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New metrology for radon at the environmental level

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Abstract

Radon gas is the largest source of public exposure to naturally occurring radioactivity. However, radon is also a useful tracer for understanding atmospheric processes, assessing the accuracy of chemical transport models, and enabling integrated emissions estimates of greenhouse gases. A sound metrological system for low level atmospheric radon observations is therefore needed for the benefit of the atmospheric, climate and radiation protection research communities. To this end, here we present a new calibration method for activity concentrations below 20 Bq m^{-3} and a prototype of the first portable radon monitor capable of achieving uncertainties of 5 % (at k=2) at these concentrations.

Compliance checking of policy-driven regulations regarding greenhouse gas (GHG) emissions is an essential component of climate change mitigation efforts. Independent, reliable "top down" methods that can be applied consistently for estimating local- to regional-scale GHG emissions (such as the Radon Tracer Method, RTM) are an essential part of this process. The RTM relies upon observed radon and GHG concentrations and measured or modelled radon fluxes. Reliable radon flux maps could also significantly aid EU member states comply with European COUNCIL DIRECTIVE 2013/59/EURATOM. This article also introduces the *traceRadon* project, key aims of which include outlining a standardised approach for application of the RTM, creating infrastructure with a traceability chain for radon concentration and radon flux measurements, and developing tools for the validation of radon flux models. Since radon progeny dominate the terrestrial gamma dose rate (TGDR), the planned *traceRadon* activities are also expected to improve the sensitivity of radiation protection early

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warning networks because of the correlation known to exist between radon flux and ambient equivalent dose rates.

Keywords: Radon, metrology, tracer, environmental measurements

1. Introduction to traceRadon

An overlapping need exists between the climate research and radiation protection communities for improved traceable low-level radon activity concentration (^{222}Rn) and radon flux measurements, combining the challenges of collecting, collating and modelling large datasets, with setting up new radiation protection services. The EMPIR project 19ENV01 traceRadon[1] works toward these goals for the benefit of both large scientific communities by providing the necessary infrastructure for measuring atmospheric radon activity concentrations from 1 Bq m^{-3} to 100 Bq m^{-3} and radon fluxes. In addition, it will generate data at four selected European sites for validation of radon flux models and inventories and will create the first standard protocol for applying the Radon Tracer Method (RTM). The latter is particularly important as a "top down" validation tool for "bottom up" greenhouse gas (GHG) emission inventories that help to support national reporting under the Paris Agreement on climate change, and for comparison of regional emissions across Europe. In this context, the following metrological activities for realisation and dissemination are carried out in the framework of the traceRadon project: i) development of radon activity standards, ii) calibration of low-level atmospheric radon concentration monitors, iii) development of reference infrastructure for radon flux from soil, iv) calibration of continuous radon flux monitors in the field, and v) validation of radon flux models and inventories.

All European countries operate automatic gamma dose rate systems and atmospheric radionuclide concentration detectors for environmental radioactivity monitoring. The results of this radiological monitoring are exchanged through the European Radiological Data Exchange Platform (EURDEP) as requested by EU legislation [1]. Atmospheric radon activity concentration and radon flux data is not yet routinely collected due to the current lack of ability to measure it accurately at the stations contributing to EURDEP. Information from this platform could be very useful to improve understanding of the spatial and temporal variability of atmospheric radon concentrations and terrestrial radon fluxes across Europe. For this reason, it is important to develop a robust metrological capacity, and potentially new instrumentation options, that suit the needs of different European and worldwide networks. Doing so would enable standardization of the quality assurance chain and increase the reliability of reported atmospheric radon concentration measurements that, at ambient environmental levels, can reach values between hundreds of mBq m^{-3} and tens of Bq m^{-3} [2, 3]. In addition, gamma spectrometry detectors are being installed at several European network sites that can provide indirect information on soil moisture content, a basic parameter to model radon flux [4]. The project traceRadon will contribute in this regard by characterizing and comparing several widely used monitors both for gamma dose rate [5] and full gamma spectrometry.

2. Advances in the realisation of the units Becquerel (Bq) and Sievert (Sv)

The central task of a National Metrology Institute (NMI) is to realize, maintain and disseminate the legal units of measurement in compliance with the International System of Units (SI). In each country an NMI sits at the top of the metrological hierarchy and serves its customers by issuing calibration certificates. These certificates document traceability to the SI system. On 20 May 2019, new specifications for SI units came into force. By this, the signatory countries of the Metre Convention adopted a fundamental reform of SI units. The kilogram, Ampere, mole, and Kelvin were redefined using natural constants. The last artefact, the original kilogram, is now obsolete. The kilogram, Ampere, Kelvin, and mole will now be defined by setting exact numerical values for the Planck constant, the elementary electric charge, the Boltzmann constant, and the Avogadro constant, respectively. The metre and candela are already defined by physical constants, subject to correction to their present definitions. While these changes are fundamental the overall change happened almost without being noticed because the new definitions improve the SI without modifying the size of any units. This ensures continuity with existing measurements despite the origin of traceability for each unit having shifted.

[1] 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.

2.1 Realisation of the Becquerel

The dissemination of the unit of activity is typically either achieved by the provision of activity standards or by the calibration of radioactive sources. In the case of radon (^{222}Rn), realisation of the Becquerel (Bq) is achieved by different absolute methods. One involves measurement of activity by a system that counts, in a defined solid angle, the alpha particles emitted by ^{222}Rn condensed on a cold point [6]. This method has the advantage that the respective radon activity can be unfrozen afterwards and transferred to create a reference atmosphere. It is therefore directly useable to disseminate the unit. Other methods of obtaining the Bq are available but they require a different line for dissemination, like Liquid Scintillation Counting (LSC) or proportional counter methods.

Considering the small uncertainty assigned to these methods, emanation standards were not able to compete with them for a long time [7]. This recently changed in the case of very low activities [8]. Source emanation is determined by measuring the activity of radium (^{226}Ra) progeny remaining in the radium source. The highest detection probabilities are needed for the low activities required to establish a traceability chain for the environmental atmospheric radon measurement, . This inspired a new technological approach: an integrated source-detection system operated in real time monitoring mode. For the first time, a novel combination of source and detector developed at Physikalisch-Technische Bundesanstalt (PTB) is used in the scope of the traceRadon project. For this purpose, an ion-implanted silicon semiconductor detector is coated in a defined manner with radium chloride ($^{226}RaCl_2$), by means of thermal vapor deposition, directly onto the dead layer of the detector. Thus, the detector is simultaneously both the source of radon and the spectrometric detector for the resulting alpha radiation, see Fig. 1.

Both the absolute activity of ^{226}Ra and the loss of ^{222}Rn can be determined directly by analysis of the α-spectrum. This yields the absolute activity of ^{222}Rn emanating from the integrated source-detection system.

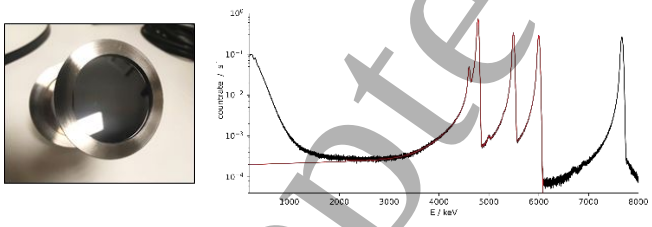


Figure 1: The integrated source-detection system: On the left, an ion-implanted silicon semiconductor detector, coated in a defined manner with $^{226}RaCl_2$ by means of thermal vapor deposition; on the right, the resulting α-spectrum. The respective peaks are well resolved. For the tailing contributions a model with mixtures of exponentially modified Gaussians is used.

2.2 Realisation of the Sievert

The traceability chain for dissemination of the Sv is not so easily explained, because this unit is used for more than one operational quantity. Here, only the ambient dose equivalent, the quantity for area monitoring, is considered. According to the International Commission on Radiological Protection (ICRP) [9], the ambient dose equivalent is defined as *"The dose equivalent at a point in a radiation field that would be produced by the corresponding expanded and aligned field in the ICRU sphere at a depth of 10 mm on the radius vector opposing the direction of the aligned field."* The ambient dose equivalent is given the symbol $H^*(10)$. The SI unit of $H^*(10)$ is the sievert (Sv).

The realisation and dissemination of the unit is done by reference fields for the radiation quality of interest (radiation type and energy range), which are defined for photons by the International Organization for Standardization in [10]. It is important to note that the quantity of interest which is determined in the unit Sv is in the following the ambient dose equivalent for photons. This results in a dose rate $dH^*(10)/dt$ for photons (e.g., γ-rays emitted by the decay of naturally occurring radionuclides in the environment) in Sv h^{-1}. Though there are several detection principles available, the challenge is to measure this quantity without interference from cosmic radiation, which creates an overresponse in most commonly used systems [11, 12, 13].

From the radiation protection point of view, the realisation of the traceability chain for Sv is important for the National Surveillance Networks which get the outdoor gamma radiation levels usually given as ambient equivalent dose rates $H^*(10)$ measured in nSv h^{-1}.

3. New reference infrastructure for environmental radon activity concentration, radon flux and dose rate measurements

3.1 Radon activity concentration

Traceability of activity concentration (Bq m^{-3}) in the concept of the revised SI system means that the basic units second (s) and metre (m) are defined by fundamental constants and are realised by an NMI. To realise the unit, the conventionally true value of the quantity to be measured has to be estimated, in this case the radon activity concentration. Afterwards this information is used to determine the calibration factor of a device. Comparison of different instrument results are only possible if all instruments are traceable to the SI system. The uncertainty of a measurement in the field will always include the uncertainty of the calibration. It is an intrinsic part of the quality of the

measurement, like the instrument itself, and should be chosen with great care.

~~Traceable calibrations enable the comparision of results, especially when they are made with different instruments.~~ For the instrument, the following physical characteristics are required before starting a measurement campaign: range of application, detection limit, traceability and uncertainty.

Until recently state of the art was the application of one of the following three procedures for the calibration of ^{222}Rn monitors -: a primary method based on a reference activity concentration realised by a primary radon gas standard and a calibration volume (both values are traceable to national standards); a secondary method based on calibration via a reference monitor enclosed in the same atmosphere as the system under test; and a primary/secondary calibration in a constant atmosphere based on a radium emanation source. The latter method being primary or secondary with respect to the components used.

Institutes that operate one or more of these methods have to be traceable according to the list given in the Calibration and Measurement Capabilities of the Bureau International des Poids et Measures (BIPM). All methods provide valid calibration factors [14, 15,16].

To be able to calibrate an instrument in a range of atmospheric radon activity concentration, the activity concentration of the reference atmosphere should either be stable, or the system under test needs to have a sufficiently high statistical response (sensitivity) to achieve small statistical uncertainties by its reading. As an example of this requirement, Fig. 2 summarises an evaluation of two systems under test with contrasting response characteristics (AlphaGUARD PQ2000Pro, Bertin Instruments; and 200 L two-filter dual-flow-loop monitor, ANSTO) to a build-up and decay phase of radon activity concentrations in a closed large-volume climate chamber at PTB. The activity concentration is generated by an emanation source (from section 2.1) in this known volume. This measurement proves that a fourth calibration procedure exits: a primary calibration in a non-constant atmosphere based on a radium emanation source if the system under test has a high sensitivity.

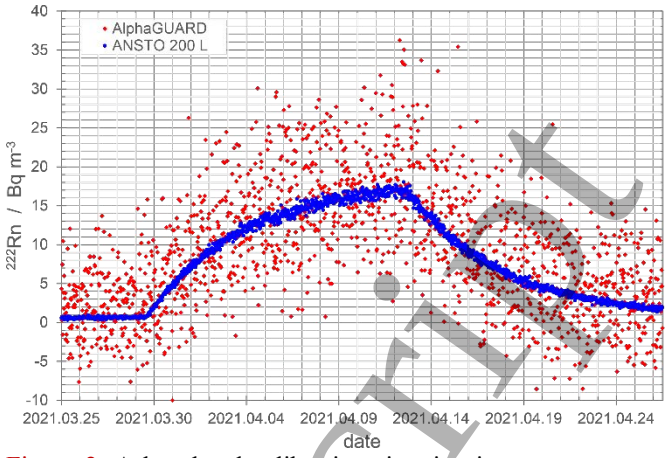


Figure 2: A low-level calibration situation in a non-constant atmosphere: The adjusted response of two types of detectors (AlphaGuard in red, ANSTO 200 L in blue) in a build-up and decay situation in a closed large-volume climate chamber at PTB.

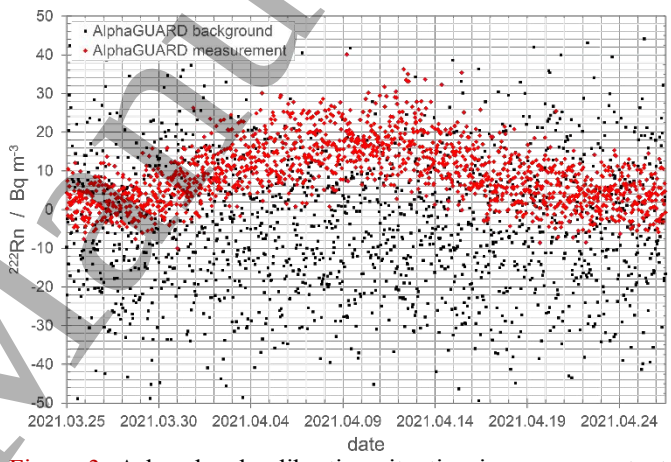


Figure 3: A low-level calibration situation in a non-constant atmosphere: One AlphaGuard in red from Fig. 2 in a build-up and decay situation in a closed large volume climate chamber at PTB. The activity concentration outside the climate chamber is given by a second AlphaGuard in black.

This preliminary primary measurement procedure resulted in a calibration factor of (26.0 ± 1.3) s Bq m^{-3} with a sensitivity of (0.0385 ± 0.0020) s^{-1} Bq^{-1} m^3 (reciprocal of the calibration factor) both for k=2, for the new ANSTO 200 L radon monitor, further described in section 4 [17]. The measurement uncertainty will be reduced even further after the intrinsic background and the radon background in the climate chamber have been determined more accurately. In contrast to this, Fig. 3 shows that the response of the AlphaGuard from Fig. 2 does not fullfill the requirements to be calibrated in a non-constant atmosphere considering the signal to background reading given in Fig. 3.

3.2 Radon flux

Fundamentally, the ^{222}Rn activity concentration in soil and its emission rate depend on the geology of an area, the porosity and permeability of the soil, the terrain structure and the associated ^{238}U mineralization. After its formation by ^{226}Ra decay ^{222}Rn escapes from soil pores to the atmosphere by diffusion. Radon exhalation from soil, also referred to as radon flux, is measured in activity (Bq) per unit time (s) and surface area (m^2). Direct measurements of radon flux are ideally made using the accumulation method [18, 19, 20], which is based on the measurement of ^{222}Rn gas accumulation in a chamber placed on the soil over a set time period.

The ability to measure radon flux from the ground, or other surfaces, can be useful for (i) application of the RTM; (ii) validation of radon flux models and inventories [4]; (iii) meeting the regulatory requirements for uranium mill tailings or phosphogypsum stacks [21], (iv) determining the radon emanation from building materials such as bricks, concrete and other surfaces [22, 23], and (v) identification of indoor radon risk areas. While present theoretical understanding of the processes controlling the release of ^{222}Rn from soil to the atmosphere is quite extensive [24, 25], the uncertainties associated with experimental measurement have not been fully evaluated. Some of these uncertainties are related to changing environmental conditions during the measurement period imposed by the chamber (e.g., temperature and air pressure inside the chamber) as well as with the chamber setup parameters (e.g., depth of the accumulation chamber or associated collar within the soil).

Aside from chamber effects, there are a range of other interrelated meteorological factors that affect the radon flux from a soil surface, including wind speed, atmospheric pressure, soil moisture, and soil temperature. It is difficult to quantify the change in radon exhalation due to any of these factors individually because they are interrelated (while a precipitation event increases soil moisture, it is often also associated with a drop in pressure, a drop in temperature, and a change in wind speed and direction). It is necessary to consider the importance of these factors when determining the optimum flux measurement system [26].

The traceRadon project aims to investigate the influences mentioned above and create infrastructure with a traceability chain enabling a correct radon flux measurement from the soil. In the scope of this project an exhalation bed facility has been designed and built, together with a radon flux reference system, to calibrate other accumulation chambers based on different designs and components.

3.3 Dose rate

A recognisable correlation (~60%) between radon flux and Terrestrial Gamma Dose Rate (TGDR) was first reported by [27] during their extensive campaign using Berthold Geiger counter dosemeters at a variety of locations throughout the entire continent of Australia. In [28] the empirical relationship obtained in [29] was applied to the extracted TGDR from the dense EURDEP network which currently comprises 5500 stations in 34 Countries across Europe monitoring outdoor radiation levels. The response of the instruments operated in these networks is subject to changes in the concentration of radon progeny either induced by changes of the radon activity concentration or by changes in the environmental conditions. The National Surveillance Networks obtain outdoor gamma radiation levels usually given as ambient equivalent dose rates $H^*(10)$ in nSv h^{-1}, and consisting in contributions from the following major sources: (a) secondary cosmic radiation, (b) terrestrial gamma radiation, (c) radiation from airborne and deposited daughter progeny of radon and thoron (^{220}Rn) attached to aerosols, and (d) inherent background of the detector.Besides these sources, the site characteristics should also be considered: close buildings, trees and other disturbance objects are important sources of bias in the ambient dose estimation. Therefore, the measured values should be corrected in order to get normalized results (with reference to an "ideal" site). When not properly corrected, systematic errors in the estimated TGDR may occur. Including intruments for the measurement of radon activity concentration at these sites will reduce these systematic errors in future. Here, the new metrology in the radon field leads to a new quality in the surveillance of the ambient equivalent dose rates. Due to the complexity of determining correct values from ambient dose rates acquired at monitoring stations an alternative method was used [30]. This method is based on deriving the TGDR from the activity concentrations of natural radionuclides in the soil (uranium, (^{238}U), thorium (^{232}Th), and potassium (^{40}K)). Knowing these activity concentrations, the TGDR values were calculated using conversion coefficients (nSv h^{-1} per Bq kg^{-1}) from [31].

Considering the increased number of gamma spectrometric detectors installed in National Surveillance Networks, another promising approach is the possibility to use these detectors to infer soil moisture, which is one of the most relevant parameters in the estimation of radon flux. [32] used Monte Carlo simulations to investigate the potential to estimate soil water content with NaI spectrometric measurements. The reliability of the simulations was validated experimentally with known soil water contents and radionuclide abundances.

As part of the traceRadon project an intensive campaign with selected dose rate monitors and spectrometric detectors will be carried out at four European sites to analyse the application of the previous models and also to study the use of the spectrometric detectors. Once the detectors have been selected, the next step will be to characterize them by experimental exposures and by Monte Carlo simulations. The inherent background, response to secondary cosmic radiation and to different radionuclides will be carried out at PTB's facilities. A pure photon reference field for the calibration of dose rate monitors is available at PTB: the low-level

underground laboratory for dosimetry and spectrometry (UDO II). Located 430 m beneath the surface in the Braunschweig-Lüneburg salt mine of the "European salt company" (esco), the muon component of the secondary cosmic radiation (SCR) at UDO II is suppressed by four orders of magnitude compared to the muon flux at the surface (sea level) due to the shielding effect of the rock overburden. The very low activity of rock salt, combined with low mean radon levels in the mine atmosphere, leads to an ambient dose equivalent rate of only (1.4 ± 0.2) nSv h^{-1} at the reference point (compared to about 100 nSv h^{-1} at the surface). Furthermore, the laboratory provides a photon calibration facility. Irradiation of the detectors is performed by using different radiation qualities (^{57}Co, ^{60}Co, ^{137}Cs, ^{226}Ra, ^{241}Am) which are traceable to primary standards. The photon fields of this calibration facility are collimated, to minimize scattering by the surroundings (walls, floor, etc.) at the reference point, 2 m from the sources. For complete characterization of a dosemeter additional response information is needed. In general, the reading A of a dosemeter is composed of the inherent background A_0, the response to terrestrial radiation $q_{Terr}\ H_{Terr}$, the response to secondary cosmic radiation $q_{SCR}\ H_{SCR}$ and artificial radiation $q_{Art}\ H_{art}$

$$A = A_0 + q_{Terr}\, H_{Terr} + q_{SCR}\, H_{SCR} + q_{Art}\, H_{Art}\ .$$

The state-of-the-art in calibrations, including detemination of the instrument response q_{Terr} , q_{SCR} , q_{Art} in the environmental range, is summarised in [13]. The dose rate systems in radiological early warning networks try to detect H_{Art} while the two components H_{Terr} and H_{SCR} vary naturally. The largest variation is induced by rain events, which concentrate captured radon progeny (polonium (^{218}Po), lead (^{214}Pb), bismuth (^{214}Bi)) near the surface. All contributions can be determined at the four PTB measuring sites: the free-field reference dosimetry site for environmental radiation, the dosimetry site for cosmic radiation, UDO II, and a plume simulation site that has been made available to study the effect of small dose rate changes on top of the natural background radiation resulting from rain events [33, 34].

Another underground European facility is under development in Romania, at IFIN-HH. The ultra-low background "microBequerel" laboratory is located within the former salt mine Unirea, from Slanic-Prahova (about 100 km north of Bucharest). The Unirea mine is about 210 m deep and was used for salt exploitation from 1943 until 1970. Its unique characteristics provide great environmental conditions for ultra-low background dose rate monitor calibrations. According to [35], the average underground dose rate was found to be (1.17 ± 0.14) nSv h^{-1}.

4. Application and impact on greenhouse gas (GHG) monitoring networks

Bringing new scientific methods and findings to bear on GHG mitigation policy is a clear 'new challenge' for the traceRadon project. The greatest opportunity for traceRadon to realise this goal is by helping to reduce the uncertainty associated with integrated local- to regional-scale GHG emissions estimates, for which the starting point is: (i) accurate GHG amount fraction observations, (ii) spatially and temporally representative radon flux maps, and (iii) widely available low-level radon activity concentration observations from instruments with traceable calibrations. Here, the existing GHG networks, the necessity and current availability of atmospheric radon measurements within these networks, as well as the importance of improving the quality and traceability of such observations are discussed.

The Integrated Carbon Observation System (ICOS) is Europe's state-of-the-art research infrastructure providing highly standardised, robust, in situ data and elaborated data products on the carbon cycle, and quantifying GHG emissions and sinks across Europe. Their atmospheric network includes stations in 13 European countries (see Fig. 4). Currently, 30 stations are labeled while 8 others are in various stages of the labeling process but are measuring GHG concentrations and in some cases already providing these data to the ICOS database. Each of the current atmospheric stations measures GHG concentrations (at least carbon dioxide, CO_2, and methane, CH_4), as well as the major meteorological parameters ('class 2' sites). Moreover, many of the sites, also measure additional observables such as carbon monoxide (CO), stable isotopic ratios, radiocarbon and (oxygen/nitrogen) O_2/N_2 ratio [36]. These measurements are usually made from near the tops of tall towers (typically >100 m above ground level), in mountainous terrain, or in remote environments. While radon is also a recommended measurement quantity at ICOS sites, due to its recognised value as a tracer of transport and mixing, as yet there is no standardised rule or protocol governing the integration of such measurements into the processing of ICOS data or in the downstream use of ICOS data (e.g., for top-down emissions estimation).

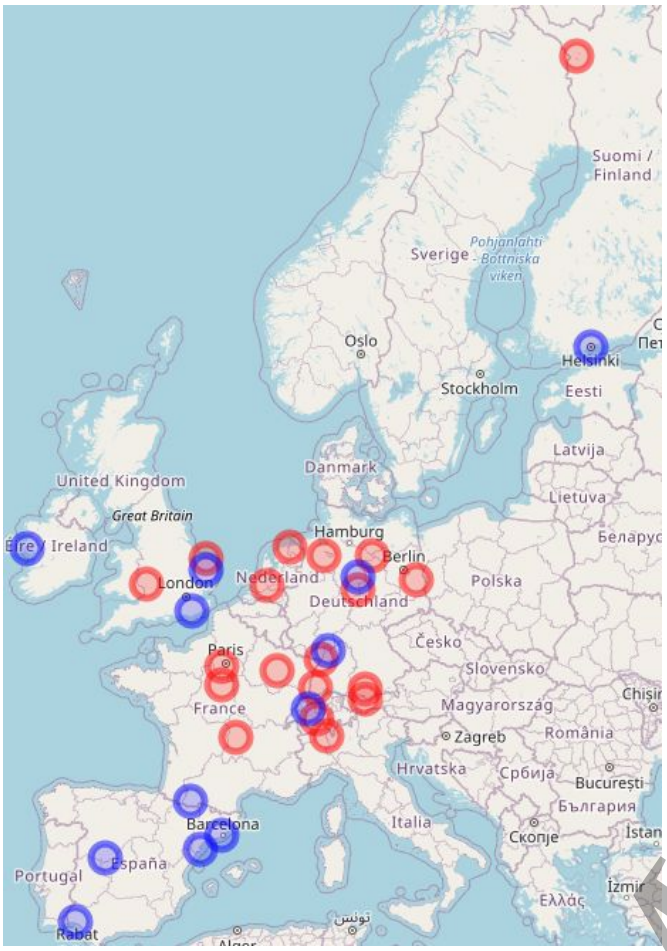


Figure 4: Location of the 31 European sites measuring atmospheric radon measurements. 19 sites are part of ICOS Atmosphere (red circles). The others in blue circles are either part of AGAGE, WMO-GAW or are not affiliated to a GHG network.

ICOS is not the only scientific enterprise measuring atmospheric composition in Europe. Several other in situ atmospheric composition networks exist (e.g., NOAAs Global Greenhouse Gas Reference Network; Scripps CO_2 programme; and the Advanced Global Atmospheric Gases Experiment), the oldest of which were initiated to understand global trends in GHGs. The World Meteorological Organisation (WMO) Global Atmospheric Watch (GAW) programme also recognise observation sites for their importance in atmospheric composition monitoring on regional and global scale, many of which are part of the specific networks mentioned above. More recently there have also been attempts to measure urban-scale emissions (e.g., the Urban Test Bed project developed by the US national measurement institute, NIST). Each of the variety of monitoring network stations has its own challenges regarding the translation of atmospheric GHG amount fraction measurements into useful, policy-relevant estimates of GHG fluxes. As well as making the best observations of GHG amount fractions (and of related tracers), a major challenge for ICOS is making additional measurements that will help the modelling community (e.g., from the VERIFY project, verify.lsce.ipsl.fr) translate GHG observations into accurate measurements of GHG fluxes.

Some 'top-down' GHG estimation methods use observations of changes in atmospheric composition, and prior information on flux magnitudes and distributions [37], typically with atmospheric transport models (together with inverse methods). Despite the robust mathematical approach of these methods, there are still large uncertainties involved, e.g., estimations of the boundary layer height, modelled transport in the atmosphere, errors related to the resolution of the model, and how all these pieces of information are combined in a statistically rigorous way.

With a view to reduce these uncertainties, a purely measurement-based method for estimating spatially-integrated GHG emissions could include the use of a surface-emitted atmospheric tracer with appropriate physical properties (e.g., simple source and sink charactertiscs) that had a spatially distributed source function. Radon (^{222}Rn) has been proposed as a potentially suitable observable for this task. Being an inert gas, ^{222}Rn does not chemically react with other atmospheric constituents, it is not involved in biogenic processes, and its low solubility makes it unlikely to be washed out by rainfall. Despite the spatial heterogeneity of its long-lived parent (^{226}Ra, $T_{1/2}$~1620 years) and soil permeability, radon's flux to the atmosphere can be modelled with smaller uncertainties than GHG fluxes. Furthermore, its only atmospheric sink is radioactive decay ($T_{1/2}$ ~3.82 days), making ^{222}Rn an approximately conservative tracer on sub-diurnal timescales, but preventing it from accumulating in the atmosphere on greater than synoptic timescales [38]. When emitted into the atmosphere, ^{222}Rn experiences the same atmospheric circumstances (i.e., transport and dilution through mixing) as all other gases with near-surface sources [39, 40, 41]. Consequently, if the ^{222}Rn flux is known, and its atmospheric concentration measured, their ratio can be determined and subsequently applied to estimate surface fluxes of other species from their observed atmospheric mixing ratios in a consistent and representative way [42].

Atmospheric radon measurements have already been used in several studies to estimate local- to regional-scale surface fluxes of GHGs to/from the atmosphere, e.g., the 'radon tracer method' (RTM) [43, 44, 45, 46, 47]. RTM is a box model approach built on the following equation:

$$j_{GHG} = j_{Rn}\ \Delta C_{GHG}\ \Delta C_{Rn}^{-1} f$$

where j is the flux, ΔC is the departure of atmospheric concentrations from regional background conditions, and f is the factor accounting the decay of ^{222}Rn during transit time. One application of this method relies on nocturnal

observations, conducted within the stable nocturnal boundary layer under non-advective conditions, when atmospheric inversion conditions inhibit mixing and allow gases to accumulate.

While such a method already exists, implementation of the RTM in a way that is comparable between different sites first requires traceability of the contributing radon measurements.

High sensitivity atmospheric radon measurements are available at 30 key European atmospheric monitoring stations, 16 of which are ICOS stations, see Fig 4.1. These radon measurements are currently being made by a wide range of different instruments, based on different measurement techniques, [48, 49], see Fig. 4.2. The most significant difference in measurement approach between the systems currently in operation is whether they make direct measurements of radon gas, or infer radon concentrations through measurement of ambient radon progeny. To achieve a consistent and reliable use of atmospheric radon measurements by the climate research community, it is imperative that every effort is made to harmonize observations across the multitude of platforms described in [48, 49] by building a chain of traceability for their calibrations and developing a full budget of their measurement uncertainty for atmospheric radon concentration. Consequently, objectives of the traceRadon project include evaluating the comparative performance of several portable atmospheric radon monitors and to develop traceable calibrations for them at very low atmospheric radon activity concentrations (e.g., 1 Bq m^{-3} to 100 Bq m^{-3}), with a view to select one or two of these instruments for use as calibration transfer standards to provide traceable measurements across the growing European radon monitoring network. For instrument selection, the type of sampling has to be considered: the so-called direct monitors remove all ambient progeny from the air, then let only radon gas and aerosol-free air into the measurement chamber. The indirect monitors, based on the measurement of radon progeny, do not do this and are dependent on the equilibrium factor as a result. Since the response times of direct radon monitors typically excede those of indirect monitors, it will be necessary to apply response time corrections to observations from these kinds of detectors when applying the RTM to validate emissions inventories.

The direct monitors of ANSTO (700 L and 1500 L dual-flow-loop two-filter radon monitors) are the most commonly used detectors within the European radon network, see Fig. 5. Their popularity is largely attributable to their low detection limit (0.040 Bq m^{-3} and 0.025 Bq m^{-3}, respectively), low statistical uncertainty, and low maintenance requirements. The response time of these instruments to step changes in radon concentration is around 45 minutes, which can be corrected for in post processing through deconvolution of the time series, as described by [50]. The indirect (radon progeny) Heidelberg University radon monitor (Levin et al., 2002) is also running at a number of European stations. In addition, several Spanish stations are currently using the direct monitor ARMON (Atmospheric Radon MONitor) [51] which can measure both ^{222}Rn and ^{220}Rn with 30-minute temporal resolution, and allows a full alpha spectral analysis of each measurement.

Figure 5: Atmospheric radon research detector types (from ANSTO, UPC, UHEI and other developers) currently used at the European atmospheric monitoring stations for research. In total there are 38 research monitors in use at 30 different sites.

In the scope of traceRadon, two direct monitors, a portable (200 L) version of the ANSTO monitor and an optimized ARMON (v2) have been designed and built.

The first instrument that was developed within the traceRadon project is a novel, portable (200 L) direct (radon gas) dual-flow-loop two-filter radon monitor. This instrument has a 30-minute temporal resolution and a sensitivity determined in section 3.1 that, for the first time, is traceable to the new sources. While it only measures ^{222}Rn, this instrument is fully remotely controllable, can fit within a 19" instrument rack, has low power requirements (~100 W at 240 VAC), is suitable for low-maintenance long-term indoor or outdoor operation, records internal environmental paramaters for STP and water vapour correction of radon concentrations, and has the capability to perform calibrations/instrumental background checks automatically in situ.

As shown in Fig 2, full characterisation of the sensitivity and uncertainty of the new 200 L ANSTO radon monitor under controlled conditions is underway at PTB. Shortly after construction, however, an approximate check of the performance of this instrument in the field was conducted by comparison with a 1500 L ANSTO monitor near Sydney, Australia, for atmospheric radon activity concentrations between 0.2 Bq m^{-3} and 8 Bq m^{-3}. The performance of the new monitor closely matched that of the larger monitor (Fig. 6), although its response time was faster. At "baseline" monitoring stations [52, 2], however, where atmospheric radon activity concentrations are frequently below 0.15 Bq m^{-3}, use of one of the larger model ANSTO radon monitors is recommended.

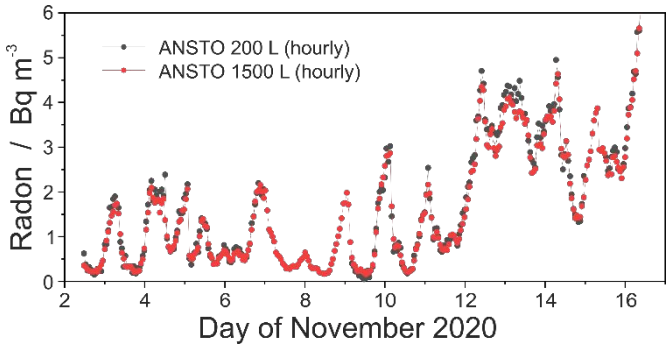


Figure 6: Two-week comparison of hourly atmospheric radon activity concentration observations between a 200 L and 1500 L ANSTO radon detector from 3 m above ground level at a site near Sydney, Australia, 15 km from the coast.

The second instrument that was developed for the traceRadon project is from the Institute of Energy Technologies (INTE) of the Universitat Politècnica de Catalunya (UPC). Originally it was designed in the mark of the project 'High efficiency monitor of atmospheric radon concentration for radiation protection and environmental applications' (MARE^{2}EA, reference: 2019-LLAV-00035) funded by the Catalan Agency for Management of University and Research Grants. Progress within traceRadon has been made to develop the detection, acquisition and drying sample modules of a new pre-prototype instrument [53]. The new modules were based on the previous model of the Atmospheric Radon MONitor (ARMON) [50] used for the measurement of atmospheric radon and thoron (^{220}Rn) concentrations. Laboratory experiments were performed at the INTE-UPC radon chamber [54] to test the PIPS detector, the detection volume, the electronics, the high voltage and the drying system components. In addition, a theoretical study of the electrostatic field generated within the detection volume was performed to improve its geometry and maximize collection of the ^{218}Po and ^{216}Po on the detector surface. Finally, a GUI has been created to remotely control the different modules and to visualize the results in real time. Preliminary results (an example is shown in Figure 7 for radon activity concentration below 100 Bq m^{-3}) indicate a sensitivity of this pre-prototype of about 0.006 s^{-1} Bq^{-1} m^3 for radon concentration, with a detection volume of only 20 L.

On the basis of this previous work, and in the mark of the traceRadon project, the INTE-UPC is now building a new version of the ARMON monitor now. It will be portable, thanks to a smart case, completely user friendly, and include real-time monitoring of radon concentrations and environmental parameters within the detection volume. Fig. 8 shows the simulated electric field within the detection volume using the COMSOL Multiphysics® software and the user panel created by LabView software.

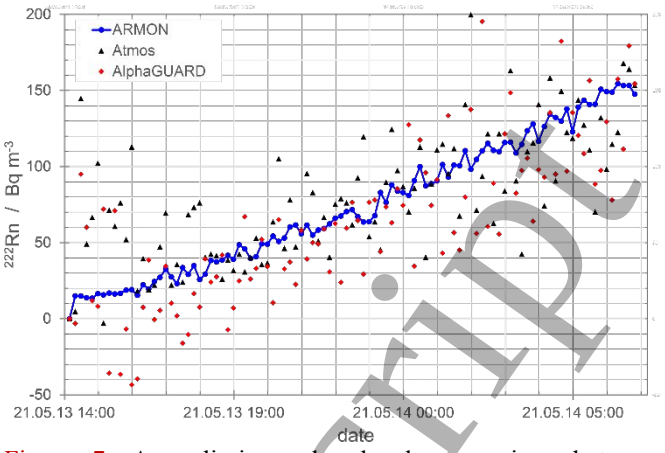
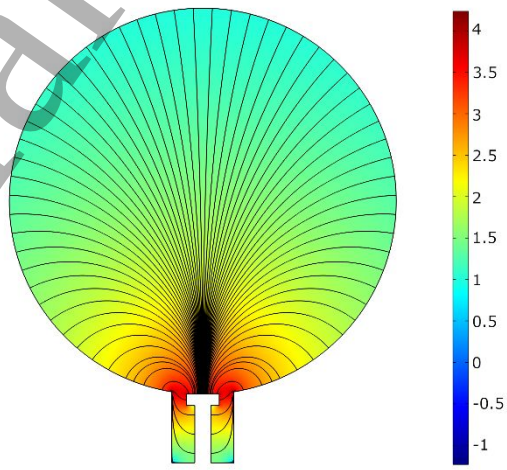


Figure 7: A preliminary low-level comparison between ARMON (in blue) and two commercial monitors (ATMOS and AlphaGUARD) (in black and red) carried out within the INTE-UPC radon chamber during an activity concentration build-up phase. INTE-UPC operates a radon chamber, categorized as a STAR according to [55].



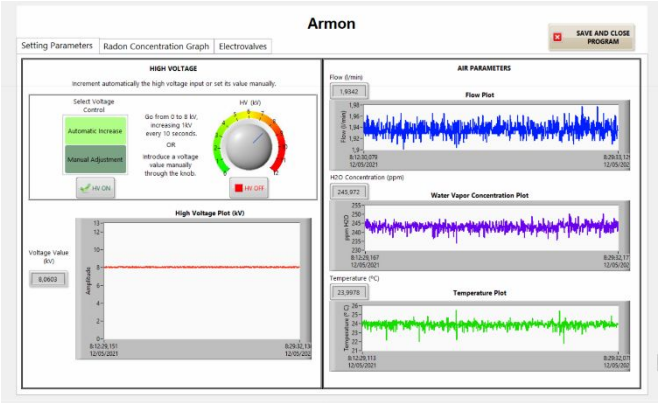


Figure 8: Upper part: Simulation of the electric field (kV m^{-1}) generated within the detection volume of the ARMON v2; Bottom part: User Interface of the ARMON v2.

The relative performance (signal stability, response time, sensitivity to changes in environmental parameters) of both the new 200 L ANSTO and ARMON v2 monitors will be compared within the traceRadon project under field conditions at the Saclay ICOS station as part of a long-term intercomparison experiment under a range of environmental conditions. In addition, a full budget analysis of the uncertainty of the measurement performed with these monitors for radon concentrations below 100 Bq m^{-3} will be performed.

An additional consideration for reliable application of the RTM is that radon soil fluxes have to be well known in time and space. The current radon flux models [4] and inventories will be validated through new low-level traceable radon flux measurements in the field, atmospheric radon measurements and Numerical Atmospheric-dispersion Modelling Environment sensitivity maps (from radon specific back trajectories that include decay correction).

To this end, the traceRadon project aims to develop a traceable metrology chain from lab to field, and by this, provide reliable data that which will allow the creation of a "first standard protocol" for application of the RTM on different spatial scales (local and regional). Within this protocol, a full budget of uncertainties will be produced and will be treated the same way for stations throughout Europe. The first standard protocol of RTM will enable long-term verification of GHG inventories.

5. Application and impact on radiation monitoring networks

Council Decision 87/600/EURATOM, 1987 [1] specifies that the results of radiological monitoring must be made available to the European Commission and all potentially affected member states. To carry out this delicate task, the EURDEP has been developed and improved over the past 30 years. EURDEP is a system for the exchange of radiological monitoring data from automatic surveillance systems in 39 countries in almost real time [56]. Assuming no radiological events occur, these measurements, in the form of gamma dose rate, essentially reflect the natural background gamma radiation from approximately 5500 fixed sensors. In addition, data from a few hundred air samplers and meteorological stations are exchanged. Fig. 9 shows the location of gamma dose rate sensors at the moment of writing this paper in some of the European countries. Significant differences in the configuration of national networks are evident.

Figure 9: The location of gamma dose rate sensors in some of the European countries in spring 2021.

The EURDEP provides current and continuous information available on https://remon.jrc.ec.europa.eu/. However, the system does not have a primary alerting role, and hence, cannot automatically be taken as an indication of increased levels of radioactivity without prior consultation with the data provider.

In the European Council directive 2013/59/Euratom, Article 103, Paragraph 3 states that Member States should identify areas where it is expected that annual average indoor radon concentration will exceed national reference level in a significant number of dwellings [57]. These areas are often called "Radon Priority Areas" (RPA). The delineation of these areas will allow the planning and prioritisation of measures within the national action plan, and has implications in that radon measurements in workplaces located in these areas may be required [58]. Further to legally binding requirements, such a prioritisation can also be useful for radon prevention for new buildings (for example, through specific building codes), as well as the promotion of actions aimed at reducing exposure to radon [58].

RPAs are most commonly estimated from indoor radon data, but geogenic data (i.e., uranium concentration in the ground, terrestrial gamma dose rate, geological units, soil units and others) could be used instead, or in addition [59]. These predictor or proxy quantities are physically and statistically related to indoor radon quantities and are at the base of the concept of Geogenic Radon Potential (GRP) [60]. Therefore, atmospheric radon concentration and radon flux are geogenic data useful to estimate the GRP and to identify RPAs.

In radioactivity monitoring it is fundamental to avoid false positives and to be as precise as possible in the determination of radioactivity amount. The EURDEP could be considered an example of a network susceptible to such problems. During or after rain and snow, solid radon progenies formerly distributed throughout the lower atmospheric column can be concentrated at, or near, the ground surface. This deposition of radionuclides can generate relatively high short-lived peaks in the radioactivity detected, called radon wash-out or simply radon peaks. These peaks in ambient dose rate can create false

positive responses in a network such as the EURDEP. Atmospheric radon and radon flux data could help to quantify, simulate and potentially remove such radon-wash out peaks [32, 33]. This will help both to prevent false alarms and to improve the detection in case of radiological/nuclear accidents by enabling the reduction of the detection threshold.

6. Conclusions and Outlook

Climate change and radiological protection both affect humankind and the environment, worldwide. In the combat against both climate change and radiation exposure [30,61, 62], measurements must be supported by traceable metrology infrastructure providing reliable data for scientists and decision makers. The new radon metrology presented here implements traceability at the environmental level and improves radon concentration monitoring and radon flux maps.These maps will help Europe to meet the World Health Organisation (WHO) and International Atomic Energy Agency (IAEA) requirements for access to validated and reliable radon exposure data according to geographical criteria. In the first step, new sources and new technology have been developed. For the first time, a source detector combination is available and two new radon monitors have been designed and built. The traceability chain is currently being extended to field measurements. If traceability is implemented there, reliable data will be produced. This is fundamental for scientific developments, application of new technologies (AI application and data mining) and steering of resources.

In terms of the outlook for future developments and data use, it can be stated that measurements of radon and its progeny have the potential to provide valuable and rare climate information that is typically not available in high temporal and spatial resolution, such as soil moisture [63, 64], or precipitation type and aerosol load [65]. Improved measurements of atmospheric radon, particularly when combined with standard meteorological observations and innovative data analysis approaches, will enable that potential to be fully realized.

Improved radon observations impact studies of surface-atmosphere exchange processes, both over land and in the marine boundary layer. In marine settings, as oceanic radon sources are typically 2-3 orders of magnitude less than their terrestrial counterparts, sufficiently accurate measurements of atmospheric radon can be used to track terrestrial influences, distinguishing the air masses influenced by land-atmosphere exchange processes from the air masses that have been in contact only with the ocean [51, 37]. Over land, particularly in environments of high release of gases from the surface, such as in volcanic settings or hot springs, radon is used for surveillance monitoring and as a proxy of other gases, such as CO_2 [66, 67]. Improved capability to measure radon in the atmosphere will contribute to better quantify surface-atmosphere exchange processes in these environments and improve understanding of natural hazards.

Furthermore, improved capability to measure radon in the atmosphere impacts the ability to understand atmospheric characteristics that are extremely relevant for climate studies, such as atmospheric ionisation and the atmospheric electric field [68, 69]. Ion production in the atmosphere is mainly driven by ionising radiation, and near the Earth's surface radon and its progeny are the major sources of ionising radiation. The electrical properties of the atmosphere play a fundamental role in climate, as they influence droplet formation and cloud microphysics, and thus earth's radiative balance. Improved radon observations will enable improved understanding of the connection between ambient radioactivity and atmospheric electricity.

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