



# NEWSLETTER

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## EMPIR 19ENV01 traceRadon: An introduction

Radon gas is the largest source of public exposure to naturally occurring radioactivity, and concentration maps based on atmospheric measurements aid developers to comply with EU Basic Safety Standard Regulations (EU-BSS). Radon can also be used as a tracer to evaluate dispersal models important for identifying successful greenhouse gas (GHG) mitigation strategies.

To increase the accuracy of both radiation protection measurements and those used for GHG modelling, traceability to SI units for radon release rates from soil, its concentration in the atmosphere and validated models for its dispersal are needed. This project will provide the necessary measurement infrastructure and use the data that this generates to apply the Radon Tracer Method (RTM) which is important for GHG emission estimates that support national reporting under the Paris Agreement on climate change. An overlapping need exists between the climate research and radiation protection communities for improved traceable low-level outdoor radon measurements, combining the challenges of collating and modelling large datasets, with setting up new radiation protection services.

Compared to the large spatiotemporal heterogeneity of GHG fluxes, radon is emitted almost homogeneously over ice-free land and has a negligible flux from oceans. Radon flux relates to the transfer process of radon activity from soil to the atmosphere per square metre and second, whilst radon activity concentration is the amount of activity of radon in the atmosphere per cubic metre. Atmospheric measurements of radon activity concentrations can be used for the assessment and improvement of atmospheric transport models (ATM). However, traceability to the environmental level does not currently exist for measurements of radon fluxes and atmospheric radon activity concentrations. Therefore, significant improvements in such measurements are needed. Climatic Atmospheric Monitoring Networks (AMN) like the European Integrated Carbon Observation System (ICOS), are infrastructures that operate GHG monitoring stations and include atmospheric radon monitors in their stations. The radon data produced from such networks can be used to improve transport modelling and the estimation of GHG emissions based on the RTM, which uses the correlation between GHG and radon concentrations. However, this radon data needs significant improvement in terms of the accuracy of both radon flux measurements and environmental radon activity concentrations in the range 1 Bq m<sup>-3</sup> to 100 Bq m<sup>-3</sup> to be able to provide robust data for use in the RTM. Similarly, for radiation monitoring, all European countries have installed networks of automatic radiation dose and airborne contamination monitoring stations and report the information gathered to the European Radiological Data Exchange Platform (EURDEP), thus supporting EU member states and the EURATOM treaty.

Currently, monitoring information on dose rates is collected from automatic surveillance systems in 39 countries, however, urgently needed data on outdoor radon activity concentrations is not yet collected due to a lack of ability to measure accurately at the low levels encountered in the environment. Furthermore, accurately detecting contamination from nuclear emergencies relies on rejecting false positive results based on radon washed from the atmosphere by rain. Therefore, improving contamination detection requires greater accuracy in determining environmental radon concentrations and their movement in the atmosphere.

At this point in time 18 partners and 12 collaborators are working toward this goal!



## News from the work packages and recent developments

### WP 1: Outdoor radon activity concentrations

The new version of the ARMON (Atmospheric Radon MONitor) has been mounted and characterized at the radon chamber of the Universitat Politècnica de Catalunya (UPC) during the past months. This monitor, based on the alpha spectrometry of positive Po-218 ions collected on a PIPS detector surface, has been designed for measuring low ambient Rn-222 activity concentration ( $<100 \text{ Bq}\cdot\text{m}^{-3}$ ). The ARMON has been sent to the Physikalisch-Technische Bundesanstalt (PTB) at the beginning of November 2021 to be calibrated and for participation in a 1-month intercomparison campaign with different atmospheric radon monitors. Fig. 1.1 show the ARMON in the transport case.



Figure 1.1: inside (left) and back (right) of the new ARMON monitor. In the inner trays, from top to bottom: i) laptop; ii) datalogger and multichannel analyser; iii) sensors and voltage transformers iv) amplifier v) detection volume vi) high voltage power supply.

At SUJCHBO, with the help of the CMI, the procedure for the graded approach field calibration was developed. Field calibration is used for instruments operated in flow measuring mode (not diffusion mode). Experiments in the SUJCHBO laboratory were performed with the AlphaGuard DF 2000. An example of one such field calibration is shown in Fig. 1.2.

Measurement of radon in outdoor air "correctly" requires metrological traceability of the activity concentration of radon in outdoor air from about  $1 \text{ Bq}\cdot\text{m}^{-3}$  to  $10 \text{ Bq}\cdot\text{m}^{-3}$ .

Because of the small number of events, this poses a great challenge to the transfer.

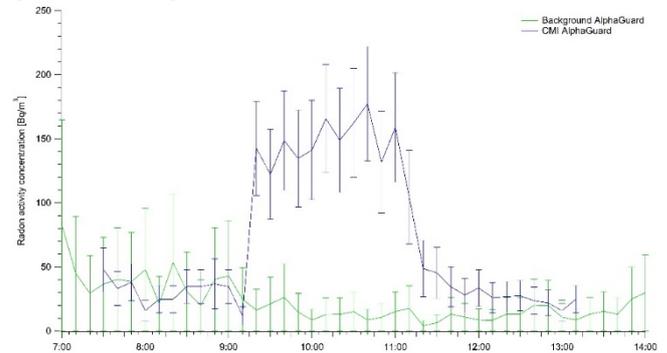


Figure 1.2: Measurement for a field calibration of an AlphaGuard DF2000 performed at SUJCHBO. The CMI source produces  $119(6) \text{ Bq}\cdot\text{m}^{-3}$ . The evaluated activity concentration is  $120(28) \text{ Bq}\cdot\text{m}^{-3}$ .

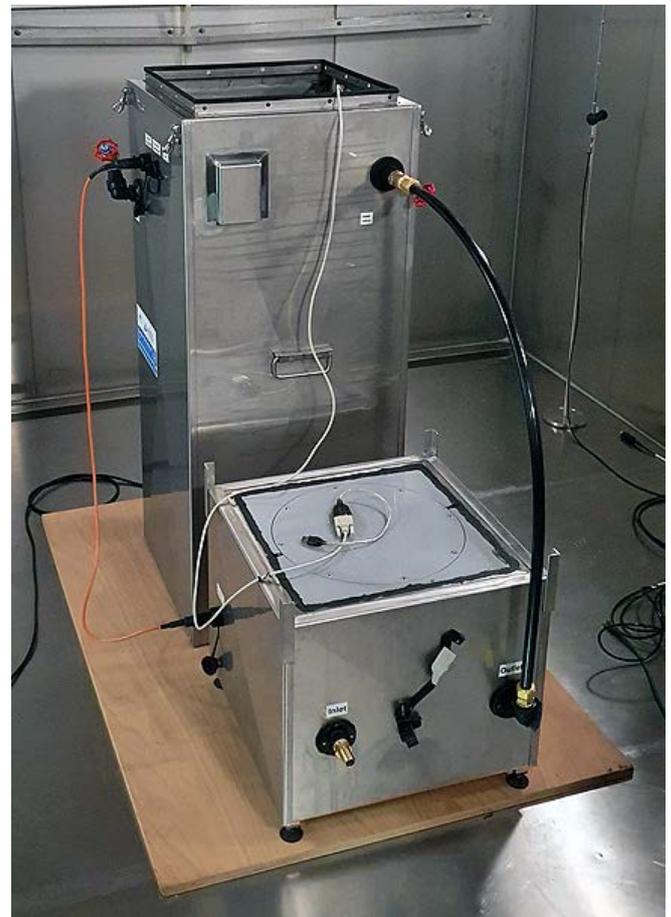


Figure 1.3: 200 L radon monitor prototype from ANSTO in PTBs' climate chamber.

This becomes obvious with the following example calculation: Assume that a radon measuring device has an active volume of 1 L. This results in counting rates of  $36 \text{ h}^{-1}$  at  $10 \text{ Bq}\cdot\text{m}^{-3}$ , which statistically results in an uncertainty (with Poisson statistics) of 17 % during a measuring time of one hour. Additionally, one has to

take into account the uncertainty of the instruments intrinsic zero effect, linearity, and calibration for such a system [Röttger 2013, Röttger 2011]. The overall measurement uncertainty, as well as the decision threshold and detection limit of such a system are not very suitable for detecting dynamic processes in the range of the activity concentration of outdoor air, even under ideal calibration conditions. One solution to the statistical problem is to increase the active volume or to measure the radon progeny with increased flow. This way, the count rates can be greatly increased. Examples of such detectors include the ANSTO monitors (active volume between 200 L and 1500 L), the Atmospheric Radon Monitor "ARMON" from UPC, and the Heidelberg Radon Monitor "HRM".

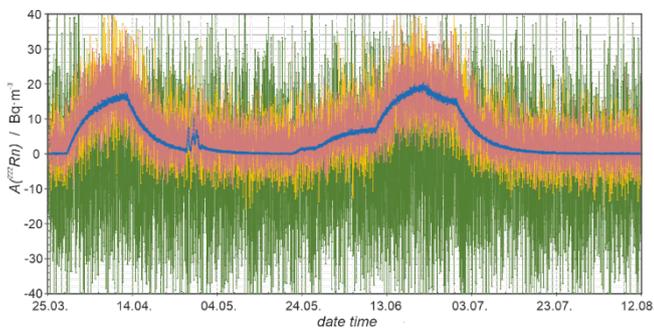


Figure 1.4: Response of the new prototype (blue) compared to radon monitors used in radiation protection measurements (red, yellow). To give an impression of the statistical problems of low volume devices according to background reading, the green curve shows an identical device to the red and yellow data. All data points are 30 min average interval.

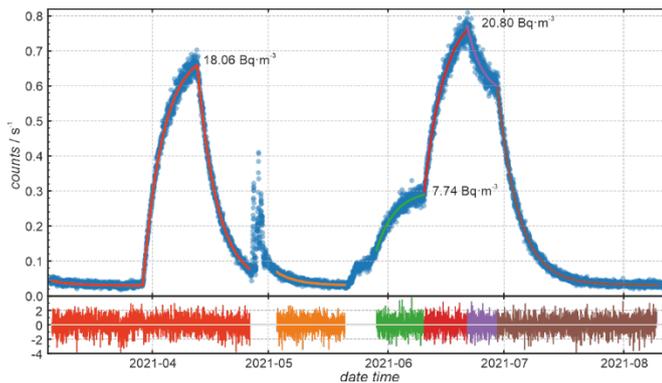


Figure 1.5: Shows the response in counts per second of the device (30 min averaging interval) according to the established activity concentration level.

Meanwhile an intensive calibration campaign in the PTB climate chamber produced some initial results regarding a possible calibration route for the 200 L radon monitor, the new prototype from ANSTO. The set-up inside the climate chamber is visible in Fig. 1.3. The preliminary results were published in [Röttger 2021] while the proof of principal is impressively shown in Fig. 1.4 and Fig. 1.5.

## WP 2: Radon flux measurements

During May and September 2021 several experiments were carried out at the University of Cantabria (UC) with the aim for testing the reliability of an exhalation bed, with a characterized radon flux, to calibrate radon flux systems under different environmental conditions. In Fig. 2.1 a typical experimental configuration is shown. Different radon flux systems, designed and built by different traceRadon project partners and collaborators, were simultaneously tested over the bed surface under dynamic or static conditions.



Figure 2.1: Typical configuration experiment with different radon flux systems.

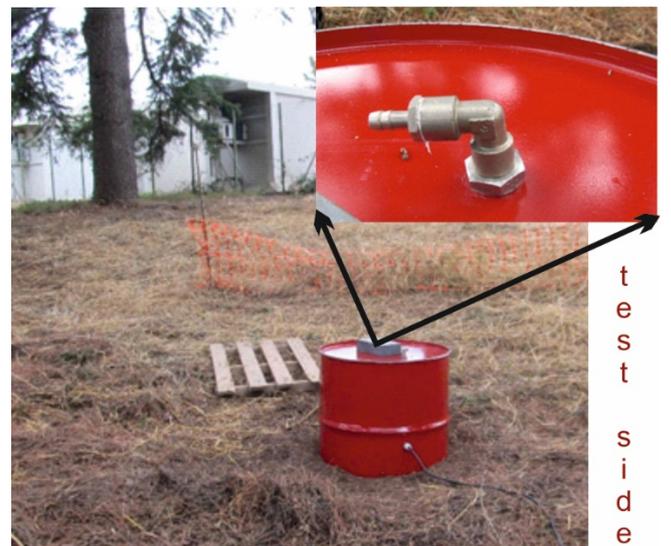


Figure 2.2: Measurement in field with ENEA's new radon flux system performed at the ENEA site during October 2021

The traceRadon partner ENEA designed and developed a new radon flux system (Fig. 2.2) which consists of an accumulation chamber with a vent port and continuous radon monitor inside. Laboratory test carried out at

ENEA shows that the leakage rate from the vent port is in the order of  $0.01 \text{ h}^{-1}$ . Field measurements were also performed at the ENEA site during October 2021.



Figure 2.3: Radon flux Intercomparison campaign at Esles de Cayón (Cantabria, Spain) between the 13<sup>th</sup> and the 28<sup>th</sup> of October 2021.

During the months of September and October 2021, UC with the support of WP2 partners, organized and performed two intercomparison campaigns of radon flux systems within the context of Task 2.2. The campaigns were conducted in high and low radon source areas with typical radon flux values of about  $2000 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  and  $50 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ , respectively. The high radon level intercomparison campaign was carried out at a Spanish uranium mine located in Saelices el Chico (Salamanca, Spain) from 6<sup>th</sup> to 8<sup>th</sup> of October 2021 and the low level at Esles de Cayón (Cantabria, Spain) between 13<sup>th</sup> and 28<sup>th</sup> of October 2021. Fig. 2.3 presents the experimental setup during the intercomparison campaign at Esles de Cayón.

### WP 3: Radon flux models and inventories

The radon flux from the soil to the atmosphere depends on the uranium concentration in the soil material, the basic soil texture (sand, silt and clay content), and the air-filled pore space in which the radon can move and diffuse into the atmosphere (see Fig. 3.1).

The air-filled pore space depends on the texture but also very much on the water content in the soil, which, depending on the frequency of precipitation, takes up part of the free pore space and effectively limits the transport of radon into the atmosphere. Previous observations have shown that the radon flux from dry soils can be more than a factor of two larger than under wet conditions. Parameterizations of these processes are implemented in the radon flux model by Karstens et al. (2015) and will be directly evaluated with data from the intensive measurement campaigns in WP2.

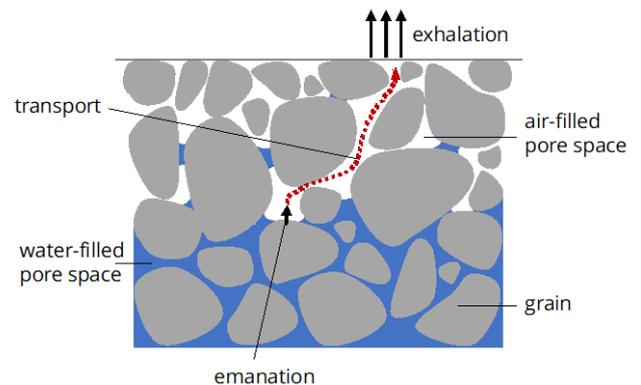


Figure 3.1: Radon transport processes in the soil.

The radon flux model is applied to calculate process-based European radon flux maps using existing soil parameter maps as well as spatially and temporally resolved soil moisture information. The radon source is parameterized using the uranium content of the soil, available in the European Atlas of Natural Radiation [Cellini et al., 2020] and an estimate of the likelihood that a radon atom can escape from soil grains (emanation rate) based on the texture of the soil. Both datasets are available for the EU region and have to be complemented with additional information from other data sets to cover all land surfaces in geographical Europe, which is a prerequisite for using the final radon flux map in atmospheric transport model studies. Spatially resolved soil moisture information is available from global soil moisture reanalysis in which soil moisture is simulated by state-of-the-art land surface models driven by numerical weather prediction models [e.g. ERA5-Land, Muñoz-Sabater et al., 2021; GLDAS-Noah, Rodell et al., 2004]. A recent evaluation study has shown that these reanalyses realistically represent the temporal variability of the soil moisture but also that they often show substantial biases of the absolute value [Li et al., 2020]. These biases in the modelled soil moisture will directly translate into biases in the calculated radon flux. Therefore, methods to scale the soil moisture and/or the resulting radon flux based on comparisons with representative measurements need to be developed. This requires careful screening of available soil moisture data for spatial representativeness and measurement accuracy.

Terrestrial ambient gamma radiation data provides additional information for the evaluation of radon flux models and maps. This requires that dose rate monitors and spectrometers are well characterized with respect to their inherent background and their sensitivity to small variations of ambient dose equivalent rate.

A spectrometric system has been developed at UPC for continuously monitoring of the environmental gamma radiation and to be used in the intensive campaigns at selected sites in the framework of the traceRadon project. The spectrometric detector system is named

DoRayMon. The system can connect to 3G/4G and automatically send the measured spectra to a database. Fig. 3.2 shows a photo of DoRayMon.



Figure 3.2: A photograph of DoRayMon.

Table 3.1:  $H^*(10)$  rates measured with DoRayMon compared with the reference values provided by PTB.

| Source                     | $H^*(10)$ [nSv/h] |                  |
|----------------------------|-------------------|------------------|
|                            | DoRayMon values   | Reference values |
| $^{60}\text{Co}$ (Dose 1)  | $34.8 \pm 0.7$    | $35.3 \pm 1.4$   |
| $^{60}\text{Co}$ (Dose 2)  | $52.1 \pm 0.6$    | $52.6 \pm 1.6$   |
| $^{137}\text{Cs}$ (Dose 1) | $11.8 \pm 0.4$    | $12.4 \pm 0.9$   |
| $^{137}\text{Cs}$ (Dose 2) | $24.1 \pm 0.5$    | $24.5 \pm 1.1$   |
| $^{137}\text{Cs}$ (Dose 3) | $50.9 \pm 0.5$    | $50.0 \pm 1.2$   |
| $^{226}\text{Ra}$          | $38.6 \pm 0.5$    | $42.0 \pm 1.6$   |

The inherent background of the detector, its response to cosmic radiation and the sensitivity to small variations of the ambient dose equivalent rates,  $H^*(10)$ , were studied by carrying out irradiations at PTBs facilities in June 2021. The DoRayMon net  $H^*(10)$  rates compared to the reference values are in good agreement as it is shown in Table 3.1.

#### WP 4: Radon and radon flux in maps

Article 103 of Basic Safety Standards Directive (Directive 2013/59/EUROATOM) requires Member States to identify areas where the radon concentration (as an annual average) in a significant number of buildings is expected to exceed the relevant national reference level, called radon priority areas (RPA). The designation of the RPA is usually done based on the indoor radon measurements or based on the geogenic radon potential. Within WP4, the traceRadon project aims to go beyond the state-of-the-art by developing improved methods for identification of RPA by including outdoor radon concentrations and radon flux measurements. For this purpose, a literature review of outdoor radon concentrations focusing on the aspect of the radiation health risk have been carried out.

Although radon in outdoor environment does not represent significant health risk to the general population, knowing it is of interest for radiation protection issues as: 1) it can serve as a baseline to assess anthropogenic contribution, 2) there are regions that have much higher annual outdoor radon concentrations than the worldwide average, 3) neglecting to correct indoor radon concentrations in surveys for outdoor radon concentrations could lead to improper estimation on percentage of dwellings exceeding a reference level, which as a consequence could lead to a misclassification of the RPA, and 4) correct assessment of radon exposure is important for epidemiological studies to detect an effect due to radon. Measurement of outdoor radon concentrations represent a challenge since outdoor radon concentrations could be a few orders of magnitude smaller than indoor radon concentrations and detectors are exposed to harsh environmental conditions. According to reviewed literature: around 60 % of integrating, 30 % of continuous and 10 % of instantaneous measurements were performed for outdoor radon measurements. Detectors were placed at different height, but mainly between 1 m and 3 m. In certain cases, special treatments of track detectors were applied in order to increase sensitivity and reduce background of detectors. Measurement duration was in accordance with type of measurements, covering from 10 minutes of grab sampling, over a few weeks, months and a whole year and up to a few years of continuous measurements: aiming to study diurnal and seasonal radon variations, influence of different parameters (meteorological, geographical,...)

According to the reviewed literature, only 5 national outdoor radon surveys were performed in Japan, Germany, Spain, Slovenia and Ireland. Outdoor radon surveys were performed at a much smaller scale compared to indoor radon ones. Correlation between outdoor radon concentration and some of the "radon quantities" such as indoor radon concentrations, exhalation rate, radon in soil gas, gamma dose rate and  $^{226}\text{Ra}$  in soil, were investigated in only a few studies.

According to the overview of literature regarding outdoor radon measurement campaign, harmonization of used measurement methods is needed, and international recommendations and measurements protocols should be set up. This could be quite challenging since outdoor radon concentrations are used in more applications than indoor radon concentrations. The literature overview imposes that outdoor radon surveys for the purpose of radiation protection issues should be performed more systematically and at a larger scale. The full results have been reported in a paper submitted to the International Journal of Environmental Research and Public Health.

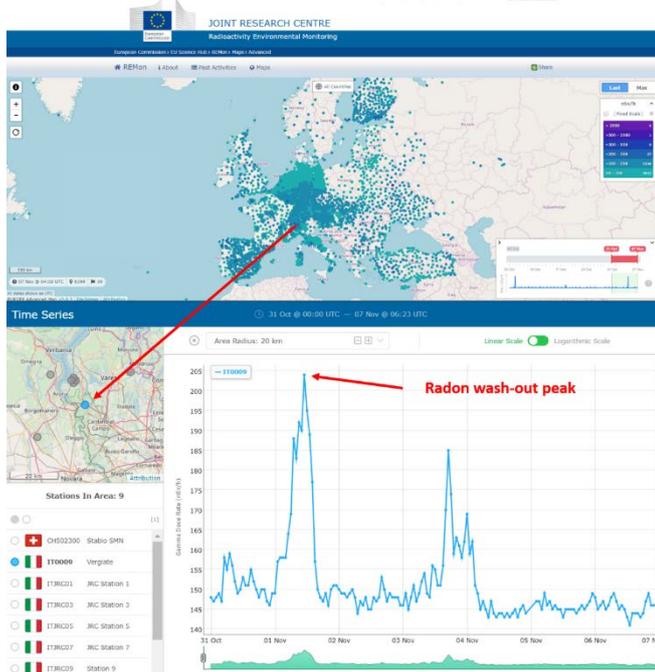


Figure 4.1: Example of radon wash-out peak in gamma dose rate measurements collected and displayed in the EURDEP system (<https://remap.jrc.ec.europa.eu/> - advanced map).

Peaks in gamma radiation are often associated with precipitation events and consequent scavenging of airborne radionuclides to the ground, mainly radon progeny Pb-214 and Bi-214. The estimation of peaks in gamma radiation time series driven by precipitation is both of scientific and practical interest. From the scientific point of view gamma radiation measured at the earth surface provides information on micro-physical processes occurring high above in the clouds, as the dominant source of radon progeny is in-cloud processes – nucleation scavenging (growth of a cloud condensation nucleus including the radionuclide in a cloud droplet) and interstitial aerosol collection by cloud or rain droplets. From the practical aspect, peaks in gamma radiation due to precipitation cause false alarms in early-warning systems (i.e. the European Radiological Data Exchange platform,

<https://remon.jrc.ec.europa.eu/>) and portals for detection of radioactive material. The automatic detection of gamma radiation peaks associated with precipitation is therefore highly relevant, and purely data-driven approaches are of particular interest, as high-resolution precipitation data co-located with gamma stations is not always available, particularly in nearly real time, see Fig. 4.1.

Thus, INESC TEC is working on the application of AI approaches for the identification of peaks associated with precipitation in gamma radiation time series. Two different types of methods are being considered: machine learning (ML) methods, and time series computational methods, namely based on the matrix profile (MP). Diverse methods are being examined as a single one-size-fits-all approach is often sub-optimal, since the performance is dependent on the structure of the gamma time series being examined and specific constrains in terms of length and type of available data. For the ML methods collocated gamma and precipitation data are being considered, while for the matrix profile approach only gamma radiation time series are used. In the ML methods the algorithms are trained to predict precipitation from the gamma radiation time series, based on simultaneous gamma and precipitation values in a training set. The rationale is that once the training is optimized, the prediction enables the identification of the gamma observations associated with precipitation, based only on gamma radiation observations, with no need of precipitation data. In the case of matrix profile algorithms, the matrix profile is used for the identification of time series motifs (typical structures in the time series), based on the fact that gamma peaks associated with precipitation have a typical shape of faster increase and final decay half-life of about 30 min.

Both ML and MP methods are currently being explored to assess their applicability and feasibility in the identification of precipitation-driven anomalies in gamma time series.

In parallel, JRC is working on a method for estimating radon wash-out peaks based on a detailed meteorological characterization of such peaks. To this purpose, EURDEP stations where in addition to gamma dose rate also outdoor radon and meteorological data, such a precipitation, wind speed, humidity, pressure and temperature, are available, have been taken as reference to perform a feasibility study and to test the methodology. These analysis and results will help to better understand radon wash-out peaks and to try to prevent false alarms in the EURDEP system due to radon wash-out effects.



## Past Events

Direct input to standard developing organizations (ISO and IEC) was generated as well as to working groups related to regulatory issues. Recent research outputs and new developments in the project are disseminated to the scientific communities via conferences, publications and workshops.

### GARRM 2021

In the scope of 15<sup>th</sup> INTERNATIONAL WORKSHOP GARRM (on the GEOLOGICAL ASPECTS OF RADON RISK MAPPING) the development of the exhalation bed a facility to calibrate radon flux devices was presented on 23<sup>rd</sup> September 2021.

### CIM 2021

The International Metrology Congress is a Unique event in Europe a showcase for industrial applications, advances in R&D and prospects dedicated to measurements, analysis and testing processes. The project traceRadon was presented in the Green Deal session on 8<sup>th</sup> September 2021.

### LIFE-Respire 2021

Under the RESPIRE - Radon rEal time monitoring System and Proactive Indoor Remediation in July 2021, the European Atlas of Natural Radiation: Indoor Radon Concentration and Geogenic Radon was presented and the link to the data from traceRadon was drawn.

### SMSI 2021

The Sensor and Measurement Science International in 2021 was held as a digital Conference. Here the new online data analysis for approximate sequential Bayesian filtering to estimate Rn-222 emanation from Ra-226 sources from spectra was presented under the session for measurement foundations on 4<sup>th</sup> May 2021.

### EGU 2021

EGU General Assembly 2021 brings together geoscientists from all over the world to one meeting covering all disciplines of the Earth, planetary, and space sciences. In the session of Geosciences Instrumentation & Data Systems the topic Geoscience applications of environmental radioactivity was addressed by a highlighted presentation on radon metrology for use in climate change observation and radiation protection at the environmental level in April 2021.

### EURAMET TC-IR 2021

The Technical Committee for Ionising Radiation (TC-IR) is concerned with the metrology of ionising radiation related to medical, industrial, environmental, scientific

and radiation protection applications. During the Annual Meeting 2021 the advances of traceRadon were presented in February 2021.

## Newest Publications

Annette Röttger *et al* 2021: New metrology for radon at the environmental level *Meas. Sci. Technol.* 32 124008; <https://doi.org/10.1088/1361-6501/ac298d>

Florian Mertes *et al* 2021: Approximate Sequential Bayesian Filtering to Estimate Rn-222 Emanation from Ra-226 Sources from Spectra, SMSI Proceedings 2021; <https://doi.org/10.5162/SMSI2021/D3.3>

Annette Röttger *et al* 2021: Radon: das Schöne und das Biest, *StrahlenschutzPraxis* 2/2021, ISSN 0947-434 X

## Upcoming Events

Staying in touch with the project is easy: Just follow us on twitter: @traceRadon:

<https://twitter.com/traceradon>

A website is available at <http://traceradon-empir.eu/>

A notice board was established on <https://www.researchgate.net/project/19ENV01-traceRadon> as well.

The consortium is currently preparing to contribute to the following meetings, conferences or workshops:

|                |                                                                  |
|----------------|------------------------------------------------------------------|
| January 2022   | EURADOS Annual Meeting 2022                                      |
| February 2022  | EURAMET Annual Meeting 2021                                      |
| May/June 2022  | 6th European Congress on Radiation Protection (IRPA)             |
| May/June 2022  | ICRM Low-Level Radioactivity Measurement Techniques (LLRMT) 2022 |
| September 2022 | ICOS Science Conference 2022                                     |

## Acknowledgements

EMPIR 19ENV01 traceRadon was launched in summer 2020. It is supported by a broad global scientific community within climate research, radiation protection and metrology. All stakeholders are united by the goal of providing new and improved data for science, the public and decision makers.



In the preparation of the project, the communication and discussion within EURADOS WG 3 turned out to be very effective. Further thanks go to EURAMET e.V., the European Association of National Metrology Institutes which made such a project possible within the EMPIR framework program.

For the time being, the project traceRadon has established the following collaborations by a Letter of Agreement (in the order of signature date): Collaborators by signed letters of agreement:

1. Universität Heidelberg, Germany
2. ANSTO, Australia's Nuclear Science and Technology Organisation, Australia
3. ERA, European Radon Association, Europe
4. Met Office, United Kingdom
5. University of Novi Sad, Serbia
6. Politecnico di Milano, Italy
7. University of Cordoba, Spain
8. EURADOS, e.V., Europe
9. Universität Siegen, Germany
10. IRSN, France
11. ARPA Piemonte, Italy
12. ARPA Valle d'Aosta, Italy



The consortium is grateful to have this powerful support from colleagues worldwide! Further collaboration interest is welcome.

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