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Coordinator: Franz Josef Maringer, BEV-PTP Tel: +43 676 735 4791 E-mail: franz-josef.maringer@boku.ac.at Project website address: www.metroradon.eu		
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RMG: -		

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1 Overview

The aim of this project was to develop reliable techniques and methodologies to enable SI traceable radon activity concentration measurements and calibrations at low radon concentrations. The results of the project have been targeted at the implementation of the European Council Directive 2013/59/EURATOM (EU-BSS), one aim of which is to reduce the risk of lung cancer for European citizens due to high radon concentrations in indoor air. The calibration methods and measurement techniques developed in the project have started to assist EU member states in the establishment of their national radon action plan, which is required under the EU-BSS.

2 Need

Radon is a radioactive, colourless, odourless, and tasteless noble gas, which occurs naturally through geological-based processes. Despite its short half-life of 3.8 days, radon gas from natural sources can accumulate in buildings, particularly in confined or unventilated spaces.

Radon is estimated to cause between 3 % and 14 % of all lung cancer cases, depending on the average radon level in the country. For Europe, this corresponds to around 15 000 to 20 000 people per year dying of lung cancer caused by radon exposure.

Before the commencement of the EU-BSS, the recommended maximum limit for the annual ^{222}Rn activity concentration at workplaces was 1 000 Bq/m^3. With the new EU-BSS, for the first time, an obligatory maximum limit for the national legal limits of the ^{222}Rn activity concentrations in indoor air of workplaces and dwellings was set for all EU member states.

Recently, the EU-BSS have been implemented into national legislation of the EU member states. The maximum reference limit set by the EU-BSS is 300 Bq/m^3. A significant improvement in the metrological infrastructure in Europe for radon calibrations, especially at low activity concentrations (< 300 Bq/m^3) was a prerequisite in order to be able to fulfil these requirements. Therefore, new procedures and traceable radon reference sources for the traceable calibration of radon measurement instruments at low activity concentrations with adequately low uncertainties needed to be developed.

Thoron (^{220}Rn) and its progeny are known to bias the results of radon (^{222}Rn) activity concentration measurements; however, information about this effect was limited. A better understanding of this effect was, therefore, needed together with techniques to reduce the influence of thoron and its progeny on radon measurements and calibrations. Traceability and quality assurance of calibrations of radon monitors and of radon calibration facilities, as well as the development of methods to conduct a large number of traceable and quality assured in-situ and laboratory measurements of radon were also required.

EU member states are bound to consider several aspects when preparing their national radon action plans (Article 103, EU-BSS), which is a strategy for conducting surveys of indoor radon concentrations. To ascertain that, the required level of safety is met for all European citizens, the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe needed to be optimised. Therefore, identification of Radon Priority Areas (RPA) was necessary in order to take appropriate actions for the protection of the public.

To reduce trade barriers and to ensure the mutual recognition of calibration certificates, general guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air have to be established. This have been so far and will be in future facilitated too by the uptake of the project’s results in the standards developing organisations and furthermore, by the end-users.

3 Objectives

The overall objective of this project was to enable the SI traceable monitoring of radon (^{222}Rn) at low radon activity concentrations including calibration and radon mapping, in particular in support of the European Council Directive 2013/59/EURATOM (EU-BSS). The project contributed to the creation of a coordinated metrological infrastructure for radon monitoring in Europe.

The specific objectives of the project were:

1. To develop novel procedures for the traceable calibration of radon (^{222}Rn) measurement instruments at low activity concentrations (100 Bq/m^3 to 300 Bq/m^3) with relative uncertainties ≤ 5 % (k = 1). As part of this, to develop new radioactive reference sources with stable and known radon emanation rates.
2. To investigate and to reduce the influence of thoron (^{220}Rn) and its progeny on radon end-user measurements and radon calibrations.
3. To compare existing radon measurement procedures in different European countries and from the results optimise the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe.
4. To analyse and develop methodologies for the identification of Radon Priority Areas (i.e. areas with high radon concentrations in soil, as defined in the EU-BSS), including the development of the concept of a Radon Hazard Index (RHI), and to investigate the relationship between soil radon exhalation rates and indoor radon concentrations.
5. To validate traceability of European radon calibration facilities, and to publish guidelines and recommendations on calibration and measurement procedures for the determination of radon concentration in air.
6. To facilitate the take up of the technology and measurement infrastructure developed by the project by end users (regulators, radiological protection bodies and policy makers), standards developing organisations (ISO/TC45, CEN/TC351, ISO/TC85, CENELEC/TC 45, IAEA) and the measurement supply chain (accredited laboratories, instrumentation manufacturers).

4 Results

4.1 *Objective 1: Procedures for traceable calibration of radon (^{222}Rn) instruments at low activity concentrations*

To achieve this goal, novel procedures for the traceable calibration of radon (^{222}Rn) measurement instruments at low activity concentrations (100 Bq/m^3 to 300 Bq/m^3) with relative uncertainties ≤ 5 % (k=1) have been successfully developed. This activity range is relevant for regulations defined by the European Council Directive 2013/59/EURATOM for indoor radon concentrations at workplaces (article 54) and dwellings (article 74).

In order to develop procedures for the traceable calibration of ^{222}Rn measurement instruments at low activity concentrations with sufficiently low relative uncertainties, it was necessary to develop new radioactive reference sources with stable and known radon emanation rates. Existing ^{222}Rn emanation sources were re-evaluated concerning their present emanation power and stability. Therefore, problems and alternative manufacturing techniques were investigated in detail. Based on these investigations, prototypes of ^{222}Rn emanation sources, which rely on electrodeposition, drop-implantation and chemisorption of radium (^{226}Ra) were developed and produced.

Additionally, a procedure for preparing thoron (^{220}Rn) emanation sources was developed and tested successfully and thoron emanation sources were produced. Furthermore, a new low-level radon chamber has been designed and built to create long-time and time stable low-level radon atmospheres and has been used to test the feasibility of the new sources.

A long-term stable low-level ^{222}Rn flow through source was designed, manufactured, and installed in two radon chambers. After checking that, a stable ^{222}Rn atmosphere was achieved, and the source was tested at different air pressure, temperature, and humidity values. A combined relative standard uncertainty of about 1 % of the activity measurement was achieved for the flow through source in both reference chambers, which is well below the target uncertainties (≤ 5 %, k = 1).

Moreover, a method for the determination of activity concentration in a steady air flow has been developed and implemented. The feasibility of the method was tested with the developed long-term stable, low-level flow through ^{222}Rn source for activity concentrations between 45 Bq/m^3 and 100 000 Bq/m^3.

A comparison protocol and schedule for the comparison of existing ^{222}Rn gas primary standards at European Metrology Institutes and Designated Laboratories in the low kBq range were developed. ^{222}Rn gas standards were produced and distributed to the participating laboratories. The results of this registered EURAMET supplementary comparison No. 1475, registered at BIPM as EURAMET.RI(II)-S8.Rn-222 were analysed and reported (*Draft B Report* for publication in Metrologia). Also, a comparison protocol and a schedule for the developed ^{220}Rn emanation sources, which were designed within this project, have been developed.

PTB, supported by JRC has developed ^{222}Rn and ^{220}Rn emanation sources including a detector system for the continuous monitoring of the emanated activity of ^{222}Rn relative to the activity of the ^{226}Ra source (and emanated ^{220}Rn relative to activity of ^{228}Th source) traceable to primary standards.

CEA has developed ^{222}Rn and ^{220}Rn emanation sources using polymers. The radon emanation of the sources is not quantitative. These sources were used by CEA and METAS for the development of a method for direct and traceable measurement of the activity concentration of ^{222}Rn and ^{220}Rn in an air flow (Figure 1).

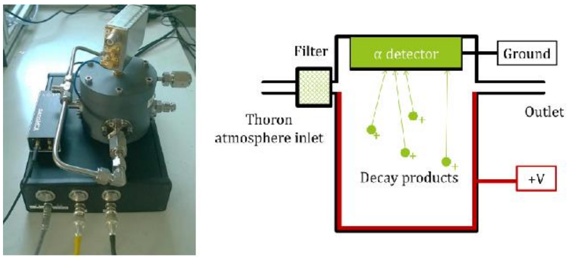

Figure 1: Set-up for measurement of activity concentration in an air flow at CEA.

CMI with support from SUJCHBO has developed a long term stable ^{222}Rn low activity emanation flow-through standard source based on a metering flow controller and dispenser generating a known ^{222}Rn concentration in an air flow.

The quantitative emanations from the two types of sources, developed at PTB and CMI, were compared with the results obtained with the “in air flow measurement system” developed at CEA.

The new ^{222}Rn emanation sources developed and compared within the project together with existing certified reference volumes were installed at BfS, BFKH, IFIN-HH, IRSN, METAS and SUJCHBO reference chambers in order to establish constant and traceable ^{222}Rn activity concentrations.

BfS, IFIN-HH, BFKH, IRSN, METAS and SUJCHBO developed calibration procedures for their reference chambers in the activity concentration range from 100 Bq/m³ to 300 Bq/m³ using ^{222}Rn gas and emanation standards (examples in Figure 1). The result is not one single and unified calibration method but very similar methods using slightly different sizes of calibration chambers and different instrumentation of sensors, flow controllers, etc. The goal to develop procedures for the calibration of radon measurement instruments at low activity concentrations (in the activity concentration range from 100 Bq/m³ to 300 Bq/m³) with sufficiently low relative uncertainties ≤ 5 % was reached. Time stable activity concentrations in this range have been established with the following uncertainties (k=1) at: BfS with 1.0 %, IRSN with 2 %, METAS with 1.5 %, and SUJCHBO with 2 %. Long exposure times of about 40 hours were used to realize uncertainties of the calibration factors for radon measurement instruments close to the uncertainties of the activity concentrations.

The novel procedures for the calibration of radon measurement instruments at low activity concentrations were used to determine the accuracy of commonly used integrated radon measurement instruments (alpha-track detectors, electrets, etc.) and novel detectors.

The ^{222}Rn comparison is registered at EURAMET under the number 1475 and at BIPM as EURAMET.RI(II)-S8.Rn-222. Samples of ^{222}Rn gas provided by the CEA were sent to the seven participants and were measured using various techniques (Figure 2). All results were consistent, within the uncertainties. No outliers were identified by the Grubbs test. Nevertheless, five results were in very good agreement whereas two others were discrepant (about 20 % from the weighted mean). The relative standard deviation between the seven results is 11.4 %.

Figure 2: Examples for reference chambers (from left to right: SUJCHBO, BfS, BFKH, IFIN-HH)

In summary, the project successfully achieved the objective with these findings and results:

- development of new ^{222}Rn and ^{220}Rn radioactive reference sources with stable and known radon emanation capacity,
- establishment of constant and stable ^{222}Rn activity concentrations in radon reference chambers,
- establishment of time stable radon activity concentrations in the relative uncertainty range between 1.0 % and 2.0% (target ≤ 5 %),
- development of calibration procedures for reference chambers in the activity concentration range from 100 Bq/m^3 to 300 Bq/m^3 using ^{222}Rn gas and emanation standards, and
- successful comparison of existing radon gas primary standards at European NMIs/Dis in the few kBq range.

4.2 Objective 2: Reducing the influence of thoron on radon measurements and calibrations

In order to investigate and to reduce the influence of thoron (^{220}Rn) and its progeny on radon end-user measurements and radon calibrations, three reference thoron atmospheres (low, intermediate and high activity concentration) have been established at the reference chamber for calibration of measurement devices with the primary thoron standard system. Before conducting the calibration of the instruments, the homogeneity of the reference atmospheres in the chambers were tested using newly developed methods based on liquid scintillation counting of aerogels and solid-state nuclear track detectors (SSNTD), as well as numerical calculations.

The calibrated measuring instruments are being used to study the influence of thoron on the radon measurement in two new exposure facilities to measure pure and mixed ^{220}Rn and ^{222}Rn atmospheres. Dynamic temperature conditions were also built with static and dynamic temperature conditions within -15 °C to + 60 °C have been created as well as predefined variable concentration profiles. Additionally, measurements have been conducted to quantify the influence of thoron on the radon measurement under field conditions.

A literature review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations provided the necessary data to start an experimental study on diffusion barriers against thoron, radon-thoron cross-interference, radon permeability data and radon transport properties at different temperatures. The findings of this study provided crucial and much needed information on how to limit the influence of thoron on radon measurements. A model for the evaluation of the performance of these polymers as thoron barriers was developed. Additionally, a method was proposed for evaluation and correction of the temperature bias when polymer foils are used as thoron barriers. The method is based on differential measurements with two detectors and has been tested successfully.

Direct experimental determination of the radon transmission through polymer foils in the volume of passive detectors for different temperatures was performed. At the same time a study of the temperature dependence of the response of widely used detectors was made. It was observed that both the radon transmission through polymer foils and the detector response showed significant temperature dependence. However, the trends in both cases are reciprocal. Radon transmission increases with temperature increase, while the detectors' response decreases. At this point, a novel concept, beyond state-of-the-art was proposed. Compensating modules were designed in which detectors are placed, so that the thoron interference on the results is eliminated and both temperature bias and influence of humidity are greatly reduced. The experimental results obtained with various passive detectors demonstrated the feasibility of this approach. This approach is capable to reduce the thoron interference and the temperature dependency of radon measurements significantly.

An experimental study of thoron interference on the results of 16 active radon monitors and 19 passive radon detectors was carried out. It was found that in most of them the thoron interference is greater than 5 % and in some of them it is greater than 20 %. These findings confirm that many radon monitors/detectors have a problem with thoron interference and justify the recommendation made: The manufacturers of radon monitors/detectors should perform testing for thoron cross-interference of their radon instruments and should include this information in the specifications of the instrument.

An analytical review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations was prepared. The properties of different filters/foils/ membranes that could potentially serve as selective thoron barriers and assessment of their radon permeability were evaluated. The radon transport properties of different polymers were studied and selective anti-thoron barriers were characterised. The major findings were:

- Diffusion barriers introduce an additional inertia in the response of active detectors.
- Gaps/pin holes chambers do not isolate the detection volume from the humidity. For such chambers the thoron interference may vary in different detectors of one and the same kind.
- Polymer foils introduce a temperature bias in the radon results.

The testing of radon monitors for thoron sensitivity should include a reference (secondary) thoron monitor that is calibrated with, or traceable to a primary thoron measuring system. A stable reference ^{220}Rn atmosphere to be created in an exposure chamber by flushing air with constant flow-rate consecutively through the ^{220}Rn source and the exposure chamber is recommended. To ensure thoron homogeneity, fan(s) should operate in the exposure chamber during exposure. The reference instrument has to be placed inside the exposure chamber throughout the exposure. Thoron homogeneity in the chamber should be checked during exposure or in another experiment under the same exposure conditions. Air pressure, temperature, humidity and air velocities (if applicable) during the tests should be recorded;

The calibration exercise which was carried out at IRSN with seven thoron measuring instruments (four AlphaGUARD and three RAD 7), showed that the ratios between the ^{220}Rn activity concentrations measured by the instrument and the reference activity concentration measured by the IRSN reference instrument is close to 1 for the four AlphaGUARD and around 0.6 for the three RAD7. The calibration factor of the AlphaGUARD was found to change with the thoron activity concentration.

The following radon monitors were tested for thoron cross-interference (*CI*): AlphaE, *AlphaGUARD* PQ2000 Pro, *AlphaGUARD* PQ2000, *AlphaGUARD* 2000 RnTh Pro, RadonEye +2, Corentium Home, Airthings Wave, Airthings Wave Plus, Corentium Pro, TSR3 – Fast and slow mode and TSR4M– Fast and slow mode, DoseMan – Fast and slow mode. All tested active thoron monitors except for RadonEye, TSR4M and DoseMan comply with the IEC 61577-2 standard requirement for *CI* < 20 %.

A minimum of three days test with a high thoron activity concentration (around 10 kBq.m^{-3} or more) is recommended to determine an accurate final *CI* of the active monitors, instead of the 4 hours at 1000 Bq.m^{-3} required in the IEC 61577-2 standard.

These passive radon detectors were tested: RSKS and Raduet by Radosys, Radtrak[2], Rapidos and Duotrak by Radonova, DPR2, DRF and Kodalpha by Algade, Easyrad by Pearl, 1 detector by STUK, 1 detector by AlphaRadon, 1 detector by Eurofins Radon Testing, 3 types of in-house detectors (1 designed by SUBG and 2 by ISS, Italy) and two types of detectors based on alpha track detection in DVD (CD). Except for EasyRAD and Kodalpha, the cross interference of thoron on the radon signal of all the instruments comply with the IEC 61577-2 standard requirement for *CI* < 20 %. No single construction of the studied radon monitors based on diffusion chambers could be recommended over the others. A low cross interference could be achieved with diffusion chambers with different constructions (in terms of volume, dimensions and inner compartments). In general, the chambers with smaller width of the air gaps (of the order of tenths of millimetre) show lower thoron sensitivity.

The manufacturers of radon monitors should perform cross-interference testing for their radon instruments and should include this information in the specifications of the instrument. For instruments for which fast reaction to rapidly changing concentrations is required a spectrometric discrimination between radon and thoron is probably the best option. For active monitors with active sampling, counting in two different time intervals can also be used. With a proper data processing such instruments can measure radon and thoron separately still keeping capacity for fast reaction to rapidly changing concentrations.

If spectral discrimination is not used but the instruments are expected to have a fast reaction, a proper approach is to incorporate a delay line either within the instrument design or as supplementary module. The parameters of such delay line (buffer volume and flow-rate) should correspond to the instrument's technical characteristics.

For continuous radon monitors which do not perform spectral discrimination, but record the temperature and for which a fast reaction is not necessarily required, the thoron influence may be reduced by packing the monitors (or their sensitive volume) in polyethylene foils.

The temperature induced bias in the radon readings can be corrected from the temperature record and using the known dependence of the radon permeability of the foil on the temperature.

For instruments for which fast reaction to rapidly changing concentrations is not required, and which work in a diffusion mode, additional diffusion barrier can be used.

For passive detectors diffusion barriers might be considered in the design and tested in the prototypes. If instruments are scheduled to work at high humidity, diffusion barriers based on polyethylene foils of low density polyethylene are recommended.

For detectors in which the usage of polyethylene packing is planned to reduce the thoron interference, it is recommended to calibrate the packed detectors and to perform the calibration at temperatures which is close to the expected mean temperature during the measurement. This is to comply with the general principle that the conditions during the calibration and the measurement should be as close as possible. Additionally, a possible temperature bias may be introduced. Three methods for simple and effective handling and taking account of this bias are proposed.

Recommendations on the construction of radon monitors that are not sensitive to thoron including the technical concepts/solutions aimed at reducing thoron-related bias in the radon signal in existing monitors were developed. Reducing the thoron interference is related to finding the best compromise between anti-thoron protection ensured and worsening of some quality of the radon measurements.

The temperature dependence of response to radon/thoron of active and passive radon/thoron monitors and radon detectors was studied in detail. For detectors that have response decreasing with the increase of the temperature, the temperature bias can be compensated if the anti-thoron polymer packing is designed as a compensation module. Different approaches to achieve the best anti-thoron protection of active radon monitors and passive radon detectors with no or minimum influence on the quality of radon response are proposed. In particular, an unexpected finding led to innovation: the possibility to reduce or compensate the thoron interference + temperature related bias + humidity influence for many widely used detectors – potentially for all detectors which radon response decreases with the increase of the temperature; **patent application submitted:** Pressyanov, D., 2019. Bulg. Pat. Appl. Reg. Nr. 112897, priority: 19.03.2019, WIPO Appl. Reg. Nr. PCT/BG2020/000003, see Figure 3.

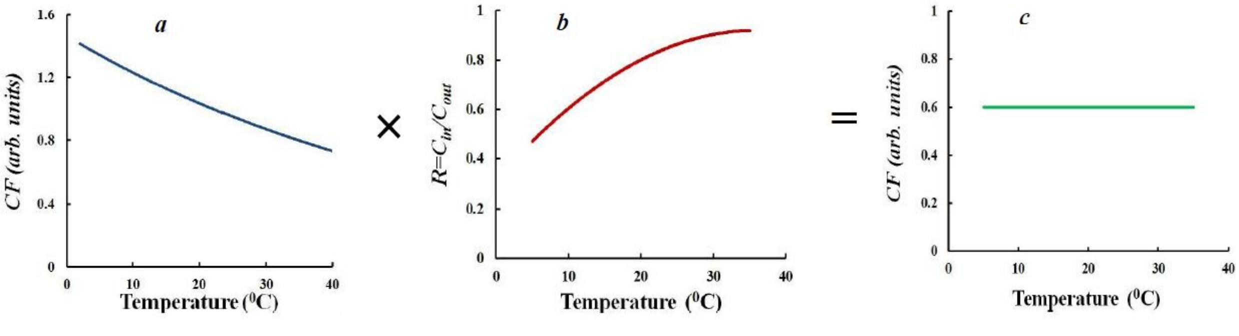

Figure 3: The concept of the compensation module design (patent pending): The temperature dependence of the response of many radon detectors (a) and that introduced by polymer anti-thoron barriers (b) are

reciprocal. This can be used to reduce/eliminate the temperature dependence (c), the thoron interference and the humidity influence

Summarizing, all planned research to achieve the objective was successfully completed with these findings and results:

- assessment of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations,
- development of novel procedures for checking the sensitivity of radon measurements to thoron,
- improvement on the construction of radon monitors that are not sensitive to thoron,
- a new technical approach aimed at reducing thoron-related bias in the radon signal in existing radon monitors, and
- development of a novel method to compensate the thoron interference, the temperature related bias and the influence of air humidity for new radon monitors.

4.3 Objective 3: Comparison of existing radon measurement procedures in different European countries and optimisation of the consistency of indoor radon measurements and soil radon exhalation rate measurements

To achieve this objective, meta-information on radon surveys performed and existing radon databases in European countries were collected and analysed, to evaluate if the data and methodologies are comparable and how they could be harmonized in case of methodical inconsistency.

In order to compare existing radon measurement procedures in different European countries and the results on consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe, a review and comparison of existing radon measurement procedures in different European countries had been conducted. The goal was to use the results of the study to optimise the consistency of indoor radon measurements and soil radon exhalation rate measurements across Europe. An extensive literature review on existing indoor radon survey and geogenic radon surveys in Europe had been conducted. Two questionnaires, one on indoor radon surveys, and one on geogenic radon surveys conducted in Europe were prepared and sent to stakeholders. The questionnaires collected missing information from the literature and obtained information on how the countries intend to transpose the EU-BSS directive into national law.

Although solid state nuclear track detectors were used in 82 % of the studies, the analysis of the results of the questionnaire on indoor radon surveys showed that the design of surveys in Europe are not comparable. According to the questionnaire, 44 % of surveys were not performed during the whole year, and seasonal corrections were not applied in all of them. Therefore, another important aspect highlighted by the questionnaire was in the harmonisation to apply seasonal corrections. This is important because seasonal variation of radon could differ within a country from region to region due to different factors, such as climate, living habits, building construction, etc. Also, the non-negligible effect of reported indoor radon concentrations could be due to thoron influence. Although a large percentage of participants knew about the interference of thoron, they did not correct or check for thoron presence.

A study of the representativeness of indoor radon measurement over different measurement periods (durations and seasons) was performed and a literature review on the applicability of the CD/DVD method for indoor radon surveys was conducted, concluding that CDs and DVDs can be used as ^{222}Rn detectors in radon surveys. Existing ISO standards EN ISO 11665-7 and ISO 11665-11, on the methodology of the radon exhalation measurement and of radon concentration in soil gas measurement, were reviewed in order to assess whether and how appropriate methodologies are for use in the present project.

In addition, an intercomparison based on variable indoor radon conditions along with ambient parameters that are subjected to change has been organised. The intercomparison site was the Laboratory of Natural Radiation located at the facilities of a former uranium mine near Salamanca, Spain. Measurements were conducted together with 20 participants. The goal of this activity was to compare the response of instruments traceably calibrated in reference radon chambers to changing conditions of radon gas concentrations. Indoor radon gas measurements for passive and continuous monitoring devices were conducted at around 200 - 300 kBq·m^{-3}·h and 2000 kBq·m^{-3}·h or higher. Radon exhalation from soil and radon concentration in soil gas measurements were conducted in the outdoor area of the laboratory. More than 80 % of the results for radon activity concentration in air were within the interval defined by the reference value and the standard deviation.

Conclusions from both literature overview and questionnaires performed on indoor radon surveys in Europe are that the overall design of surveys is quite diverse and that it is difficult to find two completely same approaches to a survey. By looking at three main aspects of the survey it can be summarised that: a) designs of surveys performed in Europe are not comparable; b) measurement methods are comparable between surveys; c) data management, statistical analysis and mapping are for some aspects comparable for others not.

The most critical part of the surveys was estimation of representativeness. An important aspect in harmonisation is to apply seasonal corrections. Furthermore, a non-negligible effect of reported indoor radon concentrations could be due to thoron influence. VINS together with AGES, BfS and JRC compiled and published a JRC technical report (*7*) and an open access journal article (*10*).

Relatively much information is available on the status of geogenic radon surveys in European countries, as well as about methodology. On the other hand, not many countries have performed geogenic radon surveys. Therefore, European coverage is poor. Again, on the other hand, surveys and data sets about quantities, which can serve as predictors (U concentration) or proxies (Ambient Dose Equivalent Rate) of the Geogenic Radon Potential, are available in many countries. So far, the data have been exploited for generating European wide geogenic radon map only in experimental trials. Current work seems more focused on developing a Geogenic Radon Hazard Index (GRHI) which relies on Europe wide available data bases (such as for geology and geochemistry), rather than on assembling regional un-harmonized datasets. Regarding methodical harmonization of geogenic quantities, a few issues have been identified. The problems can be solved, but in some cases require further experiments and partly development of procedures for harmonization.

The method employs CDs/DVDs as radon detectors (from the available stock stored indoors) and provides long term (> 1 year) retrospective indoor radon concentration results.

The main directions of usage of the CD/DVD method can be summarized as:

- retrospective dosimetry of radon and thoron (incl. for the purposes of radon mapping);
- identification of radon prone areas and buildings with radon problems;
- retrospective evaluation of the effect of building retrofits on radon levels;
- measurements in working places (incl. mines).

An intercomparison exercise has been carried out by the University of Cantabria with the support of JRC in the Laboratory of Natural Radiation (LNR) located at the facilities of the former uranium mine managed by the Spanish National Uranium Company ENUSA (Ciudad Rodrigo, Salamanca, Spain). Twenty participants from 13 countries took part in the intercomparison "radon in air", three in the "radon exhalation from soil" and five in the "radon in soil" exercise. Over 80 % of the results for radon-in-air exposure are within the interval defined by the reference value and the standard deviation. Results of the work done by UC together with JRC are published in the report *"Intercomparison of indoor radon and geogenic radon measurements under field conditions"*.

Collection of radon data is the first step in the development of any kind of radon maps. Several kinds of radon maps can be created that display for example the simple arithmetic mean, value (%) above reference level, radon potential, radon priority map, Radon Hazard Index map. Therefore, the input radon data strongly influences the output map. Radon data could be: indoor radon, soil gas radon, geogenic radon data (uranium concentration in soil and rock, terrestrial gamma dose rate, soil permeability, radon in water).

Different surveys and/or data can be harmonized in two ways as displayed in Figure 4. Firstly, all surveys are performed methodically identically. Then homogeneity is ensured by construction - this approach is called *bottom-up* and viable only if surveys are planned jointly from scratch. Realistically, surveys are conducted independently (different period, different purpose, different methods, etc.). To make surveys comparable, the results need to be normalised to a common standard using models based on physical and statistical knowledge of the used procedures in the survey. This is called *top-down* or posterior harmonization.

Our conclusion is that harmonization of radon data is partly possible and projects like the present project and intercomparison exercises are needed to study the comparability of radon data and develop procedures for harmonization. However, it is not an easy task and further studies of comparability and harmonization should be performed in the future.

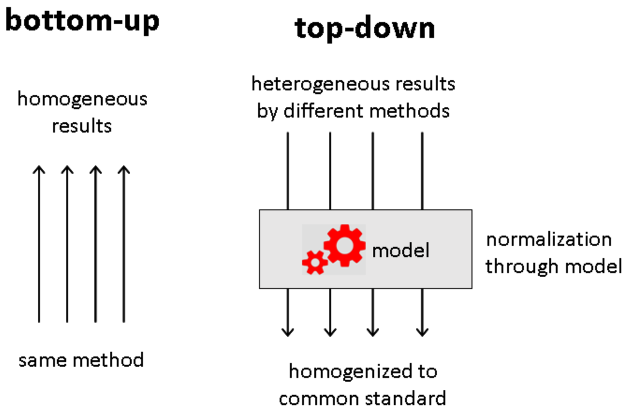

Figure 4: Bottom-up and top-down harmonization

To summarise, the project successfully achieved the objective by

- the successful evaluation of indoor and geogenic radon surveys in Europe, including their strategies, the methodologies employed, inconsistencies in the results, and potential methodologies to harmonise data and reduce inconsistencies,
- the development of options for harmonization of indoor and geogenic radon data,
- the development of a novel method of retrospective indoor radon measurements using CDs/DVDs, and
- on-site comparison of indoor radon measurements and geogenic radon measurements under field conditions together with guidelines/recommendations to assist the implementation of the EU BSS.

4.4 Objective 4: Methodologies for the identification of Radon Priority Areas and investigation of the relationship between soil radon exhalation rates and indoor radon concentrations

To achieve this target, methodologies for the identification of Radon Priority Areas have been analysed and developed, to investigate the relationships between indoor radon concentrations and quantities related to geogenic radon, including soil exhalation and to develop the concept of a “Geogenic Radon Hazard Index” (GRHI) as a tool to help identify Radon Priority Areas.

For the development of harmonised and comparable methodologies for the identification of Radon Priority Areas (RPA) it was necessary to investigate the relationship between soil radon exhalation rates and indoor radon concentrations. An extensive literature review was conducted, reviewing and evaluating the concepts and definitions of Radon Priority Areas proposed or already implemented. Methods developed in Europe to assess the “soil radon potential” which can be used for radon mapping and priority area definition were reviewed. Different approaches were selected to be tested and compared based on a case study from a region in France.

The concepts of classification uncertainty of RPAs were reviewed. Given its economic and political impact, QA for RPA estimation is crucial. In particular, this concerns the uncertainty of RPA estimates. Sources of RPA classification uncertainty were identified and ways to estimate and to quantify RPA uncertainty were proposed.

To study the various approaches for radon mapping, a mapping exercise was conducted. Exercise data for testing existing mapping methods used in various countries has been compiled and edited and is under assessment by project partners. It consists of two data sets that comprise the region of Cantabria (Spain) and six municipalities in Austria. They include indoor radon measurements, building characteristics of measured dwellings, soil air radon activity concentrations, soil permeability estimations, activity concentrations of soil samples, ambient dose rates and maps of geogenic parameters. The partners have applied their typically used mapping method to characterise radon risk. The findings were presented at the 14th International Workshop on the Geological Aspects of Radon Risk Mapping. Finally, there was a special focus on differences between radon mapping and definition of RPAs across boundaries. To study this aspect, the borders between Portugal and Spain, Spain and France, Switzerland and France, and Belgium and France were examined.

To develop a complementary approach for mapping RPAs, data on geology, geochemistry, gamma radiation, indoor radon measurements and associated buildings characteristics at a national and local scale in regions with very high radon levels in Spain, Portugal and France were collected and validated. Statistical tools were applied at both scales to identify sectors with the highest indoor radon concentrations. The identified areas have been examined.

In addition, and knowing that CD/DVDs can be used as ^{222}Rn detectors, a long-term exposure experiment of CDs and DVDs was conducted. The results had been used to assess the precision, applicability and uncertainties involved when using CDs/DVDs for retrospective radon measurements and for the identification of RPAs.

CDs/DVDs that were exposed to a radon atmosphere in the long-term exposure experiment have been analysed. Radon concentrations in a range of < 10 Bq/m³ to 147 300 Bq/m³ were found. Measurement of ^{222}Rn activity concentrations of 100 Bq/m³ to 300 Bq/m³ and even lower concentrations posed no problem. While aimed at radon exhalation and radon mitigation measurements, the scope of potential application for the CD/DVD-method appears to be much wider. It was found that the overall good correspondence between CDs/DVDs and the reference measurements can be further improved if the correlation between radon concentrations and temperature during exposure is taken into account. A modified method, based on CDs/DVDs, with improved sensitivity, was studied. This modified method can be used for measurement of low ^{222}Rn concentrations, as well as for measurements in soil gas and for measurements determining the radon exhalation from soil surface.

Conceptual and theoretical work about Radon Priority Areas is well advanced. Their generic definition according BSS is an area in which a significant proportion of indoor radon concentrations exceeds a reference level of 300 Bq/m³, at most. An important result is the comparison of residential buildings and workplaces regarding their radon characteristics. These were found to be different, in general. This is relevant, because RPAs are mostly estimated based on data of indoor radon concentration in dwellings, but legal consequences as stated in the BSS largely pertain to workplaces. For evaluating the cross-usage of concepts, different mapping methods were compared. Applying a mapping method using data sets, which were not designed for the specific requirements of the mapping method, is challenging. Different mapping methods often, but not always, deliver the same results in RPA classification, depending on the definition of RPAs. Definition of thresholds is a key factor in delineation of RPAs.

Different concepts of Radon Potential (RP) were reviewed in this research**.** One may distinguish between “top-down” approaches, whose initial variable is observed indoor radon concentration and “bottom-up” approach, which starts from control quantities. The GRP (Geogenic Radon Potential) is a particular kind of RP; it is defined physically from geogenic quantities which control radon generation and transport in the ground. Regarding mapping, the rationale of the RP in general and the GRP in particular is that the geographic pattern of IRC (Indoor Radon Concentration) mainly reflects the one of its geogenic controls.

The often poor correlations between IRC and geogenic quantities remain a challenge for further studies. Regionally developed models may not be applicable beyond the region in which they have been developed. First European-scale studies about that problem have been initiated only recently. An open question is the one of anthropogenically modified geogenic factors. This is important because most people live in altered built-up environments. It is recommended that the topic is addressed thoroughly in future investigations.

Comparisons of some examples of borders in Europe show different mapping methods and different mapping results. Main sources of inconsistency are underling data and RPA definition, as well as estimation methodology. Further studies are still necessary in European Countries to provide the technical explanations of consistency or inconsistency between radon maps across borders, as a condition to credible communication to the public.

Quality assurance of RPA delineation is often ignored. As results of an estimation procedure, RPAs are uncertain, in the sense of misclassification. This uncertainty cannot be avoided by nature of statistics, but it should be quantified. Several methods have already been developed to map RPAs. A complementary approach was tested to focus on the identification of areas that could be concerned by a significant proportion of dwellings with very high IRC of several thousands of Bq/m³. The results provide useful elements to target areas where more precise studies are needed.

As a finding of the work, the GRHI can be understood as a generalized complement and extension to the GRP to characterize susceptibility of a location to geogenic radon, as one important control of indoor radon. The GRHI is more flexible and can deal with data which usual GRP definitions cannot handle. Its main application is thought to be large-scale mapping in contrast to small-scale characterization, whose objective is supporting legislative and administrative implementation of the EU BSS (Figure 5).

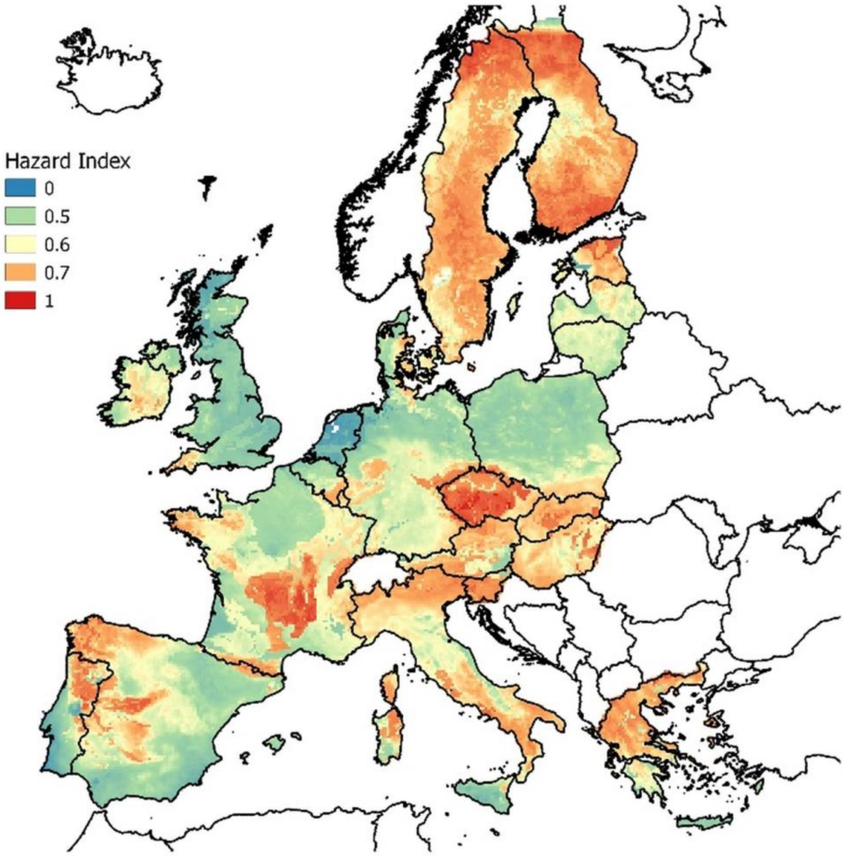

Figure 5: A tentative European GRHI (Geogenic Radon Hazard Index) map *(13)*, generated by machine learning based on numerous geogenic quantities

When finalising this objective, some important questions were identified for future investigation:

- The geogenic radon potential (GRP) is composed of radon source and radon transport. Their values acquired by grab sampling in the field reflect the condition at a certain time, which may be temporally variable to different extent. Solutions to be evaluated and compared more thoroughly include resorting to long-term measurements or fostering modelling based on temporally stable quantities.
- Radon quantities, notably indoor radon concentration (IRC) and GRP, tend to spatially and temporally extreme behaviour, resulting in the occurrence of local anomalies. The question how to estimate and map anomalies adequately will remain an issue due to its statistical complication.
- Residential buildings and workplaces and public buildings have different physical characteristics, in general, in particular concerning their “response” to geogenic radon. It turned out that the matter is complex and should be investigated further due to its practical importance.

Nevertheless, the aim of the objective have been successfully achieved by

- the evaluation of the concepts for the definitions of Radon Priority Areas (RPA),
- the analysis of the relationship between indoor radon concentration and geogenic radon,
- the harmonisation of RPAs across borders,
- a new development in estimation of RPAs,
- a guideline on the definition, estimation and uncertainty of RPA, and
- the establishment of a Geogenic Radon Hazard Index (GRHI) including an RHI map of Europe showing areas with high geogenic radon potential and conclusions on the relationships and correlation between indoor radon concentration and quantities related to geogenic radon.

4.5 Objective 5: Validation of the traceability of European radon calibration facilities, and guidelines on calibration and measurement procedures for the determination of radon concentration in air

To achieve this goal, the quality and metrological traceability of existing European radon calibration facilities over the ranges from 100 Bq/m³ to 300 Bq/m³ and 300 Bq/m³ to 10 000 Bq/m³ has been improved and validated successfully. A significant improvement in the metrological infrastructure in Europe in the field of radon calibrations at low activity concentrations have been achieved within this project in order to be able to fulfil the EU-BSS requirements. International and European comparisons (e.g. CCRI(II), EURAMET) provide confidence in the capability of European radon calibration facilities in the field of radon activity concentration monitoring in air.

In order to validate the traceability of European radon calibration facilities, a survey of European calibration facilities, institutes with primary and secondary radon calibration standards, and selected end-users had been conducted by BfS, BEV PTP, BFKH, CMI, CLOR, IFIN-HH, IRSN, JRC, STUK, SUJCHBO, and UC. The results were evaluated, and 15 facilities were selected for validation and traceability exercises using an AlphaGUARD radon monitor as a reference instrument. The comparison exercise validated the traceability, performance and precision of European radon calibration facilities in the range of 300 Bq/m³ to 10 000 Bq/m³ at three calibration points: 400 Bq/m³, 1 000 Bq/m³ and 6 000 Bq/m³. Following the development of constant radon activity concentrations in reference chambers and calibration procedures at low activity concentrations, an intercomparison exercise to validate the traceability of the secondary standards used by the European radon calibration facilities was conducted. A reference instrument was circulated through the participants who calibrated this using their usual calibration procedures.

Furthermore, the schedule and comparison protocol for the exercise to validate the traceability of European radon calibration facilities at stable radon atmospheres in the range from 100 Bq/m³ to 300 Bq/m³ was developed by CMI, BEV PTP, BFKH, IFIN-HH, STUK, BfS, CLOR, IRSN, JRC, SUJCHBO, and UC.

The first task was to mapping the relevant European radon calibration facilities. The main objective was to be able to serve European radon calibration facilities in a better way by identifying needs and work on solutions to that. For this purpose a questionnaire was created. The questionnaire asked for the performance of the laboratory and the traceability of the quantity radon activity concentration and it was divided into two parts: information about laboratory and laboratory performance and traceability. About 25 relevant European radon calibration facilities were asked to complete the questionnaire and 18 answers have been received. The following information was evaluated based on the results from the questionnaires: legal forms of laboratories, accreditation status, radon activity concentration measuring instruments which represent the highest metrological level, working standards, methods of calibration, calibration ranges, the uncertainties of calibration capabilities, size of radon chamber, climatic conditions, some additional parameters which can be monitored or controlled and the number of calibrations. This survey was comprehensive. The highest metrological level radon concentration measuring instruments are identified.

In the framework of the present project, two interlaboratory comparisons were initiated in order to validate the traceability of European radon calibration facilities and to demonstrate their performance in calibrating radon measuring instruments in the ranges from 300 Bq/m³ to 10 000 Bq/m³ organised by BfS, and 100 Bq/m³ to 300 Bq/m³ organised by SUJCHBO. Calibration services from different EU member states, which preferably represent the respective national reference for the quantity radon activity concentration in air, were encouraged to participate in the comparison. The objective of the interlaboratory comparisons was to determine the degree of agreement in the realization of the activity concentration of ^{222}Rn in air in the facilities of the participating laboratories and to create links between selected laboratories. The traceability chains of the quantity radon activity concentration in Europe were outlined.

In total 15 calibration facilities from 12 different countries of the European Union and one from Montenegro (MNE) participated in the interlaboratory comparison in the range from 300 Bq.m^{-3} to 10 000 Bq.m^{-3}. The interlaboratory comparison was conducted by the German Federal Office for Radiation Protection BfS and took place in the period from March 2018 to February 2020.

Additionally, the verification of secondary standards of European calibration laboratories was performed by SÚJCHBO, v.v.i. Kamenna at stable ^{222}Rn atmospheres in the range from 100 Bq.m^{-3} to 300 Bq.m^{-3} from October 2019 to April 2020. 8 European laboratories participated in the intercomparison of secondary standards and 9 measuring devices were calibrated.

The considerable number of participants from various European countries allowed a representative validation of the performance and quality in the calibration of radon measuring devices. Figure 6 shows the countries where laboratories participated in the comparisons.

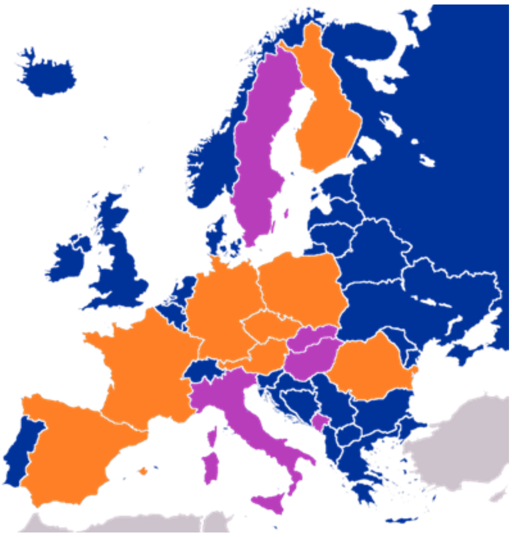

Figure 6: Calibration facilities participating in interlaboratory comparisons (violet – laboratory participating only in comparison performed by BfS, orange – laboratory participating in comparisons at BfS and SUJCHBO)

The traceability and quality assurance of calibrations of radon monitors and of radon calibration facilities itself as well as the development of methods to conduct a large number of traceable and quality assured in-situ and laboratory measurements of radon has been concerned within the present project. The electronic instruments of the type AlphaGUARD were selected as the comparison devices. The devices were compared to each participant's secondary standard, which are used for the calibration of the end-user devices.

The European radon calibration facilities trace back their primary quantities to the national metrological institutes PTB (Germany), CEA (France) and NIST (USA). The statistical analysis did not show any evidence that the different traceability chains influence the result of the intercomparison or the performance of the calibration facilities in Europe.

The interlaboratory comparison of secondary standards of European radon calibration facilities for radon calibration is a powerful tool to detect discrepancies in traceability and to ensure the quality of radon measurements in Europe. It is strongly recommended to carry out the calibrations and verifications regularly.

In summary, the project successfully achieved the objective with these results and findings:

- identification, evaluation and selection of European radon calibration facilities for validation of traceability,
- validation of the traceability, performance and precision of primary and secondary radon calibration facilities in Europe, and
- compilation of a guideline and recommendations on calibration and measurement procedures for the determination of radon concentration in air.

5 Impact

The project helped to establish a basic European metrological infrastructure for radon measurements enabling sound monitoring of radon and radon protection in Europe. Over the course of the project, about 300 European stakeholders and interested parties have been reached through the MetroRADON webpage, the newsletter, participation and organisation of workshops, presentations at scientific conferences and publications of peer-reviewed articles in scientific journals. The stakeholders consisted of manufacturers of radon monitoring equipment, companies offering radon measurements, calibration facilities, national authorities charged with the implementation of the EU Basic Safety Standards into national law, international bodies, etc. The stakeholders have been regularly updated with the project results.

The topicality of radon and the dissemination of the project's result created a strong interest in collaboration, resulting in nine academic and public organisations approaching the project to become involved as collaborators.

The project's results have been published in 15 peer reviewed scientific articles (six more papers have been drafted), used in seven technical workshops and disseminated in 70 presentations at European, international, and national conferences. The findings have been published in guidelines and best practice guides.

Impact on industrial and other user communities

This project has helped to establish a basic infrastructure for low radon activity concentration measurements and thus assist with the generation of Radon Hazard Index (RHI) maps so that metrologically sound measurements are possible to support the implementation of the EU-BSS and sound decision making for radiological protection and ensuring public health.

The impact of the project was firstly targeted to the process of the transposition of the EU-BSS, but beyond this, the project has enabled uptake and exploitation of its results and experience by all stakeholders concerned with radon, including the scientific community. As a consequence, field measurements in Europe have a higher precision, allowing more appropriate precautions and counter measures against public exposure to radon. In addition, harmonisation and standardisation has allowed the comparison and merging of different existing radon data sets on a European scale. The credibility of reported radon data has improved due to harmonised measurement procedures and has led to more consistent results. The relevant authorities and end users in EU member states have the possibility to perform accurate radon monitoring due to improved calibration methods, summarised in best practice guides and recommendations on metrologically sound calibration and measurement procedures for the determination of radon concentration in air. A guideline, prepared by JRC together with BEV PTP, PTB, AGES, BfS, summarizes the output of the project considering links between metrological standardization and radon mapping: *"Guideline on the constituents of the chain "from primary standards to radon maps" (traceability) and the links between them."*

A large number of recommendations and guidelines, such as recommendations on the construction of radon monitors that are not sensitive to thoron have been developed and technical concepts/solutions aimed at reducing thoron-related bias to the radon signal in existing monitors have been published, thus leading to improved radon measurements and the opportunity for improved instrumentation.

An industry interest group, established in the project, provided a specialized platform to disseminate the project's results to stakeholders and pursue discussion and uptake. In June 2019, an Industry Interest Group (IIG) Meeting was achieved, and 20 representatives of highly relevant industry sectors attended. At this meeting, the demand of industry on radon monitoring has been discussed based on the presented aims and interim results of the present project.

Impact on the metrology and scientific communities

Stakeholders, regulators, and end-users of radon measurement methods profited from the dissemination of the results of this project. For this purpose, an industry interest group, group of national authorities and a group of European and international bodies and associations dealing with radon issues has been formed and used as channels to disseminate relevant information to different stakeholders. To inform all stakeholders of the on-going research conducted in MetroRADON, a project newsletter has been sent to approx. 300 recipients. The newsletter was facilitating the uptake of the procedures developed in the project by the end-users. To obtain input from stakeholders, questionnaires had been sent to competent institutions in Europe to obtain information about the primary and secondary standards used, the reference atmospheres, procedures and traceability chains, as well as on indoor radon surveys and geogenic radon surveys. The results of the surveys have been used for recommendations and guidelines that have been discussed in the scientific and metrological communities and published for end-users.

Ensuring traceability for the most commonly used European radon calibration facilities, especially at low radon activity concentrations, has significantly reduced calibration uncertainties. Impact was expected in the short term as various stakeholders have requested calibration services for instruments measuring the radon concentration in air. The metrological facilities for radon measurement instruments and detectors calibration improved and updated by the outcome of this project are summarized by CMI in the *Report "Network of European calibration laboratories for radon concentration in air measurement"*.

The consideration of thoron in the newly developed calibration and measurement procedures triggered the development of new and improved methods in the scientific community and, therefore, the end-user community.

The project was facilitating the opportunity to conduct supplementary comparisons (EURAMET 1475 and BIPM/CCRI(II) as EURAMET.RI(II)-S8.Rn-222) for low radon activity concentrations. Through this, national metrology institutes (NMIs) and designated institutes (DIs) have had the ability to submit or expand their CMC entries for radon measurements. This enhanced the application of the mutual recognition arrangement (MRA) in that specific field of radionuclide metrology.

Impact on relevant standards

The project's outputs and data benefited European and international standards on radon monitoring and guidelines on radiological protection, construction products, radiation instrumentation and nuclear data. In particular, input has been given to:

- IEC TR 61577-5 "Radiation protection measurement instrumentation – Radon and radon decay product measuring instruments – Part 5: General properties of radon and radon decay products and their measurement methods" (IEC TC 45/SC 45B Radiation protection instrumentation – WG 10: Radon and radon daughter measuring instruments),
- "Measurement of radioactivity in the environment – Air: radon-222 – QA/QC for calibration facilities" (ISO/TC 85/SC 2 Radiological protection – WG 17 Radioactivity measurements), and
- Transformation of the CEN Technical Report TR 17113:2017 "Construction products: Assessment of release of dangerous substances — Radiation from construction products — Dose assessment of emitted gamma radiation" into a CEN Technical Standard (EC Action on CEN TC 351 WG 3 Construction products: Assessment of release of dangerous substances, Radiation from construction products).
- Input to a EURAMET standard comparison report: EURAMET comparisons for ^{222}Rn, not involving any Radon chambers, had been carried out about 15 years ago. The project comparison for ^{222}Rn and ^{220}Rn, done in WP1, has been successfully established as EURAMET TC IR comparison.
- Input to CENELEC/TC 45 Technical Report CLC/FprTR 62461 Radiation protection instrumentation - Determination of uncertainty in measurement - CLC/FprTR 62461:2019.
- Input to the revised IAEA Specific Safety Guide No. SSG-32 'Protection of the Public against Exposure Indoors due to Radon and Other Natural Sources of Radiation', jointly sponsored by the IAEA, WHO. BEV-PTP and other project partners support the IAEA radon section.
- Input to ICRM guideline on radon metrology. BEV-PTP and other project partners presented the progress and the interim results of the project in the ICRM Low Level Radionuclide Metrology

Techniques Working Group at the 22nd International Conference on Radionuclide Metrology and its Applications, at the University of Salamanca (May 2019).

This input has improved the traceability requirements of radon and radon daughter measuring instruments in European and international standards. The aims of which are the reduction of uncertainties at instrument calibrations and harmonization of calibration methods at low radon activity concentrations (< 300 Bq/m³ ^{222}Rn). Input has also been given to IEC standard 61577 at the end of the project.

Longer-term economic, social, and environmental impacts

In Europe thousands of lung cancer cases annually are attributed to indoor radon progeny exposure. This project contributed to improving public health through more reliable radon measurements as a basis for effective radon mitigation and prevention of radon progeny induced lung cancer in Europe, leading to improved public health and reduced healthcare costs.

6 List of publications

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7 Contact details

Franz Josef Maringer (Coordinator): franz-josef.maringer@boku.ac.at