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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 the very moment 16 partners and 8 collaborators are working on this goal!

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Introduction to the work packages and early developments

WP 1: Outdoor radon activity concentrations

The aim of this work package is to develop traceable methods for the measurement of outdoor low-level radon activity concentrations in the range of 1 Bq·m^{-3} to 100 Bq·m^{-3} with uncertainties of 10 % (k=1) to be used in climate and radiation protection networks. These methods will include two new traceable Rn-222 emanation sources below 100 Bq·m^{-3}, a transfer instrument traceably calibrated with these new sources and a calibration procedure suitable to enable a traceable calibration of environmental atmospheric radon measurement systems in the field.

The EMPIR project 16ENV10 MetroRADON developed the capability to measure SI traceable radon activity concentrations, in the range of 100 Bq·m^{-3} - 300 Bq·m^{-3} for indoor radon measurements. traceRadon will extend this metrology capability to outdoor low-level radon activity concentrations in the range of 1 Bq·m^{-3} to 100 Bq·m^{-3}.

The three main tasks of this WP are:

1.1) SI traceable low-level radon emanation source: To develop two new traceable, low level Rn-222 emanating sources (below 100 Bq·m^{-3}).

1.2) Radon activity concentration transfer standard: to develop a transfer standard for the traceable calibration of atmospheric radon monitors according to IEC 61577, at atmospheric radon levels (below 100 Bq·m^{-3}).

1.3) Traceable calibration in field: To use the Rn-222 emanating sources from Task 1.1) and the transfer standard from Task 1.2) to enable the traceable calibration of environmental atmospheric radon measurement systems in the field, with an hourly uncertainty below 15 % for k=1.

During the first 9 months period of this project, lots of activities have been carried out or have started as planned in the project proposal. Some highlights of these activities are presented here:

NPL performed a literature study of currently available radon sources for calibration of instruments capable of measuring radon activity concentrations below 100 Bq m^{-3}. Environmental parameter ranges were evaluated and a suitability list for in-field calibration was defined. That led to new needs for the characteristics of the radon source. Current commercially available radon sources are not fulfilling these characteristics at the very moment.

To fulfil these requirements, CMI with the help of SUJCHBO and PTB started the development of new low activity emanation sources. The two different principles applied are described here:

CMI developed a radon emanation source created from an emulsion of salts of fatty acids in silicone rubber, formed from the weighed standard solution. Traceability of the Ra-226 activity is established by weighing and gamma spectrometry: Using a stainless-steel cylindrical case with valves and aerosol filters, applying ultra-dried air and a mass flow controller with additional humidifier, to control the dilution of the activity concentration, a time-stable radon activity concentration is achieved.

Figure 1: Shows a picture of the CMI low-level flow-through emanation source with an activity of 1 kBq of Ra-226.

SUJCHBO uses this source to establish a time-stable radon activity concentration in their Low-Level Radon Chamber (LLRCh) as shown in Table 1.

Table 1: Examples of Rn-222 activity concentrations achieved at the SUJCHBO LLRCh with the flow rates applied and the time needed for stabilisation.

radon activity concentration in Bq·m^{-3}	flowrate in L·min^{-1}	stabilization time in min
100	1.22	27
80	1.53	18
40	3.10	8

PTB developed low-level, low activity emanation sources based on the electrodeposition from a carrier-free Ra-226 solution on a stainless-steel plate. The emanation rate of Rn-222 of these sources is followed online via γ-spectrometry using portable scintillation detectors like $LaBr_3$-Cristals. Further development applying physical vapor deposition of Ra-226 salt is in progress. The emanation of the Rn-222 follows differential equations, by nature, that include the build-up and decay of the Rn-222. Therefore, the measurement of the disequilibrium of the Ra-226 and the Rn-222 progeny is correct only for stable states. To overcome this constraint, an algorithm applying a new statistical method based on Bayes filtering (Kalman filter, assumed density filtering, see Barber 2006) has been developed and implemented (see *Figure 2*). With this

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algorithm the emanated Rn-222 in the unit atoms per second as well as the associated uncertainty is determined online from spectrometric data including the knowledge of the already measured spectra (Mertes 2021).

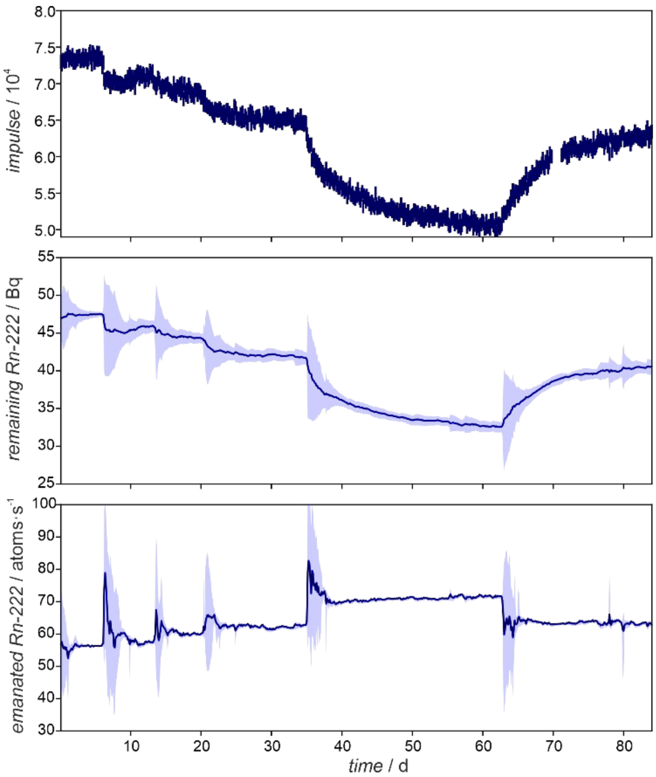

Figure 2: The first (upper) diagram shows the integral count rate above the highest energy of Ra-226. Only events from Rn-222 and its progeny are included. The variation of this count rate is achieved by variation of environmental parameters. The second diagram presents the remaining Rn-222 activity in the source, given in Bq. The light blue area represents the 90 % confidence interval for the uncertainty of the measurement. The third diagram gives the primary desired result, which is the emanated number of Rn-222 atoms, represented as atoms/s.

UPC with the help of partners and collaborators conducted a literature review of the currently available radon monitors capable of measuring the activity concentrations below 100 Bq·m^{-3}. This comprehend elaboration results in the desired matrix of properties for usable radon activity concentration monitors. The consortium has proposed suitable available monitors to be used within traceRadon accordingly.

WP 2: Radon flux measurements

The aim of this work package is to improve the accuracy of radon flux measurements for the purpose of: 1) identifying Radon Prone Areas (RPAs) for radiation protection goals; and 2) retrieving indirect greenhouse gas (GHG) fluxes using the Radon Tracer Method (RTM).

The four main tasks of this WP are:

2.1) Reference radon flux monitor: To design, establish and characterize (1) a radon “exhalation bed” as a standard reference system (by UC as ISO/IEC 17025 accredited laboratory for radon exhalation measurements), and (2) an automatic radon flux system as a Transfer Standard (TS) to calibrate existing radon flux monitors under laboratory conditions.

2.2) Continuous radon flux monitors under field conditions: To compare at least two monitors with different operational principles, each capable of making automated ongoing radon flux measurements, ‘in situ’ under environmental conditions in both low and high radon flux areas.

2.3) Measurement campaigns at selected Atmospheric Monitoring Network Stations (AMNS) and Radiological Monitoring Stations (RMS): To organize and perform radon flux measurement campaigns at AMNS and RMS using the automated radon flux monitors built and compared from tasks 2.1 and 2.2.

2.4) RTM application at AMNS: To develop a standard operating procedure for application of the RTM at AMNS to facilitate the estimation of GHG fluxes. This task will focus on the analysis of a set of radon activity concentrations, GHG mixing ratios and meteorological data from a selected AMNS measured over a broad range of atmospheric conditions. The consistency and sensitivity of several approaches for applying the RTM will be evaluated and compared with the aim to produce – for the first time – a general standard protocol for application of the RTM.

During the first 8 months of the project, several activities have been carried out in agreement with the project proposal as detailed in the following activities.

SUJCHBO, in collaboration with partners, carried out a literature review of continuous radon flux monitors in order to analyze the different radon flux systems available by identifying their strengths, weaknesses and qualities of output. UPC subsequently compiled this information as a preliminary internal report to be used in the following activities.

Based on the previous results, UPC is preparing a rental contract with ANSTO for an autoflux system (*Figure 3*), the best radon flux system option currently available, which will be tested for suitability to be used as a transfer standard in the following activities of the project.

Based on the needs identified in the previous report, UC has worked on the design of a radon “exhalation bed” as a calibration facility. The main structure of the exhalation bed is constructed from stainless steel and has a total surface area of 1 m^2, with an effective height of 0.2 m as shown in *Figure 4*.

The soil used has a radium activity of ≈ 18 $kBq \cdot kg^{-1}$, and has been dried and winnowed by a sieve to obtain a grain size of 2 mm. The resulting soil was homogenized to achieve a radon flux as spatially consistent and stable as possible under laboratory conditions. Currently, work is focusing on determining the theoretical and experimental radon flux values.

Figure 3: ANSTO autoflux system for automatic radon flux measurements.

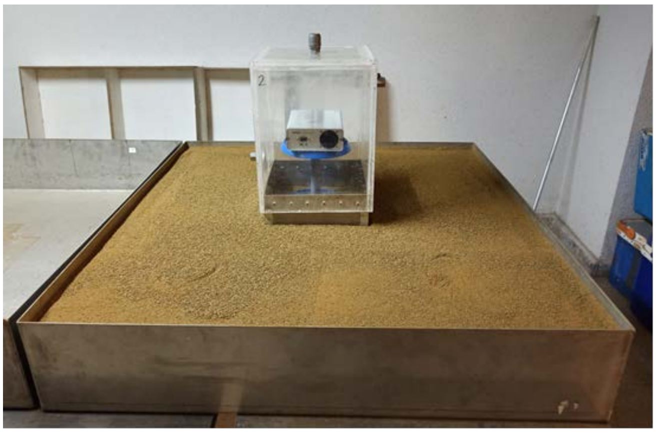

Figure 4: The UC designed exhalation bed with an accumulation chamber and a radon monitor shown for scale.

Measurement campaigns at selected Atmospheric Monitoring Network Stations (AMNS) and Radiological Monitoring Stations (RMS): Members of this activity organized a meeting in December 2020 to start a design of the radon flux campaigns. The variables and parameters to be measured were decided and the main requirements of the sites were agreed upon.

RTM application at AMNS: UVSQ, in collaboration with partners, selected the ICOS station where the RTM will be tested. The station of Saclay was chosen in agreement with its characteristics, i.e. it can make vertical radon gradient measurements and it has the capability for making GHG mixing ratio measurements at multiple heights and thus estimating GHG fluxes by two independent methods.

Figure 5: Tall tower of the atmospheric ICOS station of Saclay, France.

Saclay (*Figure 5*, SAC) is located 30 km south-west of Paris, 482.7217 °N, 2.142 °E, 160 m above mean sea level. A 3-month intercomparison of radon monitors was previously carried out at this site in 2016 (Grossi et al., 2020). GHG and meteorological data have been available in the ICOS database since 2015, with radon

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at 100 m above ground level since the end of 2016 and radon at 2 m above ground level since January 2019.

The RTM had been previously applied at the nearby Gif-sur-Yvette site, 2 km west of SAC (Belviso et al., 2013, Belviso et al., 2020). Yver et al., 2009 summarized the radon flux estimates before this date that ranged from 42(22) $Bq{\cdot}m^{-2}{\cdot}h^{-1}$ to 66(22) $Bq{\cdot}m^{-2}{\cdot}h^{-1}$ with an average of 52 $Bq{\cdot}m^{-2}{\cdot}h^{-1}$. From 2006 to 2009, additional measurements were made and used to assess a new radon map (Karstens et al., 2015). The values found for SAC varied between 18 $Bq{\cdot}m^{-2}{\cdot}h^{-1}$ and 54 $Bq{\cdot}m^{-2}{\cdot}h^{-1}$ for observations and models. Footprints of this station are available on the ICOS Carbon Portal, see https://stilt.icos-cp.eu/viewer/ for 2014 till the end of 2019. A new code to calculate the GHG flux is being developed using Jupyter python notebooks hosted on the ICOS Carbon Portal. This code allows to use the footprints and the radon map hosted on the Carbon Portal.

WP 3: Radon flux models and inventories

The aims of this work package are to validate current radon flux models and inventories using traceable measurements of radon flux and radon activity concentration supported by dosimetric and spectrometric data from the radiological early warning networks in Europe. In addition, to improve process-based radon flux maps that can be used in (1) the RTM, (2) atmospheric dispersion modelling and (3) radiation protection.

A literature review of more than 50 relevant publications was conducted to provide an overview of processes determining the production, transport and exhalation of radon from soils and strategies to estimate these fluxes based on soil parameters and/or dosimetric and spectrometric data from the radiological early warning networks in Europe. Based on this literature survey, suitable radon flux models and inventories are identi-fied.

A process-based radon flux model that describes the spatial and temporal variability of the radon flux based on soil properties, uranium content and soil moisture reanalysis will be updated in the framework of this project to cover recent time periods and increase temporal resolution from monthly to daily radon fluxes. The physical parameterization in the model will first be evaluated with data from the intensive measurement campaigns in WP2. Evaluation and uncertainty estimation of the resulting process-based radon flux maps, including required input data (e.g. soil moisture), will be performed by comparison with all radon flux measurements becoming available in traceRadon but also will they require compilation of additional data sets. Finally, all selected radon flux maps and inventories will be validated using the traceable radon flux measurements.

The dosimetric and spectrometric data from radiological early warning networks in Europe will be evaluated to further improve the radon flux models and to validate process-based radon flux maps. Suitable dose rate monitors and spectrometers will be characterized with respect to their inherent background and their sensitivity to small variations of ambient dose equivalent rate.

Figure 6: Example of a plume simulation for small variations of the ambient dose equivalent rate H(10).*

Figure 7: Low dose underground lab UDO II for absolute H(10) calibrations.*

These experiments will be conducted at the reference measuring sites at PTB, see *Figure 6* and Figure 7. Due to the Covid-19 pandemic, some of these campaigns will have to be slightly rescheduled. The calibrated detectors will then be used in the intensive measurement campaigns in WP2 to allow simultaneous measurements of radon flux and terrestrial ambient dose rate for the evaluation of radon flux parameterizations.

An additional validation of the radon flux maps will be attempted by using them as input in atmospheric transport model simulations and comparing the resulting atmospheric radon activity concentrations with continuous measurements from traceRadon measurement campaigns, the ICOS (Integrated Carbon Observation System) atmospheric measurement station network and other stations.

WP 4: Radon and radon flux in maps

This work package aims at providing and using dynamic outdoor radon concentration and radon flux maps for climate change research and radiation protection.
Useable dynamic radon activity concentration and radon flux maps will be provided through the Radioactive Environmental Monitoring web portal (REMon, https://remon.jrc.ec.europa.eu) operated by the Joint Research Centre of the European Commission and the ICOS Carbon portal. This new data will be linked to established data from EURDEP (European Radiological Data Exchange Platform) and EANR (European Atlas of Natural Radiation) and will be made available to scientists, policy makers and end users.

Concerning radiation protection, the focus is on the improvement of methods for:
• The identification of Radon Priority Areas (RPA)
• The estimation of radon wash-out peaks from total gamma dose rate data.

In the European Council directive 2013/59/Euratom, Art. 103, Paragraph 3 states that Member States should identify areas where it is expected that the annual average indoor radon concentration will exceed national reference levels in a significant number of dwellings. These areas are often called “radon priority areas - RPA”. The delineation of these areas will allow to plan and to prioritise measures within the national action plan and implies that radon measurements in workplaces located in these areas are required.
RPA can be estimated using different data, most commonly they are indoor Rn data, but geogenic quantities may be also used for estimating the so-called Geogenic Radon Potential and the Geogenic Hazard Radon Index. Geogenic quantities may be geology, tectonics, soil properties, radon activity concentration in the ground, geochemical concentrations, terrestrial dose rate and – in this project’s interest - outdoor radon activity concentration data and radon flux data. A literature review on the use of radon flux data for estimating indoor and outdoor radon activity concentrations as well as the use of the geogenic radon potential has been carried out. The literature analysis revealed great diversity in approaching outdoor radon origin and influence of different factors affecting outdoor radon measurements. The survey was somewhat complicated due to different terms used by different authors during several decades and the choice of keywords was challenging (radon flux, indoor and outdoor radon, soil gas concentration, geogenic potential). The information contained in the papers should serve as an input in the estimation of the geogenic RHI and in improving the identification of RPA.

In normal conditions (no contribution from fresh anthropogenic contamination) the ambient dose rate data represents the existing background radiation which results from natural radionuclides in soil/rock, cosmic contribution, and airborne radon. The solid progeny of radon can be washed out of the air by rain and become deposited on the ground. This results in peaks in the ambient dose rate detected. Radon wash-out events can closely mimic radioactive plume contamination deposition creating false positive responses in the EURDEP network. Therefore, better estimations of radon wash-out peaks will help to prevent false alarms in the EURDEP early warning system due to radon wash-out effects.

Past Events

Recent research outputs and new developments in the project are disseminated to the scientific communities via conferences, publications and workshops.

Kick-off meeting of traceRadon

Due to Covid-19 restrictions, this meeting was hosted as hybrid meeting: A face-to-face conference room was provided by PTB and a virtual meeting room was opened in parallel. The partners and two already involved collaborators discussed the upcoming tasks and organised the work.

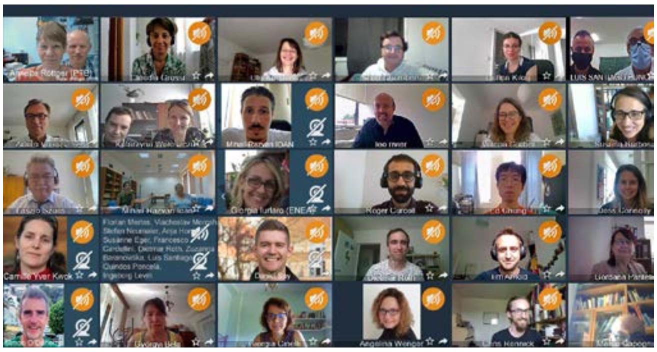

Figure 8: Kick-off meeting under Covid-19 restrictions in 2020.

First Scientific Workshop

A first scientific workshop on “Radon metrology for use in climate change observation and radiation protection at the environmental level” was set up.

The agenda of the workshop was announced via email and the EURADOS newsletter. The workshop was held at PTB Braunschweig as a hybrid meeting on 20th October 2020 and was free of charge. More than 80 scientists from all over the world gathered virtually and discussed the first results, planned research activities for the objectives of traceRadon.

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Find more information and all presentations from the scientific workshop here:

http://traceradon-empir.eu/

Scientific Workshop 20th October 2020
EMPIR 19ENV01 traceRadon

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The meeting will be held as a virtual meeting from PTB, Bundesallee 100, 38116 Braunschweig, Germany

08:00 - 09:00	Technical test of the (Web-) Conference
09:00 - 09:15	**Annette Röttger (PTB):** *Introduction to traceRadon*
09:15 - 10:15	**Florian Mertes and Stefan Röttger (PTB):** *Status of the Rn-222 emanation source development at PTB*
	Katarzyna Wołoszczuk (CLOR): *Calibration procedures of radon instruments*
	Razvan Ioan (IFIN-HH): *Capabilities at IFIN-HH*
	Scott Chambers (ANSTO): *Overview of the new portable ANSTO dual-flow-loop two-filter Radon-222 monitor*
10:15 - 10:30	Break
10:30 - 11:15	**Claudia Grossi (UPC):** *State of the art of radon flux measurements*
	Luis Quindos (UC) : *A radon bed exhalation facility*
11:15 - 12:00	**Ute Karstens (ICOS Carbon Portal Lund University) and Arturo Vargas (UPC):** *Radon flux maps and their validation using radon flux measurements and terrestrial data*
12:00 - 13:00	Lunch
13:00 - 13:45	**Giorgia Cinelli (JRC):** *Radon and radon flux in maps for radiation protection issues*
	Gordana Pantelic, Ivana Vukanac, Jelena Krneta Nikolic, Maciej Norenberg, Zuzanna Baranowska, Igor Celikovic, Milos Zivanovic (VINS): *Literature survey on the use of radon flux data for estimating indoor and outdoor radon activity concentrations*
13:45 - 14:00	Closing remarks

This project 19ENVT01 traceRadon has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme. 19ENV01 traceRadon denotes the EMPIR project reference.

Figure 9: Agenda of the first scientific workshop of traceRadon.

ACCOMC 2020

The first results from a portable two-filter dual-flow-loop radon detector developed by ANSTO in the scope of traceRadon was presented at the "Atmospheric Composition & Chemistry Observations & Modelling Conference incorporating the Cape Grim Annual Science Meeting 2020 and 16th Australia and New Zealand Aerosol Assembly".

EURADOS WG3.3

In the annual meeting of EURADOS e.V., the consortium presented first results in the WG 3.3. Additionally, the planned activities in the scope of comparisons using the environmental reference sites of PTB were presented in WG3.1.

Upcoming Events

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

https://twitter.com/traceradon

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:

23 February 21	19ENV01 traceRadon consortium meeting (M9)
19 – 30 April 21	EGU 2021
3 – 6 May 21	SMSI 2021
28 – 30 September 21	CIM 2021
In planning: 23 – 24 November 21	19ENV01 traceRadon consortium meeting (M18)

Information on the developing EMN for Radiation Protection

EURAMET's European Metrology Networks (EMNs) aim at realizing EURAMET's vision of building metrology capabilities based on high quality scientific research and on an advanced metrology infrastructure. The EMNs coordinate the identified measurement needs and formulate the measurement services and research and knowledge transfer accordingly. A CCRI webinar on "Metrology for Radiation Protection" was run on 2020-10-05, introducing among others the project traceRadon to the scientific community interested in the developing EMN on radiation protection. This seminar summarized the technical issues that need to be tackled for the coming years, including opportunities to get involved in research. The second part of the seminar covered a EURAMET initiative to engage with stakeholders and to strengthen existing knowledge transfers. The developing EMN on radiation protection is prepared by EMPIR 19NET03 supportBSS. In the scope of this, a strategic research agenda and two roadmaps are in development, covering the metrological needs of both the Euratom Treaty and Council Directive 2013/59 / Euratom.

The partners of supportBSS, together with other organisations providing a service in the field of radiation protection, are preparing a proposal for the GA of EURAMET in 2021 to establish the EMN on radiation protection.

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.

The first results are new technologies for calibrating high-sensitivity radon monitors. The consortium would like to thank the staff of project partners and collaborators of traceRadon, as well as all institutions and organisations that supported the project with recommendations.

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):

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

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

This project has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme. 19ENV01 traceRadon denotes the EMPIR project reference.

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