Edinburgh Research Explorer

New metrology for radon at the environmental level

New metrology for radon at the environmental level

View the article online for updates and enhancements.

New metrology for radon at the environmental level

Annette Röttger[1,*] , **Stefan Röttger**[1] , **Claudia Grossi**[2] , **Arturo Vargas**[2] ,
Roger Curcoll[2], **Petr Otáhal**[3] , **Miguel Ángel Hernández-Ceballos**[4] , **Giorgia Cinelli**[5],
Scott Chambers[6] , **Susana Alexandra Barbosa**[7] , **Mihail-Razvan Ioan**[8] ,
Ileana Radulescu[8] , **Dafina Kikaj**[9] , **Edward Chung**[9,10] , **Tim Arnold**[9,10] ,
Camille Yver-Kwok[11] , **Marta Fuente**[11] , **Florian Mertes**[1] and **Viacheslav Morosh**[1]

[1] Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany
[2] Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, 08028 Barcelona, Spain
[3] National Institute for NBC Protection, 26231 Milin, Czech Republic
[4] Department of Physics, University of Córdoba, 14071 Córdoba, Spain
[5] European Commission, Joint Research Centre, 21027 Ispra, Italy
[6] Australian Nuclear Science and Technology Organisation, 2234 Lucas Heights, Australia
[7] INESC TEC, 4200-465 Porto, Portugal
[8] Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, 077125 Magurele,
Romania
[9] National Physical Laboratory, Teddington, Middlesex, United Kingdom
[10] School of GeoSciences, University of Edinburgh, Edinburgh, United Kingdom
[11] Laboratoire des Sciences du Climat et de l'Environnement, (LSCE-IPSL), CEA-CNRS-UVSQ,
Université Paris-Saclay, 91191 Gif-sur-Yvette, France

E-mail: annette.roettger@ptb.de

CrossMark

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 modeled 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 standardized approach for application of the RTM, creating infrastructure with a traceability

---

* Author to whom any correspondence should be addressed.

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, the planned *traceRadon* activities are also expected to improve the sensitivity of radiation protection early warning networks because of the correlation known to exist between radon flux and ambient equivalent dose rates.

Keywords: radon, metrology, tracer, environmental measurements

(Some figures may appear in colour only in the online journal)

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 modeling large datasets, with setting up new radiation protection services. The EMPIR project 19ENV01 traceRadon[12] 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 realization and dissemination are carried out in the framework of the traceRadon project: (a) development of radon activity standards, (b) calibration of low-level atmospheric radon concentration monitors, (c) development of reference infrastructure for radon flux from soil, (d) calibration of continuous radon flux monitors in the field, and (e) 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 realization 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 Meter 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 meter 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.

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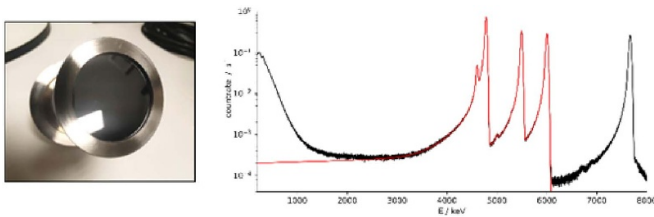
Figure 1. The integrated source-detection system: on the left, an ion-implanted silicon semiconductor detector, coated in a defined manner with $^{226}RaCl_2$ by means of thermal vapor deposition; on the right, the resulting α-spectrum. The respective peaks are well resolved. For the tailing contributions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

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 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 figure 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.

2.2. Realization 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 [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 realization 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 [11–13].

From the radiation protection point of view, the realization 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 meter (m) are defined by fundamental constants and are realized by an NMI. To realize 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.

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 realized 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. All methods provide valid calibration factors [14–16].
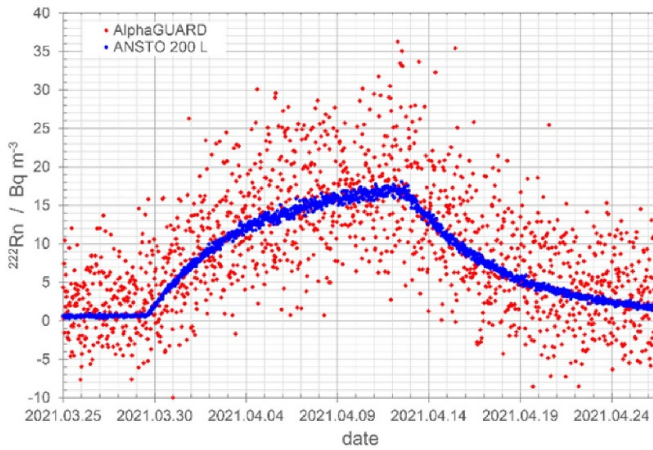
Figure 2. A low-level calibration situation in a non-constant atmosphere: the adjusted response of two types of detectors (AlphaGuard in red, ANSTO 200 l in blue) in a build-up and decay situation in a closed large-volume climate chamber at PTB.
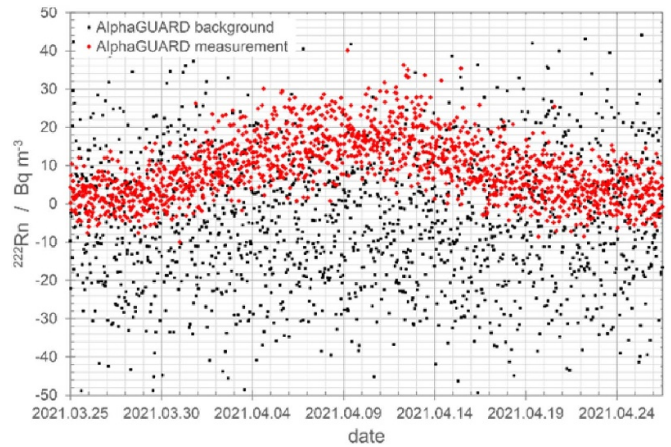

Figure 3. A low-level calibration situation in a non-constant atmosphere: One AlphaGuard in red from figure 2 in a build-up and decay situation in a closed large volume climate chamber at PTB. The activity concentration outside the climate chamber is given by a second AlphaGuard in black.

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, figure 2 summarizes 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.

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, figure 3 shows that the response of the AlphaGuard from figure 2 does not fulfill the requirements to be calibrated in a non-constant atmosphere considering the signal to background reading given in figure 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 [18–20], 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 (a) application of the RTM; (b) validation of radon flux models and inventories [4]; (c) meeting the regulatory requirements for uranium mill tailings or phosphogypsum stacks [21], (d) determining the radon emanation from building materials such as bricks, concrete and other surfaces [22, 23], and (e) 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 recognizable 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 (SCR), (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 instruments 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. Baldoncini *et al* [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 analyze 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 SCR 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 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 SCR $q_{SCR} H_{SCR}$ 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 determination of the instrument response q_{Terr}, q_{SCR}, q_{Art} in the environmental range, is summarized 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 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 realize 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: (a) accurate GHG amount fraction observations, (b) spatially and temporally representative radon flux maps, and (c) 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 standardized, 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 figure 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 recognized value as a tracer of transport and mixing, as yet there is no standardized 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).

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 recognize 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
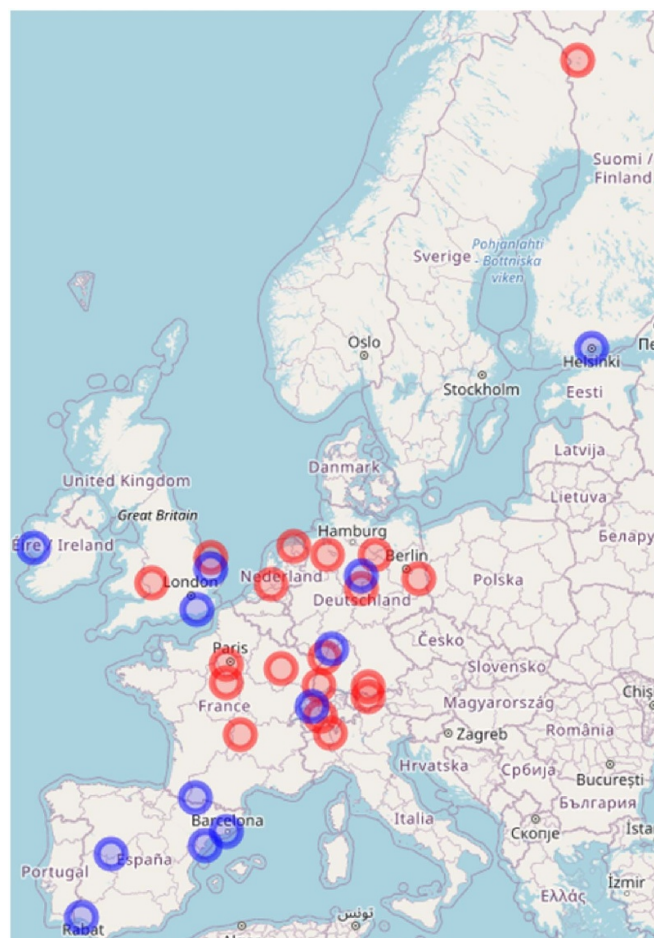

Figure 4. Location of the 31 European sites measuring atmospheric radon measurements. 19 sites are part of ICOS Atmosphere (red circles). The others in blue circles are either part of AGAGE, WMO-GAW or are not affiliated to a GHG network. OpenStreetMap contributors 2020. Distributed under a Creative Commons BY-SA License. Accessed 11 June 2021.

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 modeling 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, modeled 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 characteristics) 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, $T_{1/2} \sim 1620$ years) and soil permeability, radon's flux to the atmosphere can be modeled with smaller uncertainties than GHG fluxes. Furthermore, its only atmospheric sink is radioactive decay ($T_{1/2} \sim 3.82$ days), making ^{222}Rn an approximately conservative tracer on subdiurnal 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 [39–41]. 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 'RTM' [43–47]. 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 figure 4.1. These radon measurements are currently being made by a wide range of different instruments, based on different measurement techniques [48, 49], see figure 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
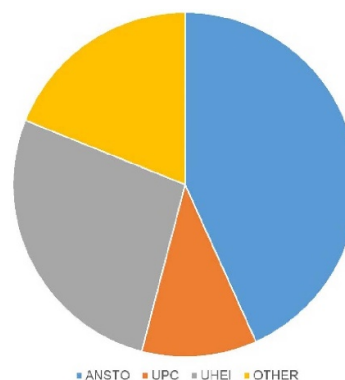

Figure 5. Atmospheric radon research detector types (from ANSTO, UPC, UHEI and other developers) currently used at the European atmospheric monitoring stations for research. In total there are 38 research monitors in use at 30 different sites.

portable atmospheric radon monitors and to develop traceable calibrations for them at very low atmospheric radon activity concentrations (e.g. 1 Bq m^{-3}–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 exceed 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 figure 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 min, 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 is also running at a number of European stations. In addition, several Spanish stations are currently using the direct monitor atmospheric radon monitor (ARMON) [51] which can measure both ^{222}Rn and ^{220}Rn with 30 min temporal resolution, and allows a full alpha spectral analysis of each measurement.

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 min temporal resolution and a sensitivity determined in section 3.1 that, for the first time, is traceable to the new
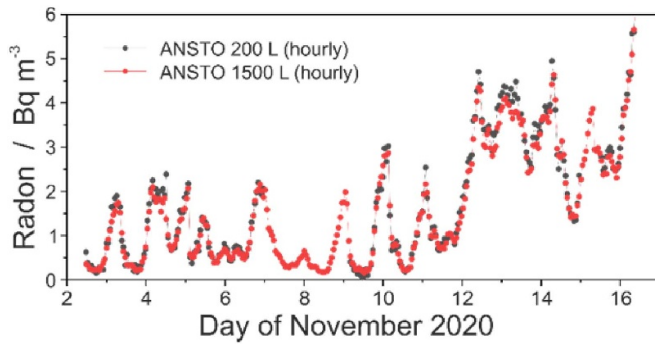
Figure 6. Two-week comparison of hourly atmospheric radon activity concentration observations between a 200 l and 1500 l ANSTO radon detector from 3 m above ground level at a site near Sydney, Australia, 15 km from the coast.
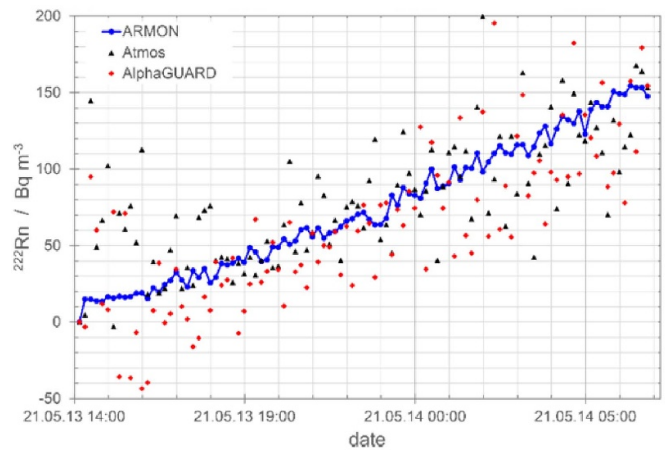

Figure 7. A preliminary low-level comparison between ARMON (in blue) and two commercial monitors (ATMOS and AlphaGUARD) (in black and red) carried out within the INTE-UPC radon chamber during an activity concentration build-up phase. INTE-UPC operates a radon chamber, categorized as a STAR according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

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 parameters for STP and water vapor correction of radon concentrations, and has the capability to perform calibrations/instrumental background checks automatically *in situ*.

As shown in figure 2, full characterization 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 (figure 6), although its response time was faster. At 'baseline' monitoring stations [2, 52], 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.

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' (MARE²EA, 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 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. Figure 8 shows the simulated electric field within the detection volume using the COMSOL Multiphysics® software and the user panel created by LabView software.

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 Modeling 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
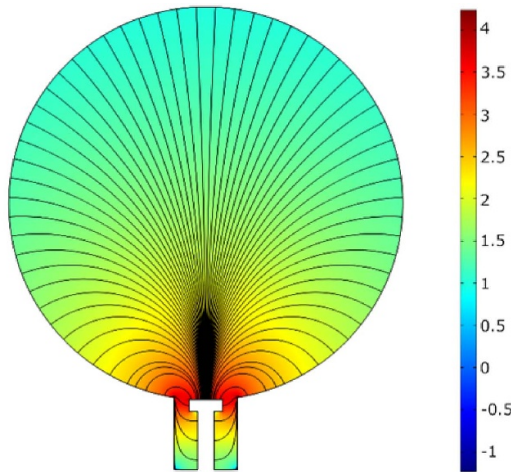
Figure 9. The location of gamma dose rate sensors in some of the European countries in spring 2021. Reproduced from remap.jrc.ec.europa.eu. CC BY 4.0. Accessed 16 August 2021.

European countries. Significant differences in the configuration of national networks are evident.

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 prioritization 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 prioritization 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

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.

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. Figure 9 shows the location of gamma dose rate sensors at the moment of writing this paper in some of the

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 and International Atomic Energy Agency 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 [37, 51]. 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 ionization and the atmospheric electric field [68, 69]. Ion production in the atmosphere is mainly driven by ionizing radiation, and near the Earth's surface radon and its progeny are the major sources of ionizing 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.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

ORCID iDs

Annette Röttger ![ORCID] https://orcid.org/0000-0001-6858-5795
Stefan Röttger ![ORCID] https://orcid.org/0000-0002-4750-9563
Claudia Grossi ![ORCID] https://orcid.org/0000-0001-6678-4440
Arturo Vargas ![ORCID] https://orcid.org/0000-0002-2576-9671
Petr Otáhal ![ORCID] https://orcid.org/0000-0001-7291-001X
Miguel Ángel Hernández-Ceballos ![ORCID] https://orcid.org/0000-0001-7868-0833

Scott Chambers ⓘ https://orcid.org/0000-0002-2521-959X
Susana Alexandra Barbosa ⓘ https://orcid.org/0000-0003-2198-3715
Mihail-Razvan Ioan ⓘ https://orcid.org/0000-0002-2657-8471
Ileana Radulescu ⓘ https://orcid.org/0000-0002-1628-9916
Dafina Kikaj ⓘ https://orcid.org/0000-0001-7801-7396
Edward Chung ⓘ https://orcid.org/0000-0001-7801-7396
Tim Arnold ⓘ https://orcid.org/0000-0001-9097-8907
Camille Yver-Kwok ⓘ https://orcid.org/0000-0002-2181-2863
Marta Fuente ⓘ https://orcid.org/0000-0002-9444-0925
Florian Mertes ⓘ https://orcid.org/0000-0003-0217-2845

References

[1] Council decision 87/600/Euratom of 14 December 1987 on community arrangements for the early exchange of information in the event of a radiological emergency, ELI (Available at: http://data.europa.eu/eli/dec/1987/600/oj) (Accessed 6 April 2021)

[2] Chambers S D *et al* 2016 Towards a universal 'baseline' characterisation of air masses for high- and low-altitude observing stations using Radon-222 *Aerosol Air Qual. Res.* **16** 885–99

[3] Grossi C *et al* 2016 Analysis of ground-based ^{222}Rn measurements over Spain: filling the gap in south-western Europe *J. Geophys. Res. Atmos.* **121** 11021–37

[4] Karstens U, Schwingshackl C, Schmithüsen D and Levin I 2015 A process-based 222radon flux map for Europe and its comparison to long-term observations *Atmos. Chem. Phys.* **15** 12845–65

[5] Kessler P, Camp A, Dombrowski H, Neumaier S, Röttger A and Vargas A 2017 Influence of radon progeny on dose rate measurements studied at PTB's radon reference chamber *Radiat. Prot. Dosim.* **177** 407–14

[6] Picolo J L 1996 Absolute measurement of radon 222 activity *Nucl. Instrum. Methods Phys. Res.* A **369** 452–7

[7] Linzmaier D and Röttger A 2014 Development of a transfer standard for the measurement of low Rn-222 activity concentration in air *Appl. Radiat. Isot.* **87** 306–9

[8] Mertes F, Röttger S and Röttger A 2020 A new primary emanation standard for Radon-222 *Appl. Radiat. Isot.* **156** 108928

[9] Annals of the ICRP 2007 The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103 *Ann. ICRP* **37** 2–4

[10] ISO 4037-1 2019 Radiological protection—x and gamma reference radiation for calibrating dosemeters and doserate meters and for determining their response as a function of photon energy—part 1: radiation characteristics and production methods (Available at: www.iso.org/standard/66872.html)

[11] Dombrowski H, Cornejo Díaz N A, Toni M P, Mihelic M and Röttger A 2018 Comparison of the uncertainties of several European low-dose calibration facilities *J. Instrum.* **13** P04023

[12] Kessler P, Behnke B, Dabrowski R, Dombrowski H, Röttger A and Neumaier S 2018 Novel spectrometers for environmental dose rate monitoring *J. Environ. Radioact.* **187** 115–21

[13] Röttger A and Kessler P 2019 Uncertainties and characteristic limits of counting and spectrometric dosimetry systems *J. Environ. Radioact.* **205–206** 48–54

[14] Röttger A and Honig A 2011 Recent developments in radon metrology: new aspects in the calibration of radon, thoron and progeny devices *Radiat. Prot. Dosim.* **145** 260–6

[15] Linzmaier D and Röttger A 2013 Development of a low-level radon reference atmosphere *Appl. Radiat. Isot.* **81** 208–11

[16] Röttger A, Honig A and Linzmaier D 2014 Calibration of comercial radon and thoron monitors at stable activity concentrations *Appl. Radiat. Isot.* **87** 44–47

[17] Chambers S D, Morosh V, Griffiths A D, Williams A G, Röttger S and Röttger A 2021 Field testing a portable two-filter dual-flow-loop 222Rn detector. Virtual poster and vPICO presentation *EGU General Assembly 2021* (*Vienna Austria*, 19–30 April 2021) Session GI6.2, abstract ID pp EGU21–196

[18] Keller G, Folkerts K H and Muth H 1982 Method for the determination of ^{222}Rn (radon) and ^{220}Rn (Thoron)—exhalation rates using alpha spectrometry *Radiat. Prot. Dosim.* **3** 83–89

[19] Keller G and Schütz M 1988 Radon exhalation from the soil *Radiat. Prot. Dosim.* **24** 43–46

[20] Grossi C, Vargas A, Camacho A, López-Coto I, Bolívar J P, Xia Y and Conen F 2011 Inter-comparison of different direct and indirect methods to determine radon flux from soil *Radiat. Meas.* **46** 112–8

[21] Dueñas C, Liger E, Cañete S, Pérez M and Bolívar J P 2007 Exhalation of 222Rn from phosphogypsum piles located at southwest of Spain *J. Environ. Radioact.* **95** 63–74

[22] Stieff L R, Kotrappa P and Bigu J 1996 Passive Eperm radon flux monitors for measuring undisturbed radon flux from the ground *1996 Int. Radon Symp., American Association of Radon Scientists and Technologists*

[23] Andersen C E 1999 Radon-222 exhalation from Danish building materials: h + H Industri A/S results Risø National Laboratory, Denmark. Forskningscenter Risoe. Risoe-R No. 1135(EN)

[24] Nazaroff W and Nero A V 1988 *Radon and Its Decay Products in Indoor Air* (New York: Wiley)

[25] Porstendörfer J 1994 Proprieties and behaviour of radon and thoron and their decay products in air *J. Aerosol. Sci.* **25** 219–63

[26] McLaughlin T 2011 Technical bases and guidance for radon flux monitoring at uranium mill tailing sites DOE CONTRACT NO. DE-AC05-06OR23100, (RFTA 11-010) DCN 2042-TR-01-0

[27] Schery S D, Whittlestone S, Hart K P and Hill S E 1989 The flux of radon and thoron from Australian soils *J. Geophys. Res.* **94** 8567–76

[28] Szegvary T, Conen F and Ciais P 2009 European ^{222}Rn inventory for applied atmospheric studies *Atmos. Environ.* **43** 1536–9

[29] Szegvary T, Leuenberger M and Conen F 2007 Predicting terrestrial ^{222}Rn flux using gamma dose rate as a proxy *Atmos. Chem. Phys.* **7** 2789–95

[30] Manohar S, Meijer H and Herber M 2013 Radon flux maps for the Netherlands and Europe using terrestrial gamma radiation derived from soil radionuclides *Atmos. Environ.* **81** 399–412

[31] UNSCEAR 2000 United Nations: sources and effects of ionizing radiation. United Nations sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR 2000 report to the general assembly with scientific annexes (Available at: www.unscear.org/unscear/en/publications/2000_1.html)

[32] Baldoncini M, Albéri M, Bottardi C, Chiarelli E, Raptisa K G C, Strati V and Mantovani F 2018 Investigating the potentialities of Monte Carlo simulation for assessing soil water content via proximal gamma-ray spectroscopy *J. Environ. Radioact.* **192** 105–16

[33] Dabrowski R, Dombrowski H, Kessler P, Röttger A and Neumaier S 2017 Detection of rain events in radiological early warning networks with spectrometric systems *J. Instrum.* **12** P10005

[34] Melintescu A, Chambers S D, Crawford J, Williams A G, Zorila B and Galeriu D 2018 Radon-222 related influence on ambient gamma dose *J. Environ. Radioact.* **189** 67–78

[35] Tudor D *et al* 2020 A facility for direct measurements for nuclear astrophysics at—FIN-HH—a 3 MV tandem accelerator and a ultra-low background laboratory *Nucl. Instrum. Methods Phys. Res.* A **953** 163178

[36] ICOS *Integrated Carbon Observation System – ICOS* (Available at: www.icos-cp.eu/) (Accessed 20 September 2020)

[37] Arnold T, Manning A, Kim J, Li S, Webster H, Thomson D, Mühle J, Weiss R F, Park S and O'Doherty S 2018 Inverse modelling of CF4 and NF3 emissions in East Asia *Atmos. Chem. Phys.* **18** 13305–20

[38] Chambers S D *et al* 2018 Characterizing atmospheric transport pathways to Antarctica and the remote Southern Ocean using radon-222 *Front. Earth Sci.* **6** 190

[39] Williams A G, Chambers S D, Conen F, Reimann S, Hill M, Griffiths A D and Crawford J 2016 Radon as a tracer of atmospheric influences on traffic-related air pollution in a small inland city *Tellus* B **68** 30967

[40] Chambers S D *et al* 2019 Skill-testing chemical transport models across contrasting atmospheric mixing states using Radon-222 *Atmosphere* **10** 25

[41] Kikaj D, Chambers S D, Kobald M, Crawford J and Vaupotič J 2020 Characterizing atmospheric controls on winter urban pollution in a topographic basin setting using Radon-222 *Atmos. Res.* **237** 104838

[42] Levin I 1987 Atmospheric CO_2 in continental Europa—an alternative approach to clean air CO_2 data *Tellus* B **39** 21–28

[43] Hirsch A I 2007 On using radon-222 and CO_2 to calculate regional-scale CO_2 fluxes *Atmos. Chem. Phys.* **7** 3737–47

[44] Levin I, Hammer S, Eichelmann E and Vogel F R 2011 Verification of greenhouse gas emission reductions: the prospect of atmospheric monitoring in the polluted areas *Phil. Trans. R. Soc.* A **369** 1906–24

[45] Lopez M, Schmidt M, Yver C, Messager C, Worthy D, Kazan V, Ramonet M, Bousquet P and Ciais P 2012 Seasonal variation of N_2O emissions over three sites in Northern France inferred from 10 years of continuous atmospheric N_2O and ^{222}Rn measurements *J. Geophys. Res.* **117** D14

[46] Wada A, Matsueda H, Murayama S, Taguchi S, Hirao S, Yamazawa H, Moriizumi J, Tsuboi K, Niwa Y and Sawa Y 2013 Quantification of emission estimates of CO_2, CH_4 and CO for East Asia derived from atmospheric radon-222 measurements over the western North Pacific *Tellus* B **65** 18037

[47] Grossi C, Vogel F R, Curcoll R, Àgueda A, Vargas A, Rodó X and Morguí J-A 2018 Study of the daily and seasonal atmospheric CH4 mixing ratio variability in a rural Spanish region using ^{222}Rn tracer *Atmos. Chem. Phys.* **18** 5847–60

[48] Schmithüsen D *et al* 2017 A European-wide ^{222}Rn and ^{222}Rn progeny comparison study *Atmos. Meas. Tech.* **10** 1299–312

[49] Grossi C *et al* 2020 Intercomparison study of atmospheric ^{222}Rn and ^{222}Rn progeny monitors *Atmos. Meas. Tech.* **13** 2241–55

[50] Griffiths A D, Chambers S D, Williams A G and Werczynski S R 2016 Increasing the accuracy and temporal resolution of two-filter radon–222 measurements by correcting for the instrument response *Atmos. Meas. Tech.* **9** 2689–707

[51] Grossi C, Arnold D, Adame J A, López-Coto I, Bolívar J P, De La Morena B A and Vargas A 2012 Atmospheric ^{222}Rn concentration and source term at El Arenosillo 100 m meteorological tower in southwest Spain *Radiat. Meas.* **47** 149–62

[52] Chambers S D, Zahorowski W, Williams A G, Crawford J and Griffiths A D 2013 Identifying tropospheric baseline air masses at Mauna Loa Observatory between 2004 and 2010 using Radon-222 and back trajectories *J. Geophys. Res. Atmos.* **118** 992–1004

[53] Curcoll Masanes R, Grossi C and Vargas A 2021 High efficiency and portable monitor of atmospheric radon concentration activity for environmental applications *EGU General Assembly 2021* (19–30 April 2021) pp EGU21–4497

[54] Vargas A, Ortega X and Martín Matarranz J L 2004 Traceability of radon-222 activity concentration in the radon chamber at the technical university of Catalonia (Spain) *Nucl. Instrum. Methods Phys. Res.* A **526** 501–9

[55] IEC 61577-1:2006 Radiation protection instrumentation—Radon and radon decay product measuring instruments—part 1: general principles (Available at: https://webstore.iec.ch/publication/5616)

[56] Sangiorgi M, Hernández-Ceballos M A, Jackson K, Cinelli G, Bogucarskis K, De Felice L, Patrascu A and De Cort M 2020 The European radiological data exchange platform (EURDEP): 25 years of monitoring data exchange *Earth Syst. Sci. Data* **12** 109–18

[57] EC 2014 European Council: council Directive 2013/59/Euratom of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation *Off. J. Eur. Union* **57** 1–73 (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32013L0059&qid=1617719433604)

[58] EC 2020 Radiation protection n.193, Radon in workplaces, implementing the requirements in Council Directive 2013/59/Euratom (https://doi.org/10.2833/552398)

[59] Bossew P 2018 Radon priorit—areas—definition, estimation and uncertainty *Nucl. Technol. Radiat. Prot.* **33** 286–92

[60] Bossew P, Cinelli G, Ciotoli G, Crowley Q G, De Cort M, Elío Medina J, Gruber V, Petermann E and Tollefsen T 2020 Development of a geogenic radon hazard index—concept, history, experiences *Int. J. Environ. Res. Public Health* **17** 4134

[61] World Health Organization (WHO) 2021 *More countries act against exposure to radon and associated cancer risks* (available at: https://www.who.int/news/item/04-02-2021-more-countries-act-against-exposure-to-radon-and-associated-cancer-risks) (Accessed 4 February 2021)

[62] Zhang B *et al* 2021 Simulation of radon-222 with the GEOS-Chem global model: emissions, seasonality, and convective transport *Atmos. Chem. Phys.* **21** 1861–87

[63] Bogena H R, Huisman J A, Güntner A, Hübner C, Kusche J, Jonard F, Vey S and Vereecken H 2015 Emerging methods for noninvasive sensing of soil moisture dynamics from field to catchment scale: a review *Wiley Interdiscip. Rev. Water* **2** 635–47

[64] Barbosa S, Huisman J A and Azevedo E B 2018 Meteorological and soil surface effects in gamma radiation time series-Implications for assessment of earthquake precursors *J. Environ. Radioact.* **195** 72–78

[65] Barbosa S M, Miranda P and Azevedo E B 2017 Short-term variability of gamma radiation at the ARM Eastern North Atlantic facility (Azores) *J. Environ. Radioact.* **172** 218–31

[66] Cigolini C *et al* 2009 Radon surveys and real-time monitoring at Stromboli volcano: influence of soil temperature,

atmospheric pressure and tidal forces on 222Rn degassing *J. Volcanol. Geotherm. Res.* **184** 381–8

[67] Girault F, Koirala B P, Bhattarai M and Perrier F 2018 Radon and carbon dioxide around remote Himalayan thermal springs *Geol. Soc. Spec. Publ.* **451** 155–81

[68] Barbosa S 2020 Ambient radioactivity and atmospheric electric field: a joint study in an urban environment *J. Environ. Radioact.* **19** 106283

[69] Balkanski Y J and Jacob D J 1990 Transport of continental air to the Subantarctic Indian Ocean *Tellus* B **42** 62–75