EMPIR EURAMET
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NEWSLETTER

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Content

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^{-3} to 100 Bq·m^{-3} 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 15 collaborators are working toward this goal!

News from the work packages and recent developments

WP 1: Outdoor radon activity concentrations

In the course of this work package two new radon (^{222}Rn) emanation sources for primary calibration of ^{222}Rn activity concentration measurement devices traceable to SI were developed. They are meant for implementation at atmospheric monitoring network stations.
One of the sources, developed at the Czech Metrology Institute (CMI) is based on an emulsion of salts of fatty acids in silicone rubber that was formed from the weighed standard solution. The emulsion was allowed to polymerize in a steel tray with the following dimensions: 70·30 mm^2. The activity of the standard was determined by the weight of the ^{226}Ra-solution, the weight of the resulting emulsion and the losses (< 0.1 %).

Figure 1: Photo of an IRSD.

The other source is a new kind of ^{222}Rn emanation source developed at PTB, called the Integrated Radon Source Detector (IRSD). The IRSD allows for quasi online, data-driven computation of the activity concentration. One of the IRSD already produced is shown in Figure 1 and the schematic process of the ^{222}Rn production and its determination in Figure 2. The basic setup is simple: a Passivated Ion implanted Planar Silicon (PIPS) detector is coated with radium chloride (^{226}RaCl$_2$) using thermal, physical vapor deposition. During the radioactive decay of ^{226}Ra and ^{222}Rn α-particles of differing energies are emitted in random direction. Those emitted in the direction of the PIPS detector can be detected. They are shown in Figure 2 as α-particles in the PIPS-Detector area, below the dashed line. Due to their different α-energy, it is possible to distinguish their origin (^{226}Ra, ^{222}Rn, ^{218}Po) and with that to determine the different contributions to the activity detected (A^S_{Ra}, A^S_{Rn}). Having this in mind, one can calculate A^V_{Rn}, which is not detected by the IRSD. This calculation is performed using a new statistical method based on Bayes filtering and state estimation.

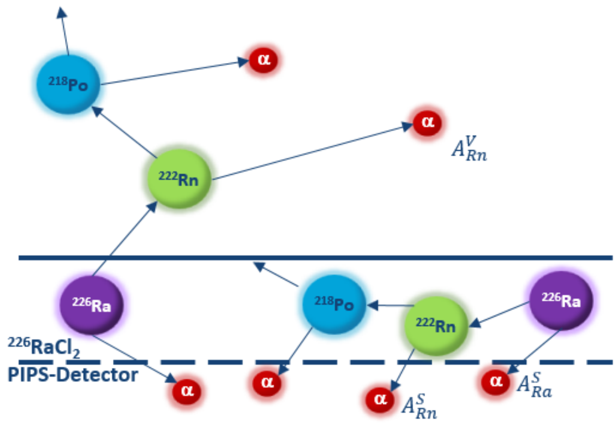

Figure 2: Schematic of the IRSD system. Shown are the Passivated Ion implanted Planar Silicon (PIPS) detector, the layer of ^{226}RaCl$_2$ on top of it and a part of the radioactive decay chain of ^{226}Ra.

Measurements with both sources at two experimental sites, SUJCHBO and PTB, were performed for quality assurance. At both sites the sources were installed in a reference volume to determine the calibration factor for a different radon activity concentration reference instrument (RRI) per site.

At SUJCHBO the sources are connected to a reference volume with the RRI inside, using a known, constant flow of radon free air through the source reference volume system.
At PTB each source, as well as the RRI, was placed inside a closed volume for several weeks to measure the build-up of ^{222}Rn inside the reference volume. The handling of the sources, and the procedure of data analysis was chosen individually according to the procedure and capabilities of both labs. The result is given in Table 1.

Table 1: Comparison of determined calibration factors with standard uncertainties at SUJCHBO and PTB for both source types.

	PTB IRSD	CMI source
PTB-RRI	1.019 ± 0.015	1.056 ± 0.019
SUJCHBO-RRI	0.88 ± 0.04	0.95 ± 0.01

It is important to note, that the performance of the sources is highly dependent on their application. The method of application is likewise important as the quality assurance and the traceability. Overall, both the comparison demonstrated an agreement but at different levels and with different methods.

The results in WP 1 overachieved the expectations: Two new source types are available, and a completely new type of instrument, the IRSD was developed. The IRSD provided the ground for a new calibration method, which is more accurate and suitable for very low activity concentrations of radon. With the IRSD, traceability for two new transfer standards (ANSTO 200 L and ARMON v2.0) was generated and with that the traceability to the SI is available in field for the first time. This impressive step forward in the metrology for the radon activity concentration is visible in the comparison of the transfer standards shown in Figure 3.

The performance of the transfer standard instruments at the intended reference activity concentrations is shown in Figure 3. To show the step forward, the measurement cycle was monitored by two commercial AlphaGUARDs in parallel. An additional AlphaGUARD was used for background monitoring of the lab.

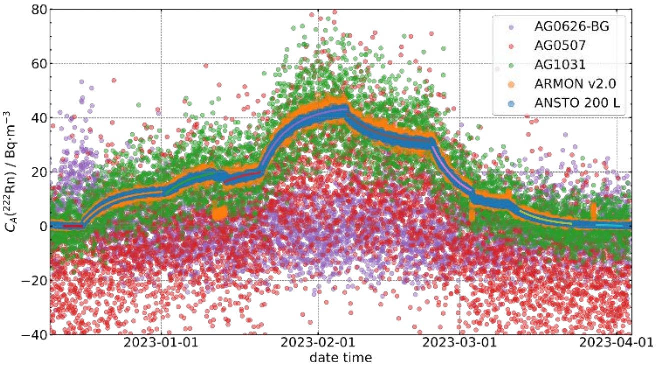

Figure 3: First presentation of the raw data of the long term comparison of five radon detectors at PTB. The newly developed detectors are the ARMON v2.0 (orange, maintenance of drying unit not removed, yet) and ANSTO 200 L (blue).

WP 2: Radon flux measurements

In agreement with the planned traceRadon project activities, after designing and building a traceability chain for continuous radon flux measurements, see Figure 4, over the first two years (2020-2021) of the project, four intense radon flux campaigns were carried out all over Europe between 2022 and 2023.

Measurements of dose rate, radon flux, water soil content, radionuclides activity in soil and physical soil parameters were performed at PTB (Germany), ENEA (Italy), SACLAY (France) and WAO (United Kingdom) stations. We observed a good agreement between measured radon fluxes and modeled ones at ENEA (Figure 5) and PTB stations on a daily basis. However, a high sub diurnal variability was observed in the radon flux measurements which is not followed by the model output. Data are currently under analysis for publication within open access journals.

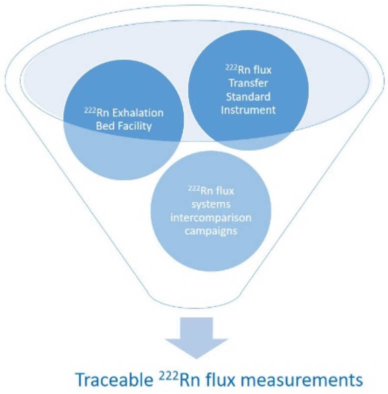

Figure 4: Traceability chain designed and built within the traceRadon project for continuous radon flux measurements.

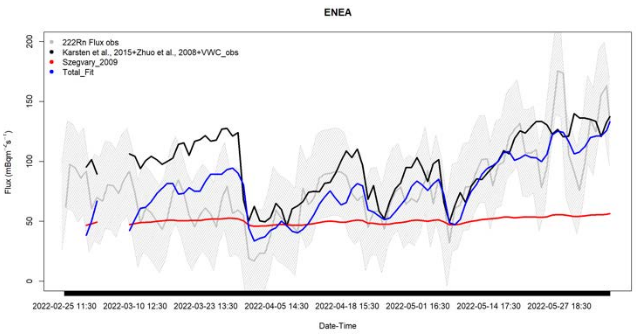

Figure 5: Daily means of radon fluxes obtained by different approaches at ENEA station.

WP 3: Radon flux models and inventories

High-resolution European radon flux maps have been calculated with the process-based radon flux model using soil parameter maps as well as spatially and temporally resolved soil moisture reanalysis datasets (ERA5-Land and GLDAS-Noah v2.1) for application in the Radon Tracer Method and in atmospheric transport modelling studies. Both versions of the maps are available for testing on the ICOS Carbon Portal (https://www.icos-cp.eu). The differences between the two versions (presented in the previous newsletter) provide a first idea on the uncertainties associated with the input data, but an evaluation of the model itself requires further comparison with actual observations.

Assessment of the process-based radon flux model include assessment of soil moisture data. For that purpose, soil moisture data from ERA5-Land and GLDAS-Noah reanalysis is compared with site measurements from the International Soil Moisture Network (ISMN) and from the Integrated Carbon Observation System (ICOS) Research Infrastructure.

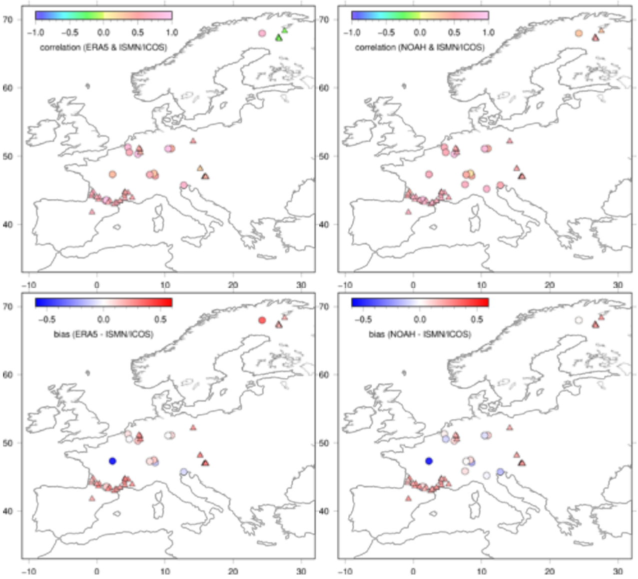

Figure 6: Comparison of soil moisture reanalysis with measurements at ISMN (triangles) and ICOS (circles) sites, correlation (upper maps) and bias (lower maps).

The correlation and bias between reanalysis and station data is shown in Figure 6. In general, the GLDAS-Noah reanalysis is in better agreement with the station data. The maps highlight the very limited spatial distribution of the available in-situ data, hindering a thorough assessment of the reanalysis data.

In-situ soil moisture measurements at the traceRadon campaign sites (PTB, ENEA, and SAC) indicate a bias below 0.1 m^3/m^3 between reanalysis and site data, being in general lower for the GLDAS-Noah reanalysis (Figure 7).

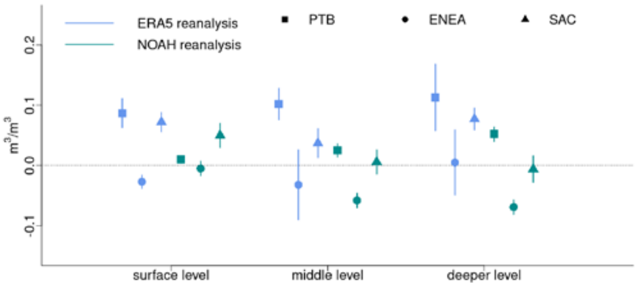

Figure 7: Average of differences between reanalysis and soil moisture measurements at the campaign sites for three depth levels.

The dose rate measurements and the soil water content based on the count rates of ^{40}K and ^{208}Tl photo-peaks have been used at the different campaigns (see Figure 8). At the stations where gravimetric measurements of soil moisture are available, they have been used for calibration of the ^{40}K and ^{208}Tl count rate methodology. Then, a calibration fit that would be usable for all the station was calculated and applied.

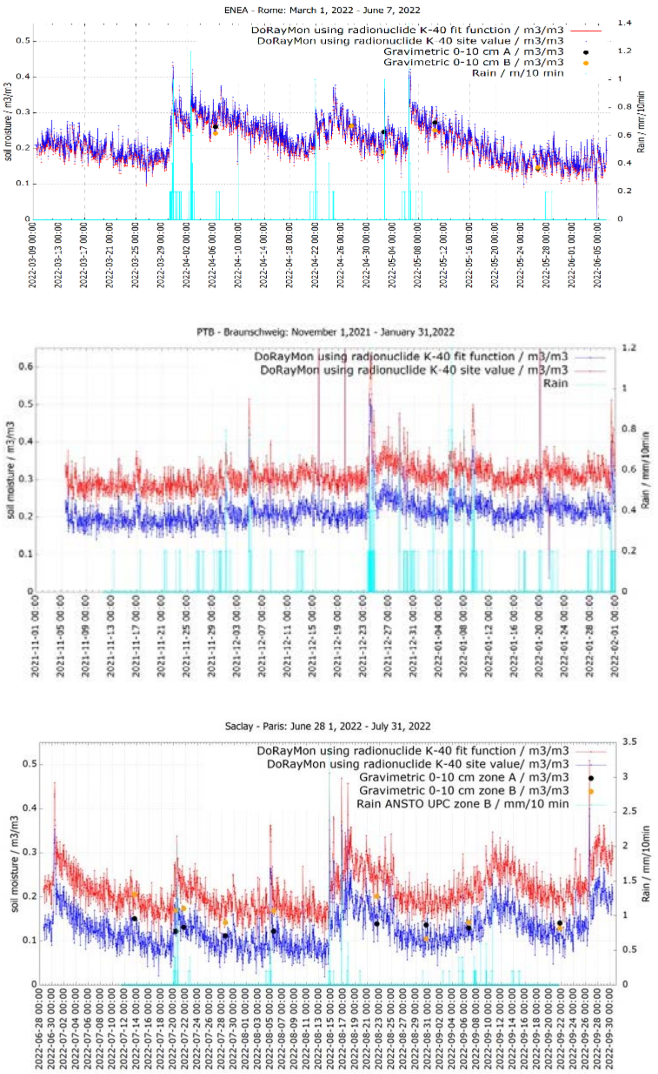

Figure 8: Comparison of soil moisture content by using site calibration equation and all-sites calibration equation for ^{40}K count rates. Top: ENEA, Middle: PTB and bottom: Saclay. Data of gravimetric measurements are also included in the plots.

In addition, the experimental time series of environmental dose rate, soil temperature and soil water content at each station were analyzed by multiparametric fits to reproduce the observed radon fluxes and to compare them with results from available radon flux models (Karstens et al., 2015) and inventories (Szegvary et al., 2009). Figure 5 shows an example at the ENEA station of daily mean radon fluxes.

Comparison of the radon flux measured during the traceRadon campaigns with the radon flux from the process-based radon flux map (Figure 9) shows large variations across sites, with lower bias obtained for radon flux maps based on the NOAH reanalysis.

Further evaluation of the two radon flux map versions is attempted by comparing the atmospheric radon activity concentration resulting from atmospheric transport model simulations based on the maps to measurements available at several stations in Europe, e.g. in the ICOS network and in the UK. Although not all these measurements are fully calibrated yet, the comparison can give first indications on the representativeness of the radon flux maps.

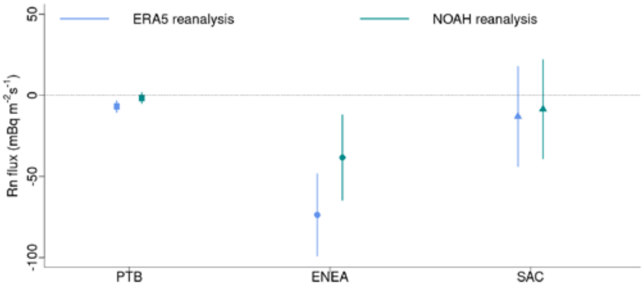

Figure 9: Average difference between radon flux measured at the three campaigns and from the process-based radon flux map based on ERA5-land and GLDAS-Noah reanalysis.

Transport model simulations have been conducted with the NAME model at NPL and UoB, and with the STILT model implemented at the ICOS Carbon Portal. Figure 10 shows monthly values of measured and STILT modelled afternoon (13:00-18:00 local time) radon activity concentration at the ICOS station Saclay (inlet at 100 m above ground). Afternoon was selected for the comparison because then the atmosphere is usually well mixed and transport models are supposed to perform better in these situations.

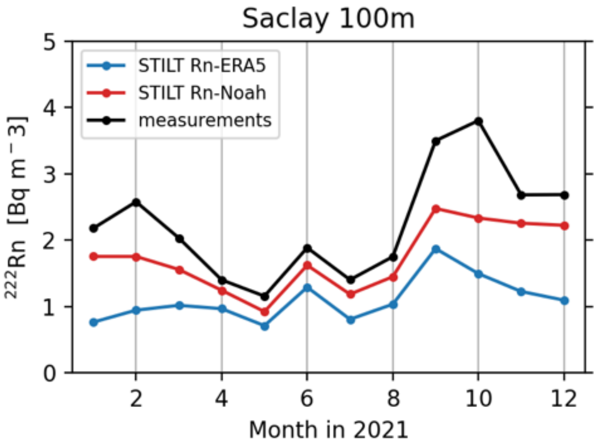

Figure 10: Figure 10: Monthly mean afternoon radon activity concentration measured at Saclay tall tower (100 m inlet height) and simulated using the atmospheric transport model STILT together with the radon flux maps based on different soil moisture reanalysis (ERA5-Land and GLDAS-Noah).

Both STILT simulations reproduce the seasonal cycle quite well, albeit underestimating the high radon concentrations in October 2021, when radon was accumulating in the lower troposphere during a stable high-pressure situation over Europe.

This indicates that disentangling deficiencies in the radon flux maps and deficiencies in the model transport requires careful selection of the situations to investigate. Overall, the ERA5-Land based simulations result in a systematic underestimation of the radon concentrations at Saclay and at many other sites, again pointing to too low radon fluxes in this version of the map, while the GLDAS-Noah based simulations show smaller differences.

WP 4: Outdoor radon and radon flux in radiation protection

Concerning the radiation protection research area, two new applications were developed and applied that involve the use of outdoor radon and radon flux data.

In the first application we investigated the possibility of using outdoor radon and radon flux to predict the radon risk of areas. We compared these two parameters with 28 other parameters used for radon risk prediction such as geological information, physical and chemical soil properties and weather data. We used the gridded indoor radon concentrations of the European Atlas of Natural Radiation as proxy for the radon potential of an area and as target variable in a machine learning workflow. We followed an approach outlined by Peterman et al. (2021) and used a random forest model for prediction on a data set covering the area of Belgium and Germany. A sufficient number of outdoor radon measurements are already available in these two countries.
We repeatedly built 500000 random forest models with different input features and evaluated the model performance. The German data set was used to train the model and as well to evaluate the model performance in a five-fold cross validation. The Belgian data was only used for performance evaluation. This reproduces one of the core ideas of an European radon potential map, where a model developed in one country can also be used in another country to predict the radon potential. To evaluate the model performance the mean square error of predictions and actual target values was used.
The models that performed best on the Belgian test data and German validation data set were selected and optimized. These two models only share one input feature (soil moisture) but otherwise use different input features. The Belgian model uses the radon flux for prediction. The impact of the input features shows high variabilities, where for the German model the performance gain of a single input feature is in the range of a few percent, whereas for the Belgian model a single input

feature (the number of coarse fragments in the top soil) more than doubles the model performance (Figure 11).

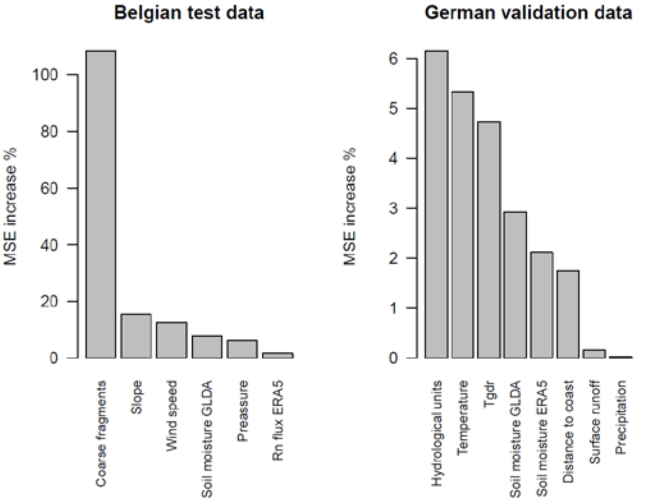

Figure 11: Two plots of feature importance for the Belgian and German model, where performance has been evaluated on the Belgian test data and the German validation data set. MSE (mean squared error) increase shows how much a model gains in performance by adding a feature, compared to the same model but without this feature.

The key message of our work is that the choice of input features to be used as predictors for the radon potential strongly depends on the setting and area. A predictor proven to work in one area might not necessarily work in another, and there is still a potential for input features not selected by our models to work in other areas. We showed that radon flux can be used for the prediction of the radon potential and improve model performance, but it depends on the area if it is needed as parameter. In addition, outdoor radon could become a valuable predictor in other settings and areas even if not selected by our models.

A promising outlook of our work is, that the accuracy of the prediction of the Belgian model is quite high (R^2 = 0.58 of target value and prediction), considering that only data from Germany has been used to build the Belgian model. Still the indoor radon concentrations of the Belgian data are essential to evaluate the model performance.

The second application relates to improve warning alarms of gamma dose rate monitoring. These real-time measurements are widely used in nuclear/radiological emergency preparedness and response systems. In order to be able to detect dose rate peaks due to artificial sources, it is important to develop automatic methods to filter false positives, i.e. peaks due to natural events. EURDEP (European Radiological Data Exchange Platform) is an example of network susceptible to such problems and could benefit of these previous commented filtering methods.

Indeed, peaks observed in ambient dose rate time series due to natural events are frequent and they can create false positive responses of the monitoring networks. Examples of the possible natural causes are: i) the so-called radon wash-out peaks due to the deposition on the ground of radon progenies after rain; ii) atmospheric radon accumulation peaks due to the variation of the height of the planetary boundary layer during the night.

In the framework of the traceRadon project a first intercomparison exercise (icp) of different automatic methods to identify and to classify ambient dose rate peaks has been organized in collaboration with researchers from EURADOS (European Radiation Dosimetry Group). The exercise was carried out in two rounds. In the 1st round, a 3-month time series of ambient dose equivalent rate $H^*(10)$ data with hourly resolution were provided to the six participants which had to design and develop method and criteria to reach the icp goal. In the 2nd round rain time series data were also provided to the participants. Each participant may provide peak occurrence dates (start-end) and their classification (natural, artificial, etc.). As an example, Figure 12 shows the results obtained from one of the participants.

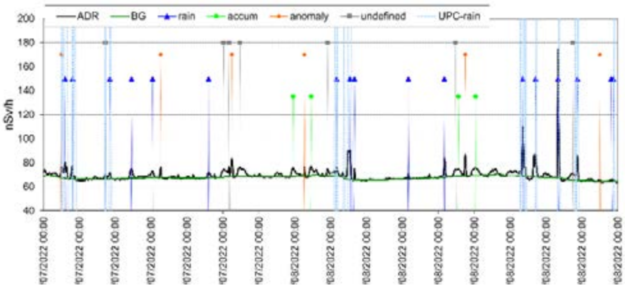

Figure 12: Ambient dose rate (ADR), background (BG) and rain trends (UPC-rain). Peaks identified and their classification (rain, night accumulation of radon (accum), anomaly and undefined).

First conclusions from the exercise are: a) sensitivities for peak identification are different for each method and b) rain information is fundamental for a good classification of the peak (in some case spectrometric data are needed).

Detailed results on both applications will be published soon in peer review journals.

Past Events

traceRadon 2nd Scientific Workshop and Training Courses

The 2nd traceRadon Scientific Workshop and two Training Courses were held in the context of this project at PTB premises on 14th and 15th March 2023. Almost 40 scientists from all over Europe came together in the beautiful city of Braunschweig, Germany, to learn about the results of the traceRadon project in both, classical talks during the Scientific Workshop, and a more practice-oriented format during the Training Courses. Due to this special setup the events were in-person only. Half of the participants were female.

The Scientific Workshop served to give an overview of the project and the results produced in its framework. Discussions were lively and continued well into the excellent dinner at a restaurant at Braunschweig city centre. This, as well as the lunch on the following day and the coffee breaks, gave the participants and presenters plenty of time to discuss the interesting ideas and their concrete implementation into scientific everyday life.

Figure 13: Group photo in front of the PTB Vieweg – building, where the 2nd traceRadon Scientific Workshop and the two Training Courses were held.

over the world were able to attend to share their expertise with each other. The theme of the conference was "Tracking progress to carbon neutrality" and the conference program covered a wide range of themes relating to primary scientific research as well as applications for industry and manufacturing.The Training courses were meant to provide practical guidance in the improvements of radon measurements and therefore less focused on results and more focused on methods. This also included a hands-on session for the application of the Radon Tracer Method. The participants were asked to bring their laptops and applied the Radon Tracer Method on-site. The Training Courses also included lab tours to show measurement devices at the lab and measurements at the outdoor reference field. This gave the participants a unique opportunity to see the size of transfer standards and laboratory equipment. Visiting the reference field (ERADOS) for outdoor radon and radon flux measurements proved the challenges that outdoor measurements present.

Together the scientists from all over Europe spend two days to improve future radon and radon flux measurements with the results achieved by traceRadon.

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.

ICOS Science Conference 2022

The 2022 ICOS Science Conference was held as a hybrid event between the 13th and 15th September 2022 in Utrecht, Netherlands and over 420 scientists from all
Dafina Kikaj, from the National Physical Laboratory (NPL) in Teddington, UK, represented traceRadon and gave a plenary talk titled Importance of harmonizing radon datasets for reducing uncertainty in greenhouse gas emission estimates. The presentation introduced the attendees to a later session, where the Radon Tracer Method and its use for greenhouse gas emission monitoring was discussed. The session consisted of several talks and poster presentations, and the traceRadon project partners provided 14 contributions for this session which are listed below.

The project coordinator, Annette Röttger (PTB, Germany), comments on the successful conference session: “To combat climate change, we need reliable data on greenhouse gas concentrations. But it is even more important to quantify greenhouse gas fluxes as discussed at the ICOS conference. The traceRadon project is making an important contribution to generating quality-assured data in this scientifically challenging field. Our measures to combat climate change are expensive: for broad acceptance in society and politics, we need the certainty that our data are of the highest possible quality. This is only possible with metrology!”

The conference provided further impetus to take up the new possibilities of radon measurement technology worldwide. Dr. Claudia Grossi, from the Technical University of Catalunya, Dr. Annette Röttger from PTB (coordinator of the traceRadon project) and Dr. Scott Chambers (ANSTO, Australia) were the conveners of the special ICOS session.

Results of these contributions are currently prepared for open access publications within the special issue of the Atmospheric Measurement Techniques: ‘Outcomes of the traceRadon project: radon metrology for use in climate change observation and radiation protection at the environmental level’.

ROOMS 2022

ERA and DSA welcomed the participation of national and international members of the scientific community, radon testers, diagnosticians, mitigators, educators, public stakeholders, businesspeople and authorities to the annual ROOMS conference in Bergen, Norway, 27th and 28th September 2022. The conference was dedicated to radon preventive measures and mitigation in new and existing buildings. State of the art of radon metrology in the field of low activity concentrations was presented by a contribution from traceRadon.

ICRM 2023

The International Committee for Radionuclide Metrology (ICRM) was pleased to announce that the conference was organized by Institutul Naţional de Cercetare-Dezvoltare pentru Fizică şi Inginerie Nucleară "Horia Hulubei" (IFIN-HH), Bucharest-Măgurele, Romania, during the period 27th-31st March 2023.
The main purpose of the conference is to enhance the international collaboration in the field of radionuclide metrology by presenting new developments and applications and facilitating the exchange of scientific information between the participants. Three contributions from the traceRadon consortium were presented and discussed.

EURAMET TC-IR 2023

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 TC-IR meeting the advances of traceRadon were presented by the Coordinator in February 2023.

EGU 2023

EGU General Assembly 2023 brings together geoscientists from all over the world to one meeting covering all disciplines of the Earth, planetary, and space sciences. Members of the consortium presented results in different sessions.

Upcoming Events

EURADOS AM 2023

The European Radiation Dosimetry Group (EURADOS) is a network of 81 European institutions (Voting Members) and more than 600 scientists (Associate Members). Presentations dealing with the results of traceRadon will be given in the WG3 of EURADOS in June 2023.

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/

Our Publications

1. Mertes, F et. al.: D3.3 Approximate sequential Bayesian filtering to estimate Rn-222 emanation from Ra-226 sources from spectra, https://doi.org/10.5162/SMSI2021/D3.3
2. Röttger, A. et al: New metrology for radon at the environmental level *2021* Meas. Sci. Technol. *32,* 124008, https://doi.org/10.1088/1361-6501/ac298d
3. Radulescu, I et al.: Inter-comparison of commercial continuous radon monitors responses, Nuclear Instruments and Methods in Physics Research Section A, Volume 1021, 2022, 165927, https://doi.org/10.1016/j.nima.2021.165927
4. Mertes, F. et. al.: Ion implantation of ^{226}Ra for a primary ^{222}Rn emanation standard, Applied Radiation and Isotopes, Volume 181, March 2022, 110093, https://doi.org/10.1016/j.apradiso.2021.110093
5. Čeliković, I. et. al.: Outdoor Radon as a Tool to Estimate Radon Priority Areas - A Literature Overview, Int. J. Environ. Res. Public Health 2022, 19, 662, https://doi.org/10.3390/ijerph19020662
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Radiation and Isotopes, Volume 196, 110726, 2023, https://doi.org/10.1016/j.apradiso.2023.110726.

13. Mertes, F., et al.: Approximate sequential Bayesian filtering to estimate ^{222}Rn emanation from 226Ra sources using spectral time series, J. Sens. Sens. Syst., 12, 147–161, 2023, https://doi.org/10.5194/jsss-12-147-2023

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

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 WG3 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 Codoba, Spain
8. EURADOS, e.V., Europe
9. Universität Siegen, Germany
10. IRSN, France
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