



## Publishable Summary for 19ENV01 traceRadon

### Radon metrology for use in climate change observation and radiation protection at the environmental level

#### Overview

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.

#### Need

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.

#### Objectives

The overall aim of this project is the development of metrological capacity (reference monitors, transfer standards and robust methodology) to measure low levels of radon in the environment, which can be used to determine emission reduction strategies of GHG and improve radiation protection of the general public.

The specific objectives are:



1. To develop traceable methods for the measurement of outdoor low-level radon activity concentration in the range of  $1 \text{ Bq m}^{-3}$  to  $100 \text{ Bq m}^{-3}$ , with uncertainties of 10 % for  $k=1$ , to be used in climate monitoring and radiation protection networks. These methods include two new traceable Rn-222 emanation sources below  $100 \text{ Bq m}^{-3}$ , a transfer instrument calibrated with these new sources to assure the traceability of the transfer instrument and a calibration procedure suitable to enable a traceable calibration of environmental atmospheric radon measurement systems in the field.
2. To develop the capability for traceable radon flux measurements in the field, based on the development of a radon exhalation reference system “exhalation bed” and a transfer standard (TS). To use this capability to harmonise existing radon flux instruments/methods by intercomparison campaigns. To develop a first standard protocol for the application of the radon tracer method (RTM) to enable retrieval of greenhouse gas fluxes at atmospheric climate gas monitoring stations and to use radon flux data for the identification of Radon Priority Areas (RPA).
3. To validate current radon flux models and inventories by the new traceable measurements of radon activity concentration and radon flux. To support the validation with dosimetric and spectrometric data from the radiological early warning networks in Europe. To improve process-based radon flux maps that will be available for use in the RTM, atmospheric dispersion modelling, and radiation protection.
4. To provide easy to use dynamic radon and radon flux maps for climate change research and radiation protection in line with Council Directive 2013/59/EURATOM, including their use to identify RPA and radon wash-out peaks.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (NMI, calibration laboratories), standards developing organisations (e.g. IEC, ISO) and end users in greenhouse gas monitoring and European radiological early warning networks.

### **Progress beyond the state of the art**

#### *Atmospheric low-level radon activity concentration measurements*

This project has begun developing the first time ever, traceable methods for measuring low-level outdoor radon activity concentrations in the range from  $1 \text{ Bq m}^{-3}$  to  $100 \text{ Bq m}^{-3}$  with uncertainties of 10 % for  $k=1$ . This is two orders of magnitude lower than the current state of the art developed in the EMPIR project 16ENV10 MetroRADON. This project will also go beyond the state of the art by targeting environmental radon activity concentrations in the outdoor air. In some Atmospheric Monitoring Network Stations (AMNS), monitors for radon activity concentrations are already in operation however their traceability has not yet been established. To be able to establish such a traceability chain this project is currently developing two new Rn-222 emanation sources below  $100 \text{ Bq m}^{-3}$  for the traceable calibration of atmospheric radon monitors. In addition, the project will develop a transfer standard for use in the field, which will then be validated in a field-based intercomparison.

#### *Radon flux measurements*

This project has begun developing a state-of-the-art radon flux calibration infrastructure, along with a transfer standard for radon flux monitors. Once the performance of the transfer standard has been validated using a radon exhalation reference system “exhalation bed”, it will be used in intercomparison campaigns to try and harmonise different radon flux measurement methods. The outcome will be a new validated radon flux dataset to assist with the identification of RPAs and the application of the RTM for GHG flux estimates.

#### *RTM procedures*

Environmental radon measurements in parallel with atmospheric GHG measurements can be used in the RTM to quantify GHG emission estimates. This project has begun work on developing the first ever procedure for the RTM approach for use at AMNS. The procedure will be systematically investigated, using traceability chains for the necessary radon activity concentration and radon flux input measurements and a validated radon flux map.

#### *Radon flux inventories and models*

Accurately representing the temporal variability and spatial distribution of the radon flux is essential for the application of radon flux maps and inventories in both atmospheric transport applications (for the climate



community) and the identification of RPAs. This project has started work that will go beyond the state of the art by traceably measuring, under field conditions, outdoor radon activity concentrations and surface radon fluxes that will be used for the validation of the radon flux maps and inventories. Further validation will include dosimetric and spectrometric data from radiological early warning networks in Europe.

#### *Dynamic radon activity concentration and radon flux maps to identify RPA*

The AMN, EURDEP and European Atlas of Natural Radiation (EANR) will provide this project with dynamic radon maps from the new traceable outdoor radon activity concentrations and radon flux measurements. This new data will be linked to established data from EURDEP and EANR and along with project data on natural radiological risks and outdoor radon activity concentrations, will be made available to scientists, policy makers and end users.

The use of radon flux data and maps in the identification of RPA has never been attempted before, due to a lack of robust data. This project has started work that will go beyond the state of the art by validating radon flux measurements and radon flux maps as parameters for the identification of RPA via the Radon Hazard Index (RHI). The RHI implemented in the preceding project 16ENV10 MetroRADON only contains static information. Therefore, this project will extend the RHI to include dynamic data (i.e. outdoor radon activity concentration and radon flux measurements) which will then be made available online.

## Results

1. *To develop traceable methods for the measurement of outdoor low-level radon activity concentration in the range of 1 Bq m<sup>-3</sup> to 100 Bq m<sup>-3</sup>, with uncertainties of 10 % for k=1, to be used in climate monitoring and radiation protection networks. These methods include two new traceable Rn-222 emanation sources below 100 Bq m<sup>-3</sup>, a transfer instrument calibrated with these new sources to assure the traceability of the transfer instrument and a calibration procedure suitable to enable a traceable calibration of environmental atmospheric radon measurement systems in the field.*

A literature study of currently available radon sources for calibration of instruments capable of measuring radon activity concentrations below 100 Bq m<sup>-3</sup> was performed. Based on this environmental parameter ranges were evaluated and a list of suitable parameters for in-field calibration was defined. This work provided the project with the necessary information on the needed characteristics for new radon sources (i.e. needs not currently met by commercially available radon sources). The development of new low activity emanation sources was started, and two different principles were used:

1. The first approach used is 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 a humidifier (to control the dilution of the activity concentration), a time-stable radon activity concentration is achieved. This approach established a radon source with time-stable radon activity concentration in a low-level radon chamber.
2. The second approach is the development of a 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 gamma-spectrometry using portable scintillation detectors such as Lanthanum Bromide (LaBr<sub>3</sub>) crystals. Further development applying physical vapor deposition of Ra-226 salt is currently in progress by the project. 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 only correct for stable states. In order to overcome this constraint, the project has developed an algorithm using a new statistical method based on Bayes filtering (Kalman filter, assumed density filtering). With this 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.

To establish the chain of traceability in field, the project has conducted a literature review of the currently available radon monitors capable of measuring the activity concentrations below 100 Bq·m<sup>-3</sup>. Based on the results the project was able to determine the desired matrix of properties suitable for usable radon activity concentration monitors and the consortium has identified suitable available monitors to be used within the next steps of the project.



2. *To develop the capability for traceable radon flux measurements in the field, based on the development of a radon exhalation reference system “exhalation bed” and a TS. To use this capability to harmonise existing radon flux instruments/methods by intercomparison campaigns. To develop a first standard protocol for the application of the RTM to enable retrieval of greenhouse gas fluxes at atmospheric climate gas monitoring stations and to use radon flux data for the identification of RPA.*

A literature review of continuous radon flux monitors was carried out including an analysis of the different radon flux systems currently available. The analysis was based on the radon flux systems' strengths, weaknesses and qualities of output and was subsequently compiled as a preliminary report. Based on the results, a suitable autoflux system, the best radon flux system option currently available, was identified for use in this project. The autoflux system will be tested for its suitability to be used as a transfer standard (TS) in the project.

Based on the needs identified in the preliminary report on the radon flux systems, a radon “exhalation bed” to be used as a calibration facility has been designed and constructed. The main structure of the exhalation bed is constructed from stainless steel and has a total surface area of 1 m<sup>2</sup>, with an effective height of 0.2 m. The soil used has a radium activity of  $\approx 18 \text{ kBq}\cdot\text{kg}^{-1}$ , and has been dried and winnowed by a sieve to obtain a grain size of 2 mm. The resulting soil was homogenised 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 for the radon “exhalation bed”.

The project has begun planning the measurement campaigns at selected AMNS and RMS. This has included deciding: (i) the design of the radon flux campaigns, (ii) the variables and parameters to be measured and (iii) the main requirements of the sites. RTM application at the AMNS was also planned by the consortium: including the selection of the station where the RTM will be tested. The station of Saclay was chosen due to its characteristics, i.e. it can make vertical radon gradient measurements, it has the capability for making GHG mixing ratio measurements at multiple heights and thus it can estimate GHG fluxes by two independent methods. Saclay is located 30 km south-west of Paris, 48.27217 °N, 2.142 °E, 160 m above mean sea level. The values for radon activity found at Saclay varied between  $18 \text{ Bq m}^{-2}\cdot\text{h}^{-1}$  and  $54 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$  for observations and models. The footprints of the Saclay 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 by the project using Jupyter python notebooks hosted on the ICOS Carbon Portal. This code will allow GHG flux to be calculated using the footprints and the radon map hosted on the ICOS Carbon Portal.

3. *To validate current radon flux models and inventories by the new traceable measurements of radon activity concentration and radon flux. To support the validation with dosimetric and spectrometric data from the radiological early warning networks in Europe. To improve process-based radon flux maps that will be available for use in the RTM, atmospheric dispersion modelling, and 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. The review was done to be able to estimate these radon fluxes based on soil parameters and/or dosimetric and spectrometric data from the radiological early warning networks in Europe. Based on this literature review, suitable radon flux models and inventories were identified. This has included 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, which is currently being developed by the project.

The dosimetric and spectrometric data from radiological early warning networks in Europe are being extracted to improve the radon flux models and to validate process-based radon flux maps. Suitable dose rate monitors and spectrometers are currently being calibrated and characterised in calibration laboratories of the partners. The schedule for the characterisation of the dose rate monitors and spectrometers with respect to their inherent background as well as their sensitivity to small variations of ambient dose equivalent rates has been agreed by the partners and the corresponding measurements will be performed at reference measuring sites at PTB.

4. *To provide easy to use dynamic radon and radon flux maps for climate change research and radiation protection in line with Council Directive 2013/59/EURATOM, including their use to identify RPA and radon wash-out peaks.*



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 review revealed significant diversity in approaches used to estimate outdoor radon origin and influence of different factors affecting outdoor radon measurements. The literature review was more complicated than expected due to different terms used by different authors over several decades which meant that the project's choice of keywords was challenging (radon flux, indoor and outdoor radon, soil gas concentration, geogenic potential). However, the project overcame these challenges and the information collated from the literature review will be used as input for the estimation of geogenic RHI and for improving the identification of RPA.

### Impact

The project has created a website at <http://traceradon-empir.eu/> and a traceRadon newsletter in order to promote itself to end users. A Twitter account for the traceRadon project has also been created <https://twitter.com/traceradon>. The account is used to post updates on the traceRadon project and re-tweet relevant climate and atmospheric science posts.

Further to this, the project has been presented at 6 different events such as the "Atmospheric Composition & Chemistry Observations & Modelling Conference, the Romanian Society for Radiological Protection and the ICOS MSA Atmosphere Meeting.

#### *Impact on industrial and other user communities*

European climate observation groups and radiological protection groups will both benefit from this project i.e. (i) climate related AMNS (e.g. ICOS), and (ii) the EURDEP and the EANR. By improving the traceability of low-level radon and radon flux measurements this project will support collaboration between these currently independent groups. Such interdisciplinary collaboration could provide new insight and understanding on the links between geology, the atmosphere, and anthropogenic activity and their combined impact. Accurate knowledge of environmental outdoor radon activity concentrations and radon flux is key for improving GHG flux estimates for climate observation and radiological protection. Current climate related AMNS were established for measurements of GHGs in order to support interpretation of the ATM and to better understand GHG levels using long-term observations. Atmospheric radon measurements are carried out at such AMNS and therefore, this project will support European AMNS in performing atmospheric radon and radon flux measurements for a variety of radon tracer applications. The project will do this through its development of new low activity Rn-222 emanation sources, a reference instrument for atmospheric radon measurements and a traceability chain for low radon activity concentration measurements (from 1 Bq m<sup>-3</sup> to 100 Bq m<sup>-3</sup>). All of which will support the comparability of real time atmospheric radon activity concentration data between different measurement sites and over time provide these radon measurements with the required traceability to the SI. This, in turn, will lead to more reliable data for policy and decision makers to use in the combat against climate change. Further to this, the project will provide the necessary measurement infrastructure and use the data that this generates to validate and to improve the RTM to make independent GHG emission estimates that support regular national reporting in the framework of the Paris agreement on climate change.

The project has set-up a Stakeholder Committee in order to support its engagement with industry and other user communities. The Stakeholder Committee currently has 20 members and includes high impact, multi-national stakeholders such as: the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the World Meteorological Organization (WMO), the International Commission on Radiological Protection (ICRP), the International Committee for Radionuclide Metrology (ICRM), the European Radon Association (ERA), the European Radiation Dosimetry Group (EURADOS) and EURAMET's Technical Committee for Ionising Radiation (TC-IR). Further to this, the stakeholder committee also includes standardisation bodies such as the Commission Electrotechnique Internationale (IEC) and the International Organization for Standardization (ISO) as well as representative organisations from individual nations e.g., Australia's Nuclear Science and Technology Organisation (ANTSO), Australia's Climate Science Centre, Oceans & Atmosphere (CSIRO), Germany's Federal Office For Radiation Protection (Bundesamt für Strahlenschutz), Germany's Meteorological Service (Deutscher Wetterdienst), Spain's Center for Energy, Environmental and Technological Research (CIEMAT), Environment and Climate Change Canada, Ireland's Environmental Protection Agency Office of Radiation Protection and Environmental Monitoring, the UK's Society for Radiological Protection (SRP), the UK's Met Office, the National Metrology Institute of South Africa (NMISA), Japan's National Institutes for Quantum and Radiological Science and Technology (QST), Italy's Politecnico di Milano - Department of Energy, Italy's National Research Council/ Biometeorology lab



(IBIMET- CNR), Romania's National Commission for Nuclear Activities Control (CNCAN) and the University of Novi Sad (Serbia).

#### *Impact on the metrology and scientific communities*

The project's data on outdoor radon activity concentration and radon flux measurements can be used to provide key information on atmospheric radon activity concentrations; one of the greatest natural radiological risks. The project's data will be made available online for scientists, policy and decision makers and end users. The project's developments in techniques for measuring low-level environmental radon activity concentration and radon fluxes will also be useful for the metrological community working in this field, for regulatory authorities, civil protection or official measuring bodies, and for manufacturers of radon monitors or dosimeters. In addition, the project will significantly advance radon flux metrology, which is still in an early stage of development. It will do this by providing a calibration infrastructure, including a radon exhalation reference system "exhalation bed" and a transfer standard. The project will then use this capability to harmonise existing radon flux instruments/methods using field-based comparisons. Radon flux measurements carried out over Europe during the project will be used to validate existing European radon flux models and inventories in order to obtain online real-time European radon maps. These radon and radon flux maps can be used for atmospheric studies and for radiological protection such as the identification of RPAs. Furthermore, radon and radon flux maps will enable Europe to meet the World Health Organisation (WHO) and International Atomic Energy Agency (IAEA) requirements for access to validated and reliable radon exposure data according to geographical criteria.

The project has already provided training to the metrology and scientific communities at 4 events; (i) a workshop on New Procedures for Radon Monitoring, (ii) a training course on New procedures, guidelines and methodologies for radon instrument calibration and measurements, (iii) a Scientific Workshop on traceRadon and (iv) a Gap workshop on radiation protection metrology.

Further to this the project is collaborating with organisations in the scientific community including the ERA, EURADOS, the UK's Metrological Office, the Universität Heidelberg, Germany, the University of Novi Sad, Serbia, the Politecnico di Milano, Italy, the University of Codoba, Spain, the Universität Siegen, Germany and ANSTO.

#### *Impact on relevant standards*

So far the project has provided input to HERCA (Heads of the European Radiological Protection Competent Authorities), the Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU), DKE GK 851 Activity measuring devices for radiation protection, ISO/TC 85 Nuclear energy, nuclear technologies, and radiological protection, BIPM and CIPM CCRI (I) (x- and gamma rays, charged particles), EURAMET TC-IR (Ionising Radiation) and EURADOS WG3 Environmental dosimetry.

In addition the project intends to provide future input to: IEC/TC 45 Nuclear Instrumentation SC45B Radiation protection instruments, WG9 Detectors and systems, WG18 Mobile unmanned automated systems, ISO/TC146 Air quality SC1 Stationary source emission, ISO/TC 207 Environmental management SC7 GHG management and related activities, ICRM (Gamma-Ray Spectrometry WG, Alpha-Particle Spectrometry WG and Low Level Measurement Techniques WG) and BIPM CCRI Section II: Measurement of radionuclides.

#### *Longer-term economic, social and environmental impacts*

Climate change and radiological protection both affect humankind and the environment, worldwide. Hence in the combat against both climate change and radiation exposure measurements must be supported by reliable metrology. By addressing a topic (i.e. the measurement of low levels of radon in the environment) that supports both climate observation and global radiological protection this project simultaneously supports the long-term economic, social and environmental work of ICOS, the Integrated Pollution Prevention and Control (IPPC) Directive 2008/1/EC, the IAEA, Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) and WHO. The project's data on low level measurement of radon in the environment will improve ATMs and their ability to estimate GHGs fluxes which in turn will support the EU Emissions Trading System (EU ETS). The EU ETS is a cornerstone of the EU's long-term policy to tackle climate change through a cost-effective reduction of emissions of carbon dioxide (CO<sub>2</sub>) and other GHG in the power, aviation and industrial sectors. The project will thus support Europe in its movement towards a competitive low carbon economy. At the same time, the project will provide the EC (through project partner the JRC) with access to reliable data of outdoor radon activity concentrations, which can be used in combination with soil exhalation flux

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measurements, for dynamic mapping of radon in the environment. By supporting the provision of accurate knowledge of RPA this project will support European radiation protection measures and thus longer-term will help to lower radiation protection costs.

Project start date and duration:		01 June 2020, 36 months
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