters, such as relative humidity and temperature, since these influence the effective diffusion coefficient. In order to investigate these dependencies, novel sources were developed at PTB with the metrological possibility of continuous determination of the emanation [8]. Thus, it could be shown that at low penetration depth of the ion-implanted ^{226}Ra there is no measurable dependence of the emanation on the ambient humidity and only a small dependence on the temperature [9]. The absolute determination of the activity of ^{226}Ra is done for this type of sources by a-spectrometry under defined solid angle, while the released fraction of ^{222}Rn is quantified by g-spectrometric comparison with radon-dense sources of the same design. Figure 1 shows three different activity standards in an overview.

On this basis, several primary ^{222}Rn activity standards were fabricated that are suitable for representing the unit activity in the activity concentration range < 100 Bq·m^{-3} in large-volume climate chambers. This is due to the fact that typical sizes of walk-in climate chambers are 10 m³ to 30 m³.

A completely new approach in traceability and thus a milestone in metrological development is represented by the device. Starting from the challenge to require high temporal resolutions and at the same time to generate high counting statistics and thus to be able to achieve low uncertainties at low activities, the highest possible realizable detection efficiency is needed. The innovative approach was to coat commercially available silicon detectors with a thin layer of ^{226}Ra: Integrated ^{226}Ra Source/Detector (IRSD). After vapor deposition, ^{226}Ra is located on the surface of the silicon detector, so that about 50% of the radon produced is released. Both the alpha particles of the ^{226}Ra and those of the remaining radon can thus be detected spectrometrically under ambient pressure with a counting yield of about 50%, separately from each other and continuously. Since the total amount of radon is a conservation quantity, the continuously recorded alpha spectra allow the amount of released radon to be determined. This type of measurement represents a mathematical inversion and requires special analytical procedures. Based on the Kalman filter for state estimation in linear dynamic systems, novel algorithms have been designed and implemented, which can be used to calculate the time course of the released activity of radon and the associated uncertainty from the continuously acquired measurement data. By the described method, even the release of one radon atom per second (about 2 $\mu Bq \cdot s^{-1}$) can be determined on average within few hours integration time still with uncertainty around 2% (k = 1). In the future, the IRSD will be used to calibrate radon monitors continuously, automatically and with feedback at radon chambers but even in the field and under changing climatic conditions.

3. NEW REFERENCE ATMOSPHERES FOR CALIBRATION OF RADON MONITORS

The importance of exposure to radon in the population and workers was noticed by the European Union where in the 2013 Council Directive 2013/59/Euratom- the Basic Safety Standards Directive (BSS) was published. A significant part of this Directive refers to the issues related to the radon hazard in dwellings and workplaces. The EU member states were required to establish national radon action plans addressing long-term risks from radon exposures in dwellings and workplaces. Hence, member states needed to establish national reference levels for indoor radon concentrations, which should not be higher than 300 $Bq \cdot m^{-3}$. This created new challenges for the radon dosimetry and calibrations because the previous limit used in certain EU member states was much higher and was 1000 $Bq \cdot m^{-3}$. Among others arose the need for harmonization of existing radon measurement procedures and creating of new ones, ensuring the traceable calibration of radon measurement instruments at low radon activity concentrations with low uncertainties. These aspects have been solved over the years, inter alia, due to research projects like MetroRADON, www.metroradon.eu .

The radon is not only addressed as the largest natural source of public exposure to ionizing radiation but also as a useful tracer for understanding atmospheric processes and estimating greenhouse gas emissions [10]. That is why reliable measurements of low-level radon activity concentrations, such as those found in the environment (< 20 $Bq \cdot m^{-3}$), are important both for organizations responsible for radiation protection and climate research. Despite the enormous changes in radon metrology that have occurred in recent years the activity concentrations below 100 $Bq \cdot m^{-3}$ have not been subject to metrological research so far. This evokes new challenges which are the development of traceable methods and robust technology for measurements of environmental low-level radon activity concentrations and radon fluxes. Both are important to derive information on greenhouse gas fluxes in the environment and therefore important for reduction strategy planning.

Properly defined closed volume is necessary to create the reference radon atmosphere. In calibration laboratories two types of radon chambers are used. The first type is a large container, typically the volume of such chambers is greater than 10 m^3. Often designed as a walk-in chamber with an air-lock, allowing entry and exit with the minimum disturbance in the radon atmosphere. The second type of radon chambers are small containers only for the equipment under test. The volumes are usually less than 1 m^3. The large radon chambers, due to their large volume, allow to perform intercalibrations of active radon monitors or calibration of devices measuring potential alpha energy concentration defined as [11]: “*The concentration of short-lived radon-222 or radon-220 progeny in air in terms of the alpha energy emitted during complete decay from radon-222 progeny to lead-210 or from radon-220 progeny to lead-208 of any mixture of short-lived radon-222 or radon-220 in a unit volume of air. The SI unit for potential alpha energy concentration is J m^{-3}*”. The smaller radon chambers are usually used to exposition of passive detectors in reference radon activity concentration. Radon chambers with a volume of about 1 m^3 represent an interesting option for calibration laboratories, allowing a successful approach for the calibration of both active and passive radon monitors.

Radon chamber is not only “radon container”, but also a system that ensure the maintenance of radon concentration inside and appropriate environmental conditions. The most frequently monitored parameters are temperature, relative humidity, atmospheric pressure, the ambient aerosol concentration, size distribution of radioactive aerosols, the radon decay products concentration and fractionalization, equilibrium factor of radium-radon gamma-ray dose or dose rate. The System for Test Atmospheres with Radon (STAR), also called “Radon Chamber”, has four inseparable parts: the equipment to containing the atmosphere, the equipment to produce the atmosphere, the reference atmosphere thus created, and the equipment and methods to monitoring the atmosphere [12]. Devices used to characterise the atmosphere shall be traceable to a primary standard.

Table 1 shows radon chambers features presented in this article. These chambers fulfil the requirements of a STAR. One essential part of the system is the reference radon monitors used to give the radon concentration inside. In the three cases exposed, the radon monitors are calibrated in the Bundesamt für Strahlenschutz (BfS), accredited by the national accreditation body for the Federal Republic of Germany DAkkS according to the ISO standard 17025, which is traceable to the national standards of the National Metrology Institute (PTB) [13]. In order to validate the two types of low-level ^{222}Rn emanation sources developed within the traceRadon project, comparisons measurements will be performed in low-level radon calibration chambers. Based on a protocol these sources are going to be measured also at IFIN-HH radon chamber. The Ionizing Radiation Metrology Laboratory (LMRI) has developed this facility in the framework of national and European joint research projects [14]. The chamber is cylindrical in shape, with a volume of 1 m^3 and has accessories used to produce radon reference atmospheres, to assure the international metrological traceability and equivalence of radon activity measurements and the possibility to perform reliable radon activity concentration measurements. Recent testing and improvement of the radon chamber tightness and traceability capacity have been done using the reference instruments and standard radon gas sources, thus obtaining designation by the Romanian Nuclear Authority, National Commission for Nuclear Activities Control (CNCAN) as Calibration Laboratory for instruments measuring radon activity concentration in air, according to the ISO standard 17025. The laboratory is performing already calibrations of in-field radon measurement instruments, belonging to various Romanian users.

Two radon chambers are located in Central Laboratory for Radiological Protection, which is the only laboratory in Poland that has accreditation (in accordance to ISO/IEC 17025) in the field of calibration of radon activity concentration and potential alpha energy concentration monitors. One of them is the walk-in radon chamber with a volume of 12.3 m^3. The chamber has the option to set the temperature and relative humidity in a wide range. The second one is a small cylindrical shape, tight container with a volume of 1 m^3. This newly developed radon chamber is planned to be used for creating low-level reference radon atmospheres using new low-activity sources developed under the traceRadon project. So far, to generate the reference radon atmospheres, two certified dry flow-through radon sources by Pylon Electronic Development Co. have been used. Their activities are 137.3 kBq and 502.5 kBq.

The radon chamber of the University of Cantabria (UC) is the only Spanish one for calibrate radon activity concentration of radon monitors and radon exposure of passive detectors according to ISO/IEC 17025 accreditation. It is a cubic shape stainless steel container with 3.25 mm wall thickness and an internal volume of 1 m^3. Access to the chamber is provided by removing the top lid of the chamber, it also allows access through three circular holes of 80 mm diameter, commonly used to introduce and remove passive detectors without disturbing the radon inside during the process. Inside, radon measurement equipment can be inserted at different heights. To homogenize the internal concentration, a fan is provided which is always working during the exposures. The radon sources available to generate the reference atmosphere are certified sources model Pylon RN-1025 (Pylon Electronics Inc.) and sources prepared from powder with high ^{226}Ra content and encapsulated in PVC jar which provide a constant radon diffusion rate. The radon monitors with traceability to international standards are the Atmos12 (Gammadata Instruments AB) and the AlphaGUARD (Saphymo GmbH). These devices are connected to a computer located outside the chamber in order to know the concentration in real time. The radon concentration inside the chamber can be controlled and modified through the use of an open-air circuit that includes a pump that extracts air from inside the chamber.

Table 1: Radon chambers features of CLOR, IFIN-HH and UC. All chambers provide services like calibration of active monitors, and exposure of passive detectors.

	CLOR	IFIN-HH	UC
V (m^3)	12	1	1
Radon concentration range (Bq·m^{-3})	100 to 50 000	100 to 10 000	250 to 11 000
Calibration and Measurement Capability (CMC) (k=2)	7%	5%	5%

Accreditation		According to ISO 17025	Designation according to ISO 17025	According to ISO 17025
Reference devices		AlphaGUARD	AlphaGUARD DF2000	AlphaGUARD Atmos12
Traceability		To BfS Reference Chamber		
Environ-mental conditions	Adjustable	Temperature, Humidity, Ambient aerosol concentration	-	-
	Monitored	Temperature, Humidity, Pressure, Size distribution of radioactive aerosols	Temperature, Humidity, Pressure	Temperature, Humidity, Pressure

4. FROM SOIL TO ATMOSPHERE: REFERENCE FIELDS FOR RADON FLUX MONITORS

The exhalation bed facility held in the Laboratory of Environmental Radioactivity, University of Cantabria (LaRUC), acts as a standard flux source of radon activity across a defined area per time. Thus, its aim is to be used to calibrate continuous radon flux systems, that can be used as a transfer standard for test validation of existing radon flux monitors by comparison campaign in field or on the same exhalation bed under laboratory conditions. The exhalation bed facility consists in two exhalation beds, one with a high radon exhalation rate and another with a low radon exhalation rate (see Figure 2). The design of each exhalation bed consists in five stainless steel welded plates shaping a box with the upper part open with an effective surface of 1 m^3. This configuration avoids the leakages through the plates and force radon to escape on the top surface. The materials chosen to constitute the exhalation beds were selected in base of its radioactive content and properties such as soil texture, structure, dry bulk density, porosity, etc. The soil for the high exhalation bed was collected from the former Spanish uranium mine located in Saelices el Chico (Salamanca, Spain), managed by the Spanish National Uranium Company ENUSA. In case of the low exhalation bed soil, it was taken from the Fos-Bucraa mine (western Sahara). The materials were dried, sieved and homogenized before putting into each container.

During the intercomparison campaigns in the framework of the traceRadon project two different areas were tested [17]. The high radon flux field is within the land of a former uranium mine managed by the Spanish Uranium Company (ENUSA), located in Saelices el Chico (Salamanca, Spain). The average soil radium concentration in this area is (814 ± 65) Bq·kg^{-1} (k=2). The reference radon flux value obtained during the intercomparison campaign was 2364 Bq·m^{-2}·h^{-1} with a standard deviation of 1172 Bq·m^{-2} h^{-1}. The low radon flux field is located in Esles de Cayón (Cantabria, Spain), a usual soil with an average radium concentration of (29 ± 3) Bq·kg^{-1} (k=2). In the low area the reference value obtained was 50 Bq·m^{-2}·h^{-1} with a standard deviation of 15 Bq·m^{-2}·h^{-1}. Moreover, the is a good agreement between the consensus values obtained experimentally and the output of the model proposed by [18] correcting the radium concentration considered using the data measured in field.

Figure 2: Picture of the Exhalation Beds: Brown, to the top: high radon flux, grey, to the bottom: low radon flux.

5. SUMMARY AND CONCLUSION

The activities developed and performed within the traceRadon project took steps forward to increase the metrological capabilities of low atmospheric radon concentrations calibration and monitoring in the range below 100 Bq m^{-3}. This includes up to now:

— the development of new primary ^{222}Rn activity standards suitable for representing the unit activity in the range below 100 Bq m^{-3} in the established walk-in radon chambers.

— the first successful calibrations in the new reference atmospheres radon monitors using the new developed activity standards.

— the development of laboratory exhalation beds tests for the calibration of radon flux monitors.

— intercomparison campaigns in two types of fields, low and high radon exhalation rates.

The results obtained in the above-mentioned activities feed into the traceRadon project with respect to radionuclide metrology and radiation protection at the environmental level. Knowledge of such joint efforts can offer a solid background in providing more accurate and traceable results for these measurement methods, being even more challenging due to the outdoor environment.

6. ACKNOWLEDGEMENT

This project 19ENV01 traceRadon has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme. The following institutes are involved as project partners in EMPIR 19ENV01 traceRadon: Physikalisch-Technische Bundesanstalt, Germany, Budapest Főváros Kormányhivatala Hungary, Cesky Metrologicky Institut, Czech Republic, Agenzia Nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile, Italy, Institutul National de Cercetare-Dezvoltare pentru Fizica si Inginerie Nucleara "Horia Hulubei", Romania, NPL Management Limited, United Kingdom, Institut Za Nuklearne Nauke Vinca, Serbia, Österreichische Agentur für Gesundheit und Ernährungssicherheit GmbH, Austria, Centralne Laboratorium Ochrony Radiologicznej, Poland, Instituto de Engenharia de Sistemas e Computadores, Tecnologia e Ciência, Portugal, Joint Research Centre - European Commission, Lunds Universitet, Sweden, Státní ústav jaderné, chemické a biologické ochrany, Czech Republic, Universidad De Cantabria, Spain, University of Bristol, United Kingdom, Universitat Politècnica de Catalunya, Spain, Université de Versailles Saint-Quentin-en-Yvelines, France, IDEAS Hungary Betéti Társaság, Hungary.

The consortium is supported by the following collaborating partners: University of Heidelberg, Germany, ANSTO, Australia's Nuclear Science and Technology Organisation, Australia, ERA, European Radon Association, Europe, Met Office, United Kingdom, University of Novi Sad, Serbia, Politecnico di Milano, Italy, University of Cordoba, Spain, EURADOS, e.V., Europe, University of Siegen, Germany, Institut de Radioprotection et de Sûreté Nucléaire, France, ARPA Piemonte, Italy, ARPA Valle d'Aosta, Italy as well as the LIFE-Respire project. The interest and support for the project extends far beyond Europe: the members of the Stakeholder Committee, for example, come from four different continents.

Thanks are due to Maria Sahagia (IFIN-HH) for some useful suggestions aimed to improve the manuscript.

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