

# 16ENV10 MetroRADON

Deliverable D8

Guideline and recommendations on calibration and measurement procedures for the determination of radon concentration in air

Czech Metrology Institute (CMI)

Due date: April 2020 Submission: November 2020



The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

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## 1 Scope

The guideline lays down common principles and recommendations that are necessary from a metrological point of view in order to promote further harmonization of protection against elevated exposures to radon at the workplace and at home. The guideline focuses on the metrological requirements for determining the activity concentration of radon-222 in ambient air.

The proposed organizational and technical measures and precautions are aimed at promoting the quality and comparability of the measurements and thus creating confidence in the results and acceptance of the derived protection measures. The harmonization of the different radon measurement techniques used across Europe would give the possibility to develop a consistent measurement framework and combine measurement data at a European level.

The guideline should give input for CENELEC/TC 45B, CEN/TC 430, IEC/SC 45B, and ISO TC 85/SC 2 committees to further development of technical standards in this area.

## 2 Instruments measuring the activity concentration of radon-222 in ambient air

Commonly used instruments differ by means of their sampling and measurement methods. The sampling can be passive or active. The latter actively conveys air into the sensitive volume using a pump. Passive sampling utilizes either adsorption onto activated charcoal or the diffusion of radon through a diffusion barrier (aerosol filter). Radon-222 and radon-220 collected inside the sensitive volume disintegrate, and form the decay products consecutively. By applying an appropriate measurement method, the emitted radiation (mostly alpha particles) are measured via direct generation of free charge carriers in the detector, or via secondary effects of the interaction of radiation with the detector (e.g. scintillation light emission, creating latent tracks in solid state polymer detectors).

Passive or active sampling in combination with their respective measurement methods can be used for different measurement tasks:

- a) Instrument with grab or continuous sampling: When a grab sampling is used, an air sample is taken and analyzed immediately thereafter. Grab sampling is used to determine the value of the radon activity concentration in the air at the moment of sampling.
- b) In the case of continuous sampling, the sensitive volume is permanently penetrated by radon. For that purpose, radon containing air enters the sensitive volume either by establishing a continuous air flow using a pump (active sampling), or by diffusion through an aerosol filter (passive sampling). Radon decay products containing in the surrounding air are impeded by an aerosol filter from penetrating the sensitive volume. The continuous sampling enables to acquire the radon activity concentration in the course of time, and to determine its fluctuations and trends. Ionization chambers, scintillation detectors and semiconductor detectors are commonly used for the measurements.
- c) Instrument with long-term passive sampling: The sensitive volume (sensor) is deposed from the analysis unit. It contains a detector, which accumulates and stores the radiation effects over a long time. This is called integrating measurement. The integration over a long period of time has the advantage to average out short-term fluctuations. After the exposure, the detector is analyzed using separate equipment. The accumulated radiation effects are a measure for the exposure to radon. The average radon activity concentration can be determined by dividing the exposure to radon by the time duration of exposure.
- d) Instrument with short-term passive sampling using adsorption: A canister containing an adsorbents (e.g. activated charcoal) is exposed bare to air and collects radon by means of adsorption. The duration of the exposure lasts 2 to 3 days. After the exposure, the activity of the adsorbents is a measure for the average radon activity concentration.

Generally, the sampling and the measurements do not differ between radon-222 and radon-220. Since both occur in conjunction very often, it can be, however, important to discriminate between both. The very different half-lives of both radionuclides are being exploited hereto. A well-dimensioned diffusion barrier extends the diffusion time into the instrument's sensitive measurement volume. Because of its relative short half-live of below 1 min, the major part of radon-220 disintegrates before it can contribute to the measurement effect. In ideal case, radon-222 is measured only. A double chamber system, for example, with and without an extended diffusion barrier makes available the measurement of each of both radon nuclides. Other design solutions exploiting the different half-lives are possible. Discrimination between radon-222 and radon-220 can also be achieved by spectrometric measurements of gammas and alphas emitted by the decay products formed in the sensitive volume.

The EU COUNCIL DIRECTIVE 2013/59/EURATOM on basic safety standards for protection against the dangers arising from exposure to ionizing radiation requires that Member States shall establish a reference level for indoor air concentrations above which it is judged inappropriate to allow exposures to occur. The reference level is stipulated as annual average activity concentration of radon-222. It is recommended to use only instruments suitable for long-term measurements with active or passive sampling for testing the reference level.

## 3 Metrological framework for radon measurements

#### 3.1 Purpose and responsibilities

Metrological framework comprises the organizational and technical measures and arrangements aimed at promoting the quality and comparability of measurements and thus creating confidence in the results and acceptance of the derived protection measures. It is addressed

- 1. to manufacturers concerning the placing on the market of instruments that satisfy common performance requirements,
- 2. to metrological authorities concerning the establishment of appropriate metrological control, and
- 3. to end-users concerning the use of the instruments, quality assurance of measurements and documentation of results.

Metrological control is part of these requirements and means the official surveillance of quality in the performance of measurement tasks. Its implementation in the field of radon is necessary for reasons of high public interest and to ensure public health and safety. The metrological control should be implemented in particular for measurements taken to test the reference level.

#### 3.2 Comparisons

Comparisons are carried out for national (secondary) reference laboratories performing the quantity radon-222 activity concentration in air. They shall be used to harmonize the presentation of the quantity within the national reference laboratories.

The comparison of European radon reference laboratories is a powerful tool to detect discrepancies in traceability and to ensure the quality of radon measurements in Europe. It is recommended to carry out the comparison regularly at intervals of 2 years.

#### 3.3 Technical requirements on radon measuring instruments

IEC 61577-2 stipulates specific metrological requirements and test procedures for radon measuring instruments. Table 1 outlines a selection of radiological and climatic test conditions and the corresponding maximum relative errors  $f_{max}$  which, in general, should be accepted in order to consider a type of radon instruments as appropriate for indoor and outdoor use.

#### 3.4 Type test of radon measuring instruments

By a type test, one or more radon instruments representative for the production are checked for compliance with requirements committed to the intended use. It covers mechanical, electrical and radiological examinations which can be carried out by the manufacturer himself or by an approved testing laboratory. Specific requirements for radon measuring instruments are laid down in the standard series IEC 61577.

Influence quantity	Standard test condition	Reference condition	f <sub>max</sub>	
Aerosol particle concentration* <sup>)</sup>	10 <sup>2</sup> cm <sup>-3</sup> to 10 <sup>6</sup> cm <sup>-3</sup>	10 <sup>4</sup> cm <sup>-3</sup>	25 %	
Ambient gamma dose rate	1 μSv.h <sup>-1</sup>	< 0.1 µSv.h <sup>-1</sup> (natural radiation background)	to be specified by the manufacturer	
Ambient temperature	- 5 °C to 45 °C	20 °C	10 %	
Relative humidity	up to 90 % at 30 °C	65 %	10 %	
Moisture **)	Condensed moisture	20 °C, 65 % r. H.	50 %	
Atmospheric pressure	960 hPa to 1060 hPa	1013 hPa	10 %	

 Table 1: Standard test conditions and the maximum accepted relative errors (after IEC 61577-2)

\*) A polydisperse aerosol distribution shall be used for the tests. The activity median aerodynamic diameter of the aerosol particle should lie within a range from 150 nm to 300 nm and the geometric standard deviation, GSD, between 1.5 and 2.5.

\*\*) only if not explicit excluded by the operating manual of the instrument

#### 3.5 Calibration of radon measuring instruments

A radon service shall ensure the traceability of its measurements to relevant primary standards by an unbroken chain of calibrations. Resulting from calibration, the relationship between values indicated by the measuring instrument and the corresponding known values of activity concentration of radon is ascertained. The relationship is expressed as calibration factor or function. Calibrations should be regularly repeated after two years at least, but also after technical modifications or maintenance procedures, or when the instrument will be used under conditions not covered by previous calibrations.

Calibrations of radon instruments are carried out in reference atmospheres and should be undertaken by approved calibration laboratories. A reference atmosphere is a radioactive atmosphere with parameters being controllable during the calibration procedure. Reference atmospheres can be set up in small containers with several tens of liters up to chambers with volumes of several cubic meters. The quantity <sup>222</sup>Rn activity concentration is established by transferring <sup>222</sup>Rn from a certified standard into a well-known volume. In many cases radon monitors provided by national metrological institutes and calibrated against a radon primary standard are used for transferring radon quantities as well. A reference atmosphere of <sup>222</sup>Rn shall be free of <sup>220</sup>Rn (thoron), otherwise the activity concentration of <sup>220</sup>Rn shall be measured.

Besides the radon activity concentration, relevant parameters are aerosol particle concentration and size distribution as well as climatic parameters. The parameters of the reference atmosphere should be corresponded to the location at which the instrument is intended to be used (e.g. in houses). Special requirements on reference atmospheres are set out in IEC 61577-4.

#### 3.6 Guidelines for interlaboratory comparisons

3.6.1 General considerations

An interlaboratory radon comparison includes the organization, examination and evaluation of instruments exposed to radon. As a rule, comparisons are conducted by recognized reference laboratories. Interlaboratory radon comparisons contribute to ensure a uniform quality standard and will preferably organized for passive radon instruments (nuclear track detectors, activated charcoal detectors, electrets). The comparisons are often used for the purposes of determining the performance and the surveillance of approved radon services.

Intercomparisons are designed for instruments using solid state nuclear track detectors, electrets or activated charcoal and run with similar procedures: Radon services submit a sufficient number of instruments of the same type to the provider. Depending on the applied test scheme devices are randomized and grouped according to the number of exposures. The number of instruments to be submitted depends on the number of exposures and the need for additional transfer instruments being used for measurement effects during storage and delivery. After exposure, radon instruments are delivered to radon services in order to determine the exposures to radon and report the results to the provider of the intercomparison. The provider prepares a report with the measurements and reference data.

Radon services being interested in a radon intercomparison can get further information about providers and organizational conditions from the European Information System on Proficiency Testing Schemes (eptis) available via Internet.

#### 3.6.2 The comparison devices

Due to the metrological characteristics, general criteria for the suitability of an instrument as a reference should be devised, such as linearity, robustness, repeatability, ease of use, data management and storage, etc. This type of measurement device is a standard instrument for radon measurements. As the questionnaire has shown, most European calibration laboratories use the same type of instrument as a reference device.

The instrument is robust and reliable under various environmental conditions and is easy to use. The measurement results are stored unchangeably in an internal memory with sufficient capacity for comparison exercises.

The instrument should be calibrated and checked (linearity and background) before, during and after the interlaboratory comparison in order to ensure the repeatability and traceability of the measurements.

Before the comparison starts, the comparison device should be calibrated at reference laboratory in different atmospheres with stable radon activity concentrations. The calibration factor,  $k_c$ , is given as  $k_c = C_{ref}/(I_{tcd} - I_{tcd,0})$ , where  $C_{ref}$  is the reference activity concentration of radon-222 established in the calibration facility.  $I_{tcd}$  is the indication of the comparison device expressed in the unit Becquerel per cubic meter. The background indication is represented by  $I_{tcd,0}$ .

As the linearity of the indication is a key requirement for the use of the comparison device in the interlaboratory comparison, an additional test for linearity of the indication should be performed. Radon-222 supplied by a gas standard can be transferred into an airtight chamber in which the comparison device was previously placed to measure the radon decrease. When the atmosphere is confined in an airtight chamber, the decrease follows the radioactive decay with a rate, which is equivalent to the radioactive decay constant of radon-222. The empirically found rate of decrease corresponds to the decay constant of radon-222. On a semilogarithmic scale, the measured radon activity concentration is linearly correlated with time, which represents the actual radon activity concentration in the atmosphere.

The calibration measurements should be also flanked by regular background measurements. For this purpose, the device can be enclosed in a volume that was flushed with low-radon air. Low-radon air can be attained by ageing the air. For this, the air delivered in pressure bottles should be stored for several weeks before use. In this case the resulting radon concentration in the volume can be considered negligible (zero). Due to device-specific data processing, the background can also have negative values and it can be assumed that the background indications are normally distributed around the average. The background of the comparison device should be measured before each run.

Visual inspections of the comparison device for damage, including damage to the diffusion filter, checking for proper functioning and checking the set measurement parameters (e.g. calibration factor) are also recommended before deploying the device for the next run. The checks performed should be recorded in the documentation of the comparison.

#### 3.6.3 Procedures

To infom each participant about the course of the comparison and the handling of the comparison device a protocol in attachement A can be used. For legal protection, a cooperation agreement on mutual rights and obligations should be concluded between the reference laboratory and each participant.

The comparison device (calibrated at the reference laboratory) can be sent to each participant for predefined time duration, in order to perform the exposures. The calibrated transfer device has to be compared with the secondary standard used at the participating laboratory. After performing the exposures, the device has to be returned to the reference laboratory. During the intermediate time, when the device is at reference laboratory, the proper operation of the device and its compliance with metrological requirements has to be checked.

Another approach that can be used is to send secondary standard from participating laboratory to reference laboratory. In this case of the comparison the calibration can be performed directly at the reference laboratory. Reference device has to be placed into the radon chamber together with a participant's device.

#### 3.6.4 Recommendation for radon exposures

Different exposure levels have to be selected: one each at a low, medium and high value in the specified range of radon activity concentration. Because the background of the reference device can be affected by the repeated calibrations it is recommended to use one reference device for the calibration in high radon concentrations in air and another one reference device in low radon concentrations in air.

For example, in the case of calibration in the range between 300 Bq/m<sup>3</sup> and 10 000 Bq/m<sup>3</sup> the three different exposure levels can be selected: one each at a low, medium and high value in the specified range of radon activity concentration (Table 2).

No.	Nominal value	Range of accepted deviation
_	(Bq/m³)	(Bq/m³)
1	400	350 – 450
2	1 000	900 - 1 100
3	6 000	5 500 - 6 500

Table 2: Nominal levels of the radon activity concentrations forthe exposure of the comparison device

The medium and high nominal values of 1 000 Bq/m<sup>3</sup> and 6 000 Bq/m<sup>3</sup> represent radon activity concentrations, which are frequently requested calibration points by end-users.

In practice, the participants cannot exactly adhere to the specified nominal values, deviations must be accepted. Therefore, the respective activity concentration ranges should be realized.

The calibration of secondary standards performed at low level activity concentrations (range between 100 Bq/m<sup>3</sup> and 300 Bq/m<sup>3</sup>) can be realized at two levels of radon activity concentration (e. g. 200 Bq.m<sup>-3</sup> and 300 Bq.m<sup>-3</sup>). For calibrations at such low radon activity concentrations it is highly recommended to calibrate secondary standards at the reference laboratory where the stable low radon activity concentration can be established. The design of the device has to be based on a constant supply and constant ventilation model. For the steady-state ( $t = \infty$ ) at a constant air exchange intensity and constant radon input rate, the following applies:

$$C_{V,Rn} = R_{Rn} / (O_{\text{settled}} \cdot \frac{M.p_{\text{ at Q calibration}}}{T_{\text{ at Q calibration}}} / \frac{M.p_{\text{ at confrontation}}}{T_{\text{ at confrontation}}} + I \cdot V)$$
(1)

С <sub>V,Rn</sub>	radon activity concentration (Bq·m <sup>-3</sup> )
Q <sub>settled</sub>	flow rate (m <sup>3</sup> ·h <sup>-1</sup> )
М	molar mass (kg·mol <sup>-1</sup> )
$oldsymbol{p}$ at Q calibration	air pressure 1013,25 (hPa)
$T_{\text{at Q calibration}}$	temperature 273,16 (K)
$oldsymbol{ ho}$ at Rn confrontation	air pressure (Pa)
$T_{\rm atRnconfrontation}$	temperature (K)
1	radon decay constant (h <sup>-1</sup> )
V	volume of radon chamber (m <sup>3</sup> )
<i>R</i> <sub>Rn</sub>	radon emanation power (Bq·h <sup>-1</sup> )

#### 3.6.5 Provided reports

The participants have to be requested to record the checks of the comparison device carried out on-site and the essential data to verify the exposures. In order to facilitate and standardize the records, the form given in Annex A was developed. The information on the comparative measurements of the participants as well as the results and uncertainties has to be given in the calibration reports of results. Example of the report is in the Annex B.

In order to determine the metrological status, the participant have to be asked to provide a brief description of the procedures to establish the exposures, information on the type of local reference instrument and how it works, and information on traceability to primary standards. The exposure data should be presented in a table containing the values of the radon activity concentration,  $C_{\text{RefLab}}$ , being averaged over the respective relevant reference period and the attributed measurement uncertainty.  $C_{\text{RefLab}}$  is determined with the equipment of the participant. The results should be reported for the local climatic conditions at the time of exposure. Additionally, the mean values of temperature, air pressure and relative humidity should be given. Optionally, the participant could also make a correction for climatic conditions to correct the results for standard room conditions (temperature of 20 °C, relative humidity of 50 % or air pressure of 1013 hPa).

The measurement uncertainties have to be given as extended uncertainties resulting from the standard uncertainties of measurement calculated according to the procedures of the participant and multiplied by a coverage factor k = 2.

#### 3.6.6 Validation method

The quantity which allows to assess the performance of the participant is the ratio, R, of the radon activity concentration,  $C_{\text{RefLab}}$ , provided by the participant's secondary standard as average value for the relevant reference period to the mean radon activity concentration of the comparison device,  $C_{\text{CD}}$ , measured within the same time interval,

$$R = \frac{C_{\text{RefLab}}}{C_{\text{CD}}}$$
(2)

The standard uncertainty  $\Delta R = u(R)$  is calculated from the uncertainty propagation of Equation ((2). The relative uncertainty is given by

$$\frac{\Delta R}{R} = \sqrt{\left(\frac{\Delta C_{\text{RefLab}}}{C_{\text{RefLab}}}\right)^2 + \left(\frac{\Delta C_{\text{CD}}}{C_{\text{CD}}}\right)^2} \tag{3}$$

 $\Delta C_{\text{RefLab}} = u(C_{\text{RefLab}})$  is the combined uncertainty for a coverage factor of k = 1 as provided by the participant determined according to its own procedures (respectively from the participant's secondary standard calibrated at the reference laboratory). The uncertainty includes the statistical variation from repeated observations (type A evaluation of standard uncertainty) and contributions from other sources, in particular from data provided in calibration and other certificates (type B evaluation of standard uncertainty) [1].  $\Delta C_{\text{CD}} = s(C_{\text{CD}})$  is the uncertainty of the mean radon activity concentration determined by the comparison device. It is given by the standard deviation,  $s(C_{\text{CD}})$ , of the mean,

$$s(C_{\rm CD}) = \sqrt{\frac{\sum (C_{\rm CD,j} - C_{\rm CD})^2}{n(n-1)}}$$
 (4)

 $C_{CD,j}$  is the  $j^{th}$  of *n* measurements taken with the comparison device. Other contributions to the uncertainty, particularly due to calibration factor are not considered. This is due to the basic purpose of the comparison device, which is not to measure the exact value of the radon activity concentration, but to provide an indication that depends linearly on the true value of the radon activity concentration. Linearity has to be warranted at least within the accepted ranges of nominal values of the radon activity concentration.

It should be noted that the simple averaging of the measurements performed with the comparison device and the use of Equation (4) is valid if the activity concentration is kept constant during the relevant reference period (suitable for calibrations at reference laboratory which is able to achieve stable low level radon atmosphere). This cannot be ensured at the participant's laboratory. Therefore, the change in activity concentration over time must be well known and taken into account when determining the comparative value,  $C_{CD}$ . In such cases, the participant has to provide information on how the comparative value is determined from the readings of the comparison device. In general the radon activity concentration established in a confined atmosphere decreases due to radioactive decay. The comparative value at the reference time  $t_{ref}$  is given by averaging the measured values corrected for the reference time,

$$C_{\rm CD}(t_{\rm ref}) = \frac{1}{n} \sum_{j} C_{\rm CD,j} \ e^{-\lambda(t_{\rm ref} - t_j)}$$
(5)

where  $t_j$  represents the measurement time of  $C_{CD,j}$  and I the decay constant for radon-222. Equation (5) must be modified accordingly if the rate of decrease differs from that of radioactive decay.

When the parameter  $R_i$  denotes the ratio R calculated for the *i*<sup>th</sup> of *n* participants and  $u_i$  is the standard uncertainty attributed to  $R_i$ , the collective average,  $R_w$ , is determined by

$$R_{w} = \frac{\frac{R_{1}}{u_{1}^{2} + \dots + \frac{R_{n}}{u_{n}^{2}}}{\frac{1}{u_{1}^{2} + \dots + \frac{1}{u_{n}^{2}}}} = \sum_{i=1}^{n} w_{i}R_{i}$$
(6)

where  $w_i$  represents the weighting for the ratio  $R_{i_i}$ 

$$w_i = \frac{1/u_i^2}{\sum_{i=1}^n 1/u_i^2}$$
(7)

As in the calculation of the collective average the ratio of the individual participant is weighted according to the attributed uncertainty, the impact on the collective average of participants with lower uncertainties is stronger than for other participants. Assuming that  $R_w$  is normally distributed with weights defined in Equation (7) and that the  $R_i$  are independent, the corresponding variance is given by [2], [3]

$$\sigma^{2}(R_{w}) = u^{2}(R_{w}) = \frac{1}{\frac{1}{u_{1}^{2}} + \dots + \frac{1}{u_{n}^{2}}} = \frac{1}{\sum_{i=1}^{n} \frac{1}{u_{i}^{2}}}$$
(8)

#### 3.6.7 Consistency check

A check of mutual consistency is required by the BIPM consultative committee CCQM [2]. It aims to test the hypothesis that the participants have a common mean value and that the deviations from this value are normally distributed.

The consistency check is performed by a chi-squared test over the number of *n* measurements (or participants). The observed test parameter  $\chi^2_{obs}$  is calculated by

$$\chi_{\rm obs}^2 = \sum_{i=1}^n \left(\frac{R_i - R_{\rm w}}{u_i}\right)^2$$
(9)

According to CCQM [2] the test parameter is compared with the quantile of the chi-squared distribution for the significance level  $1 - \alpha$  with  $\alpha = 0.05$ . The following decisions have to be made:

- a) If  $\chi^2_{obs} < n 1$  the results are mutually consistent and the uncertainties account fully for the observed dispersion of the values;
- b) If  $n 1 \le \chi^2_{obs} < \chi^2_{0,05;n-1}$  the data provide no strong evidence that the reported uncertainties are inappropriate, but there remains a risk that additional factors are contributing to the dispersion;
- c) If  $\chi^2_{\text{obs}} > \chi^2_{0.05;n-1}$  the data should be considered as mutually inconsistent.

The tests have to be performed for each exposure level and for the complete data set of all levels including singular exposures.

#### 3.6.8 The uncertainty-weighted collective average

The uncertainty-weighted collective average,  $R_w$ , for the different exposure levels is calculated from Equation (6). The square root of the variance from Equation (8) is the standard uncertainty,  $u(R_w)$ .

Assuming that the data of the comparison device always determine the respective true value of the radon activity concentration over the whole range and the measurements performed with participant's secondary standards are normally distributed around this true value, then it is expected that the uncertainty-weighted collective average is compensated, resulting in  $R_w = 1$ .

#### 3.6.9 The comparison reference value (CRV)

In order to verify the performance of the participant's secondary standards a comparator is needed, enabling the normalization of the different levels of the radon activity concentration. The comparison device provided by the coordinator is used as the comparator. As the comparison device is characterized by an indication, which is verifiably linear over the entire range, the comparison of the different radon activity concentrations measured by participant's secondary standards is made possible by their ratio to the indication of the comparison device as is given in Equation (2). From the ratios the uncertainty-weighted collective average,  $R_w$ , is calculated by Equation (6).

The measurements taken by the comparison device are subject to uncertainties, which in turn are caused by uncertainties in the realisation of the radon activity concentration in the facility of the coordinator and the calibration of the device in it. For this reason, greater importance is attached to the average value, which includes all participants. This average value should therefore be used as a reference for the interlaboratory comparison.

In order to overcome the above disadvantages and to device a balanced mean that is accepted as a reference value, the parameter  $R_i^*$  is calculated for the  $i^{th}$  participant, defined by the ratio

$$R_i^* = \frac{R_i}{R_w} \tag{10}$$

The expectation value,  $E(R_i^*)$ , is the weighted sum over each participant,

$$E(R_i^*) = \sum_{i=1}^n w_i R_i^* = \frac{1}{R_w} \sum_{i=1}^n w_i R_i = 1$$
(11)

The weights,  $w_i$ , are given by Equation (7). The new parameter  $R_i^*$  excludes the influence of the comparison device. It implies that the ratios  $R_i$  calculated for the participants are normally distributed around the common mean value given by the expectation value  $E(R_i^*) = 1$ . The expectation value is the comparison average and represents the KCRV.

The variance of the KCRV is the weighted root mean square deviation,

$$\sigma^{2} = \sum_{i=1}^{n} w_{i} (R_{i}^{*} - 1)^{2} = \sum_{i=1}^{n} w_{i} (R_{i}^{*2} - 2R_{i}^{*} + 1) = \sum_{i=1}^{n} w_{i} R_{i}^{*2} - 1$$
(12)

After replacing the weights by Equation (7), it follows

$$\sigma^{2} = \frac{\sum_{i=1}^{n} \frac{1}{u_{i}^{2}} \sum_{i=1}^{n} \frac{R_{i}^{2}}{u_{i}^{2}}}{\left(\sum_{i=1}^{n} \frac{R_{i}}{u_{i}^{2}}\right)^{2}} - 1$$
(13)

The standard uncertainty associated with the comparison reference value (CRV) is the square root of the variance. It expresses the range of variation within which a certain radon activity concentration is realized in the atmospheres of European radon calibration facilities and is thus a measure of the degree of agreement between the participants.

#### 3.6.10 Alternative selection of the comparison reference value

In case that the values for  $u(R_w)$  are not reasonable for quantifying the uncertainty of the CRV, the method of the power-moderated mean is proposed as an alternative for the determination of the uncertainty [4]. The method applies to data which are mutually independent and normally distributed around the same value. Its results are generally intermediate between arithmetic and weighted mean [4]. The method is recommended in cases where the condition  $n - 1 \le \chi^2_{obs}$  is obtained.

The calculations of the power-moderated mean can be performed using the Excel-sheet *MET511639suppdata.xlm*, which is available for download on the Internet [5]. The automatic algorithm for moderating the relative weighting should be used.

The approach provided by the Excel-sheet MET511639suppdata.xlm should be also used for the identification of extreme values. Extreme values are indicated when the difference between the measured ratio and the power-moderated mean,  $d_i = R_i - R_{w,pm'}$  exceeds the constraint specified by [4].

$$|d_i| > k \cdot u(R_{w,pm}) \sqrt{\left(\frac{1}{w_i} + 1\right)}$$
(14)

#### 3.6.11 Influence of climatic conditions during the calibrations

Standard room conditions are: temperature of 20 °C, relative humidity of 50 % or air pressure of 1013 hPa. The different climatic conditions raise the question of their influence on the results of this study especially in the case when the secondary standard is compared to the transfer standard at the participant laboratory. The multiple correlation method can be used for the test. It describes the power of the association between a specified random variable and a group of independent random variables.

For a given exposure level, the multiple correlation coefficient,  $r_{R,(T,p,rH)}$ , of the ratios  $R_i^*$  according to Equation (10) and the group of the climatic parameters temperature (*T*), air pressure (*p*) and relative humidity (*rH*) is determined from the square root of [6], [7], [9]

$$r_{R,(T,p,rH)}^{2} = \begin{pmatrix} r_{R,T} \\ r_{R,p} \\ r_{R,rH} \end{pmatrix}^{T} \begin{pmatrix} \mathbf{1} & r_{T,p} & r_{T,rH} \\ r_{T,p} & \mathbf{1} & r_{p,rH} \\ r_{T,rH} & r_{p,rH} & \mathbf{1} \end{pmatrix}^{-1} \begin{pmatrix} r_{R,T} \\ r_{R,p} \\ r_{R,rH} \end{pmatrix}$$
(15)

where  $r_{x,y}$  represents the Pearson correlation coefficient [6] for the variables x and y. The Pearson correlation coefficient is a measure of the strength of a linear association between the two variables and is calculated by

$$r_{x,y} = \frac{\sum_{i=1}^{o} (x_i - \bar{x}_i) (y_i - \bar{y}_i)}{\sqrt{\sum_{i=1}^{o} (x_i - \bar{x}_i)^2 \sum_{i=1}^{o} (y_i - \bar{y}_i)^2}}$$
(16)

The summations run over the number of participants, o = 9, involved in the test. The parameter  $r_{R,(T,p,rH)}^2$  is also denoted as coefficient of determination of the multiple correlation.

In order to assess the significance of a given  $r_{R,(T,p,rH)}^2$ , the ratio F is computed as [6], [7], [8]

$$F = \frac{r_{R,(T,p,rH)}^2(o-1-q)}{q(1-r_{R,(T,p,rH)}^2)}$$
(17)

The ratio *F* is distributed under the null hypothesis as a Fisher distribution with *q* and o - 1 - q degrees of freedom. The value for *q* is the number of the independent variables temperature, air pressure and humidity, therefore, q = 3. The coefficient of determination,  $r_{R,(T,p,rH)}^2$ , is significant for a level 1 - a, if  $F \ge F_{q,o-1-q;1-\alpha}$ .

Besides the values for multiple correlation, the coefficients of determination for the pairwise correlations,  $r_{x,y}^2$ , are also given. The ratio *F* is compared to the tabulated quantile,  $F_{3,5;0,95}$ , of the Fisher distribution for a = 0.05. Since  $F > F_{3,5;0,95}$  a correlation must be assumed.

#### 3.6.12 Traceability and correlations between the participants

The participants have to be requested to provide information on how the traceability of the radon activity concentration is realized. The radon activity concentration is a combined quantity consisting of the activity of the gaseous nuclide radon-222 and the volume. The volume is the capacity of the enclosed space containing the atmosphere for the realization of the quantity.

Due to the existence of the different branches in traceability, it cannot be excluded that discrepancies in the realizations of the quantity are transferred to the following facilities in the traceability chain. Such correlations would affect the prerequisite of a normally distributed data set.

The data set should be tested for correlation using the Pearson correlation coefficient [6]. The coefficient,  $r_{R_i,R_k}$  is calculated using the relevant data according to

$$r_{R_{i},R_{k}} = \frac{\sum_{j=1}^{o} (R_{i,j} - \bar{R}_{i,j}) (R_{k,j} - \bar{R}_{k,j})}{\sqrt{\sum_{j=1}^{o} (R_{i,j} - \bar{R}_{i,j})^{2} \sum_{j=1}^{o} (R_{k,j} - \bar{R}_{k,j})^{2}}}$$
(18)

where  $R_{i,j}$  and  $R_{k,j}$  are the ratios corresponding to Equation (10) for the exposure level j of the  $i^{th}$  or  $k^{th}$  participant, respectively.  $\overline{R}_{i,j}$  and  $\overline{R}_{k,j}$  are the respective mean ratios for the  $i^{th}$  or  $k^{th}$  participant.

The hypothesis that there is no correlation between the  $i^{th}$  and  $k^{th}$  participant is tested against the hypothesis that there is a correlation using the test parameter [6]

$$t = \frac{r_{R_i, R_k} \sqrt{o - 2}}{\sqrt{1 - r_{R_i, R_k}^2}}$$
(19)

The hypothesis of no correlation has to be rejected if  $|t| > t_{o-2;1-\alpha/2}$ , where  $t_{o-2;1-\alpha/2}$  is the quantile of the Student's t-distribution for the two-sided test with a degree of freedom of o - 2 = 1 at the significance level  $1 - \frac{\alpha}{2}$ .

## 4 Consideration / elimination of thoron influence on radon measurements

The influence of thoron (<sup>220</sup>Rn) on the radon activity concentration measurements has been observed in some radon monitors and detectors. This influence, if not properly corrected, can introduce bias in the radon results. To avoid or correct this bias radon monitors/detectors should be tested for thoron cross-interference. In the future, the manufacturers of radon instruments should include this information in the specifications of the instrument. Within the MetroRADON project, WP2 aimed to investigate and to reduce the influence of thoron (<sup>220</sup>Rn) and its progeny on radon (<sup>222</sup>Rn) end-user measurements and radon calibrations.

The following basic guidelines can be recommended to follow for assessment and reduction of thoron interference on radon monitors and detectors:

- Both thoron and its progeny (<sup>216</sup>Po, <sup>212</sup>Pb, <sup>212</sup>Bi+<sup>212</sup>Po/<sup>208</sup>Tl) need to be taken into account in testing for thoron interference of radon monitors/detectors and analysis of the results.
- The testing of radon monitors for thoron sensitivity should include reference (secondary) thoron monitor that is calibrated with, or traceable to a primary thoron measuring system. We recommend a stable reference thoron atmosphere to be created in an exposure chamber by flushing air with constant flow-rate consecutively through the thoron source and the exposure chamber. To ensure thoron homogeneity fan(s) should operate in the exposure chamber during exposure. We recommend the reference instrument 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. The air pressure, temperature, humidity and air velocities (if applicable) during the tests should be recorded.
- The cross-interference tests should be planned taking into account the possible contribution from thoron progeny (<sup>216</sup>Po, <sup>212</sup>Pb, <sup>212</sup>Bi+<sup>212</sup>Po/<sup>208</sup>TI). After the end of thoron exposure the integrated passive detectors should be left for at least three days at low thoron/radon levels before analysis. This is needed to leave thoron progeny deposited in their volume to decay.
- A minimum of three days test with a high thoron activity concentration (around 10 kBq.m<sup>-3</sup> or more) is recommended to determine an accurate final thoron cross interference of the active monitors, instead of the 4 hours at 1 000 Bq.m<sup>-3</sup> required in the IEC 61577-2 standard.
- 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.
- 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 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 interference 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 passive detectors diffusion barriers might be considered in the design and tested in the prototypes. If instruments are scheduled to work at high humidity we recommend diffusion barriers based on polyethylene foils of low density polyethylene.
- 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 temperature 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.
- For detectors in which the usage of polyethylene packing is planned to reduce the thoron interference, a possible temperature bias may be introduced. In particular, for detectors that have response decreasing with the increase of the temperature, also the temperature bias can be compensated if the anti-thoron polymer packing is designed as a compensation module, as described in Deliverable 2. Such compensation module can reduce/compensate the temperature dependence of the detectors' response + thoron interference + humidity influence.

### 5 Influence of calibration on radon measurements

In chapter 3.5 it was already mentioned that a radon calibration service shall ensure the traceability of its measurements to relevant primary standards by an unbroken chain of calibrations. With other words, the <sup>222</sup>Rn activity concentration in air in a calibration chamber must be traceable to primary <sup>222</sup>Rn activity standards and to primary volume standards. In addition, the uncertainty of the <sup>222</sup>Rn activity concentration in air in the calibration chamber should be reasonable low in order to establish calibration factors for radon measurement instruments with low uncertainties.

In chapter 3.6.3 different levels of <sup>222</sup>Rn activity concentrations are described that are typically used for the calibration of instruments. Traceable activity concentrations higher than 1 kBq/m<sup>3</sup> are usually established by transferring a certified <sup>222</sup>Rn gas activity standard into a calibration chamber with a certified volume.

For lower activity concentrations, a decaying <sup>222</sup>Rn gas activity standard is not the best choice. Within the MetroRadon project, the aim of work package 1 (WP1) was the development of novel procedures for the traceable calibration of radon (<sup>222</sup>Rn) measurement instruments at low activity concentrations (100 Bq/m<sup>3</sup> to 300 Bq/m<sup>3</sup>) with relative uncertainties  $\leq 5 \%$  (*k*=1). The results are described in detail in the Deliverable report D1. 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 reach these uncertainties, new <sup>222</sup>Rn and <sup>220</sup>Rn radioactive reference sources with stable and known radon emanation capacity were developed in a first step. PTB, supported by JRC has developed <sup>222</sup>Rn and <sup>220</sup>Rn emanation sources including a detector system for the continuous monitoring of the emanated activity of <sup>222</sup>Rn relative to the activity of the <sup>226</sup>Ra source (and emanated <sup>220</sup>Rn relative to activity of <sup>228</sup>Th source) traceable to primary standards. CEA has developed <sup>222</sup>Rn and <sup>220</sup>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 <sup>222</sup>Rn and <sup>220</sup>Rn in an air flow. CMI with support from SUJCHBO has developed a long term stable <sup>222</sup>Rn low activity emanation flow-through standard source based on a metering flow controller and dispenser generating a known <sup>222</sup>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 realization of constant and stable <sup>222</sup>Rn radon activity concentrations in reference chambers and development of procedures for the calibration of radon measurement instruments at low activity concentrations was the second step. The new <sup>222</sup>Rn emanation sources developed and compared in the first step 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 <sup>222</sup>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/m<sup>3</sup> to 300 Bq/m<sup>3</sup> using <sup>222</sup>Rn gas and emanation standards. The result is not one single and unified calibration method but very similar methods using slightly different sizes of calibration chambers and a bit different instrumentation of sensors, flow controllers, etc. The comparability of the calibrations at the six mentioned facilities is documented in Deliverable D7 "Validation report on the traceability of primary and secondary radon calibration facilities in Europe". The goal to develop procedures for the calibration of radon measurement instruments at low activity concentrations (in the activity concentration range from 100 Bq/m<sup>3</sup> to 300 Bq/m<sup>3</sup>) with sufficiently low relative uncertainties  $\leq 5$  % 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 of the activity concentrations.

## 6 Conclusions

This document provides guidelines and recommendations on how to evaluate secondary standards at participant's laboratory or directly at the reference laboratory. The first approach is suitable for cases when the secondary standard is calibrated in the range between 300 Bq/m<sup>3</sup> and 10 000 Bq/m<sup>3</sup>. For lower radon concentrations, calibration in a stable radon atmosphere at the reference laboratory is recommended. It means that the comparison device is sent to each participant or each participant is requested to send their secondary standard to reference laboratory.

The parameter allowing to assess the performance of the participant is the ratio of the radon activity concentration measured by the secondary standard for the relevant reference period to the mean of the values measured by the comparison device over the same period.

The interlaboratory comparison of European radon calibration facilities 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 interlaboratory comparisons regularly.

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Annex A: Standardisation of the records

Transfer comparison device		
Designation		
Туре		
Serial No.		

1	Checking the device for measurement capability				
		Records and statements			
	Battery power	o.k.?			
	Mode of operation				
	Time interval of integration				
	Indication of date and time	o.k.?, time difference to local time?			
	Check diffusion filter for damages	Damages: yes/no			
	Visual inspection of general state	o.k.?			
Date	Date and Signature				

2	Exposures			
		Records and statements		
2.1	Exposure No. 1			
	Level of radon activity concentration in $Bq \cdot m^{-3}$			
	Turn on the device Place device in radon atmosphere, (Plug in power supply, if necessary)	o.k.?		
	Date and time of commencement of exposure	Local date and time		
	Further relevant information about the operation of the facility and monitoring the atmosphere (e.g. air pressure in hPa, temperature in °C)			
	Date and time of finalizing the exposure	Local date and time		

2.2	Exposure No. 2	
	Level of radon activity concentration in $Bq \cdot m^{-3}$	
	Turn on the device Place device in radon atmosphere, (Plug in power supply, if necessary)	o.k.?
	Date and time of commencement of exposure	Local date and time
	Further relevant information about the operation of the facility and monitoring the atmosphere (e.g. air pressure in hPa, temperature in °C)	
	Date and time of finalizing the exposure	Local date and time

2.3	Exposure No. 3	
	Level of radon activity concentration in Bq·m <sup>-3</sup>	
	Turn on the device Place device in radon atmosphere, (Plug in power supply, if necessary)	o.k.?
	Date and time of commencement of exposure	Local date and time
	Further relevant information about the operation of the facility and monitoring the atmosphere (e.g. air pressure in hPa, temperature in °C)	
	Date and time of finalizing the exposure	Local date and time

3	Preparation for Shipment			
		Records and statements		
	Turn off the device	o.k.?		
	Are the device and included accessories (according to list) safely packed in the transport box?	o.k.?		
	Prepare the transport box for shipment (tightly closed and labelled)	o.k.?		
Date and Signature				

Annex B: Standard results report

Comparison device	
Designation	
Туре	
Serial No.	

Short description of the procedures applied for exposing the transfer comparison device, Information about the local reference instrument for the radon activity concentration, Information about traceability to primary standards

The results of the intercomparison are summarized in the table. Besides the exposure data, the mean values of temperature, *T*, air pressure, *p*, and relative humidity, *r*.*H*., of the radon atmospheres are given. This serves the purpose to characterise the measurement conditions during the exposures.

No	C <sub>M,net</sub> [Bq∙m <sup>-3</sup> ]	<i>U</i> ( <i>C<sub>M,net</sub>) [Bq∙m⁻³]</i>	C <sub>RefLab</sub> [Bq∙m <sup>-3</sup> ]	<i>U(C<sub>RefLab</sub>)</i> [Bq∙m <sup>-3</sup> ]	<i>Т</i> [°С]	р [hPa]	r.H. [%]
1							
2							
3							

 Table: Summary of results for the exposures No. 1, 2 and 3

 $C_{M,net}$  is the mean value of radon activity concentration measured by the comparison device. This value is determined from the collocated list of single measurement data provided by reference laboratory after reading out the data from the device. The single measurement data are provided with a correction for background and the application of the calibration factor of reference laboratory. For the determination of  $C_{M,net}$  only those measurement data are considered, which are taken in the decisive reference period.

 $C_{RefLab}$  is the mean value of the radon activity concentration established in the radon atmosphere of the laboratory. This value is determined with the equipment of the participant's laboratory. For the determination of  $C_{RefLab}$  only those measurement data are considered, which are taken in the decisive reference period.

The measurement uncertainty given as extended uncertainty resulting from the standard uncertainty of the corresponding mean value multiplied with a coverage factor k = 2. The standard uncertainty of the mean value is calculated from the statistical variation of the single measurements around this mean value. The following extended uncertainties are included in the table:

 $U(C_{RefLab})$  Measurement uncertainty of  $C_{RefLab}$ 

 $U(C_{M,net})$  Measurement uncertainty of  $C_{M,net}$ 

..... Date and Signature

Name and e-mail of the person in charge (use capital letters)