

The radon activity concentration –

An Euromet and BIPM supplementary comparison

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Abstract: For the first time a comparison for the radon activity concentration has been performed within the scope of Euromet. In the project 657 ‘Comparison of calibration facilities for the radon activity concentration’ 12 participants from 9 countries compared different radon reference atmospheres at 1 kBq/m³, 3 kBq/m³ and 10 kBq/m³ via a transfer standard. The comparison was listed as BIPM supplementary comparison. The results of the most participants are correlated due to a common traceability to one single radon gas standard producer. This makes a careful correlation analysis necessary to achieve the comparison reference value. The results of the comparison as well as the complex analysis of the correlated set of data is presented and discussed.

1. Introduction

Inhalation of radon progeny renders about half of the natural ionising radiation exposure of the general public. This exposure is considered as second largest source of lung cancer after cigarette smoking. In homes and most workplaces, radon gas activity concentration is closely correlated with radon progeny exposure. Radon gas can be measured with inexpensive passive detectors. Thus, Public Health authorities in most countries have issued recommendations for limits and the reduction of radon gas activity concentration in homes and workplaces. A high quality of measuring services for radon gas activity concentration requires calibration facilities where radon detectors and instruments can be calibrated under well-defined conditions traceable to national or international standards.

These facilities, generally known as ‘radon chambers’, have never been subject to a Euromet or BIPM comparison before. Euromet project 657, ‘Comparison of calibration facilities for the radon activity concentration’, provides such a comparison for the most important quantity in operating a radon chamber: the activity concentration of radon in Bq/m³. The comparison meets two needs: (1) harmonization of metrology within the scope of the MRA - the 'Arrangement of mutual recognition of equivalence of national standards and of calibration cer-

tificates issued by national metrology institutes'; and (2) compliance with the increased demands of the EURATOM directive, transferred into national radiation protection regulations, with regard to natural radioactivity and its quality-assured measurements.

Ten years after Euromet project 239 '222 Rn measurement systems' (Dean and Burke, 1994) dealing with the quantity activity of radon, Euromet project 657 'Comparison of calibration facilities for the radon activity concentration' conducted a comparison for the quantity radon activity concentration.

2. Comparison procedure

In April 2002 the planning of the project and development of the Euromet proposal took place. The quality systems of the national institutes provided the basis for running the comparison. A direct comparison of reference atmospheres created in radon chambers is not possible. Instead, interchange of an electronic radon measuring instrument was chosen as a convenient means for a relative comparison of radon reference atmospheres. The feasibility of this procedure was tested in a pilot study (Butterweck et al., 2002, Röttger et al. 2004). The instrument, which has demonstrated ruggedness during shipping in the past, is referred to as the Comparison Device (CD). For CD an instrument from the company Genitron was chosen, type AlphaGuard S/N 1145. The project used the CD to derive a calibration factor for the CD for each chamber, which is the quantity which is compared here.

During the period November 2002 to April 2004, 12 institutes from 9 countries calibrated the CD in accordance with their quality systems at three different activity concentrations: 1 kBq/m³, 3 kBq/m³ and 10 kBq/m³. In June 2004 the preliminary results were presented during a Radon Metrology Meeting in Braunschweig and in January 2005 the final report was published, s. Röttger et al.. 2005.

Between the calibrations at the participating laboratories, the CD was inspected at PTB in respect to function and background. The progress of the comparison was communicated to the participants by comparison newsletters via e-mail. The complete history of the CD exposures

(activity concentration, temperature, pressure, humidity) was documented electronically and in an instrument journal. Figure 1 shows the exposure history and the increase of the instrument background during the comparison and the semi-annual automatic correction by the CD.

Figure 1

After each calibration was finished, the complete results (results of the measurements together with their assigned uncertainties) were reported to PTB in a closed envelope. Since the PTB also took part in the measurements, the comparison exercise started at PTB and its own results were immediately reported to the EUROMET TC-IR chair.

Aside from the inevitable variation of the background, the following parameters were considered to be potential influence parameters of the calibration:

1. Time (drift of response of the CD during the comparison)
2. Linearity of the response at different activity concentrations
3. Environmental parameters (temperature, humidity, pressure)

A drift in the response of the instrument would be observable by comparing the collected results of all participants at all three calibration points, since the calibrations were performed in series. No drift was found within the assigned uncertainties.

A non-linear response of the CD would be observable by looking at the results (calibration factor) as a function of the activity concentration. There was no non-linearity detected within the range of activity concentrations of the comparison and its assigned uncertainties.

And finally: the calibrations were performed over quite a wide range of environmental parameters: temperature from 15 °C to 28 °C, air pressure from 930 hPa to 1025 hPa and relative humidity from 29 % to 60 %. There was no correlation between the values of environmental parameters and the results of the calibration detected within the assigned uncertainty for the CD.

The CD thus meets the stability requirements of a transfer standard, supported by the described procedures of quality assurance devised for this comparison.

3. Traceability and correlation

The traceability of the radon activity concentration in a reference chamber is established by using either a reference atmosphere (primary standard) or one or more reference instruments (secondary standard).

Calibration by a primary standard is based on the knowledge of an activity concentration, by means of a radon gas activity standard and a vessel of known volume which is gas-tight to noble gases. The system under test and the radon gas activity standard are enclosed in the vessel. The activity concentration chosen for the point of calibration is calculated and compared to the reading of the system under test for the calculation of the calibration factor. After calibration, the system under test can now be used as a secondary standard, or reference instrument.

A calibration by a secondary standard is based on a comparison of an instrument with a reference instrument, which was calibrated in or traceable to a reference atmosphere in the past. The radon activity concentration chosen for the point of calibration is established, and both systems are placed in the same atmosphere. The reading of the reference instrument and the system under test are observed simultaneously for the calculation of the calibration factor. The traceability chains in their length as well as in their origin are fundamental for the correct analysis of the comparison data. The length of each individual traceability chain is summarized in the absolute value of the uncertainty which can be reached. While the uncertainty increases with the length of the traceability chain, the correlation (if there is one) decreases.

An overview of some traceability chains for the radon activity and the radon activity concentration in Europe is shown in figure 2.

Figure 2

In this comparison, the CD was calibrated using a primary standard by IRSN (using a standard traceable to Laboratoire National Henri Becquerel (LNHB)) and by PTB (using a standard traceable to PTB, 6.11, activity lab). These are the only ones with calibrations directly to

a reference atmosphere (in the case of PTB, only for two of the three calibration points of the comparison: 3 kBq m⁻³ and 10 kBq m⁻³).

In these cases the calibration factor is derived using the equations:

$$c_r = \frac{A}{V_r - V_c} \quad , \quad k_c = \frac{c_r}{c_c - c_{c,bg}} = \frac{A}{V_r - V_c} (c_c - c_{c,bg})^{-1} \quad (1)$$

Here A is the activity of the radon gas standard in Bq, V_r is the volume of the reference chamber, V_c is the displaced volume by the comparison device (all volumes in m³), c_r is the activity concentration of the reference atmosphere, c_c is the activity concentration measured by the comparison device and c_{c,bg} is the background activity concentration of the comparison device, due to contamination (all activity concentrations in Bq m⁻³). The result of the calibration is represented by k_c the calibration factor of the comparison device.

The calibrations of the CD by secondary standards were traceable to three different primary standards. Inte-UPC, HPA-RP (formerly NRPB), PSI, PTB and SUJCHBO were traceable to PTB (6.11, activity lab), IRSN was traceable to LNHB, and SSI was traceable to NIST (National Institute of Standards and Technology, USA). In these cases, an activity standard was used to calibrate the secondary standard, or reference instrument, which was used to calibrate the CD.

In the cases of ARCS, BEV, BfS, MPA, PTB and STUK, the secondary standard, or reference instrument, was sent to PTB and calibrated in the PTB 6.12 radon reference chamber. The secondary standard was then used to calibrate the CD.

In these cases the calibration factor is derived using the equations:

$$k_t = \frac{c_r}{c_t - c_{t,bg}} = \frac{A}{V_r - V_t} (c_t - c_{t,bg})^{-1} \quad , \quad k_c = k_t \frac{c_t - c_{t,bg}}{c_c - c_{c,bg}} \quad (2)$$

Here c_t is the activity concentration measured by the transfer standard, c_{t,bg} the background activity concentration of the transfer standard, due to contamination (all activity concentrations in Bq m⁻³) and V_t is the displaced volume by the transfer standard (in m³). The calibration factor of the transfer standard is k_t. The result of the calibration is represented again by k_c the calibration factor of the CD.

4. Data analysis and results

The number of participating institutes is $N = 12$, by numbers $i = 1...N$ (1: PTB, 2: BfS, 3: STUK, 4: BEV, 5: ARCS, 6: Inte-UPC, 7: IRSN, 8: SUJCHBO, 9: PSI, 10: SSI, 11: HPA, 12: MPA). The input quantities to the evaluation are the institutes' measurements, denoted by $x_i = k_{c,i}$ and the standard uncertainties associated with these values, denoted by $u(x_i)$.

In a first step, arithmetic mean and standard deviation are calculated. In a second step weighted mean and its assigned uncertainty is obtained. Finally the comparison reference value (CRV) and the coverage interval for 95% is determined. The coverage interval so obtained is central with respect to probability, i.e. 2.5 % of the distribution of possible values lies to the left of the interval and 2.5 % to the right. It is not generally the shortest interval, unless the distribution is symmetric, e.g. ISO, 1995; DAKC, 2003; Siebert, B. 2004; Sommer, K. 2004.

The comparison reference value (CRV) Y is calculated using the result vector $X = \{x_1, \dots, x_{12}\}$, the design matrix A and the covariance matrix U_X , where $u(x_i, x_k) = u(x_i) u(x_k) r(x_i, x_k)$ with the correlation coefficient $r(x_i, x_k)$.

$$Y = (A^T U_X^{-1} A)^{-1} (A^T U_X^{-1} X) \quad , \quad (3)$$

$$A = \begin{pmatrix} 1 \\ \vdots \\ \vdots \\ \vdots \\ \vdots \\ 1 \end{pmatrix} \quad , \quad U_X = \begin{bmatrix} u^2(x_1) & u(x_1, x_2) & \cdots & \cdots & u(x_1, x_{12}) \\ u(x_2, x_1) & u^2(x_2) & \vdots & \vdots & \vdots \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ u(x_{12}, x_1) & \cdots & \cdots & \cdots & u^2(x_{12}) \end{bmatrix}$$

Correlated results x_i are expressed in this formula by the non-diagonal elements of the matrix U_X . If all results would be uncorrelated, the matrix had only diagonal elements and the weighted mean value would be identical to the result obtained by equation (3). The complete results, as well as the comparison reference value are given in figure 3.

Figure 3

Table 1 summarizes the results (calibration factors) in form of arithmetic mean, weighted mean and comparison reference value with their assigned uncertainties.

Table 1

5. Conclusions

The Euromet project 657 was performed successfully according to the guidelines for Euromet projects and BIPM supplementary comparisons. The degree of equivalence was calculated with regard to the CRV as well as to each participant. The details are available in the BIPM database.

The results of all participants are consistent within the scope of the assigned uncertainties. Nevertheless, the comparison initiated a detailed discussion of results, procedures, data analysis and calculation of uncertainties between the participants. This discussion is supported by the preparation of a BIPM key comparison for the quantity ‘radon gas activity’, which will be performed during 2005/2006. Thus the basis for a new comparison of the radon activity concentration is set. Since the worldwide importance of the ‘radon problem’ in regulation, health and from the metrological point of view, has increased in recent years, the next comparison will probably have even more participants than this one.

6. Acknowledgement

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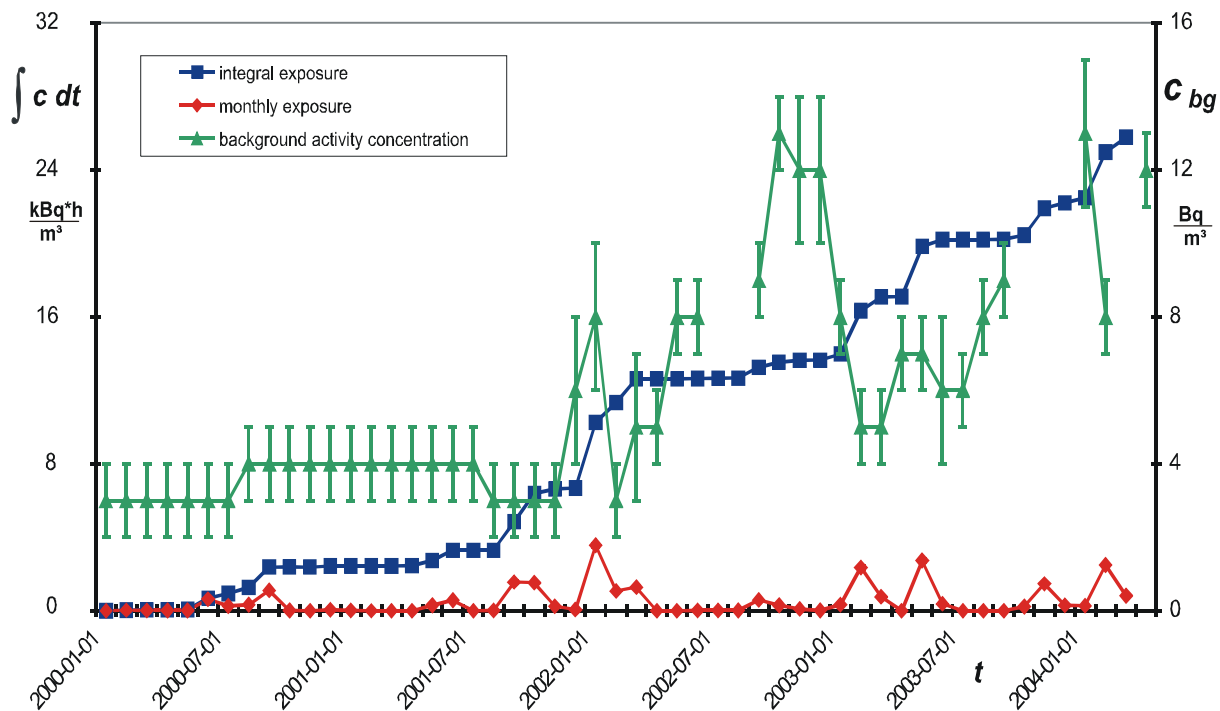


Figure 1: Background reading of the CD in comparison to the monthly integral (from 0 to 1 month) and overall integral activity concentration. The automatic background correction for this instrument is observable by the decrease of the background activity concentration twice a year.

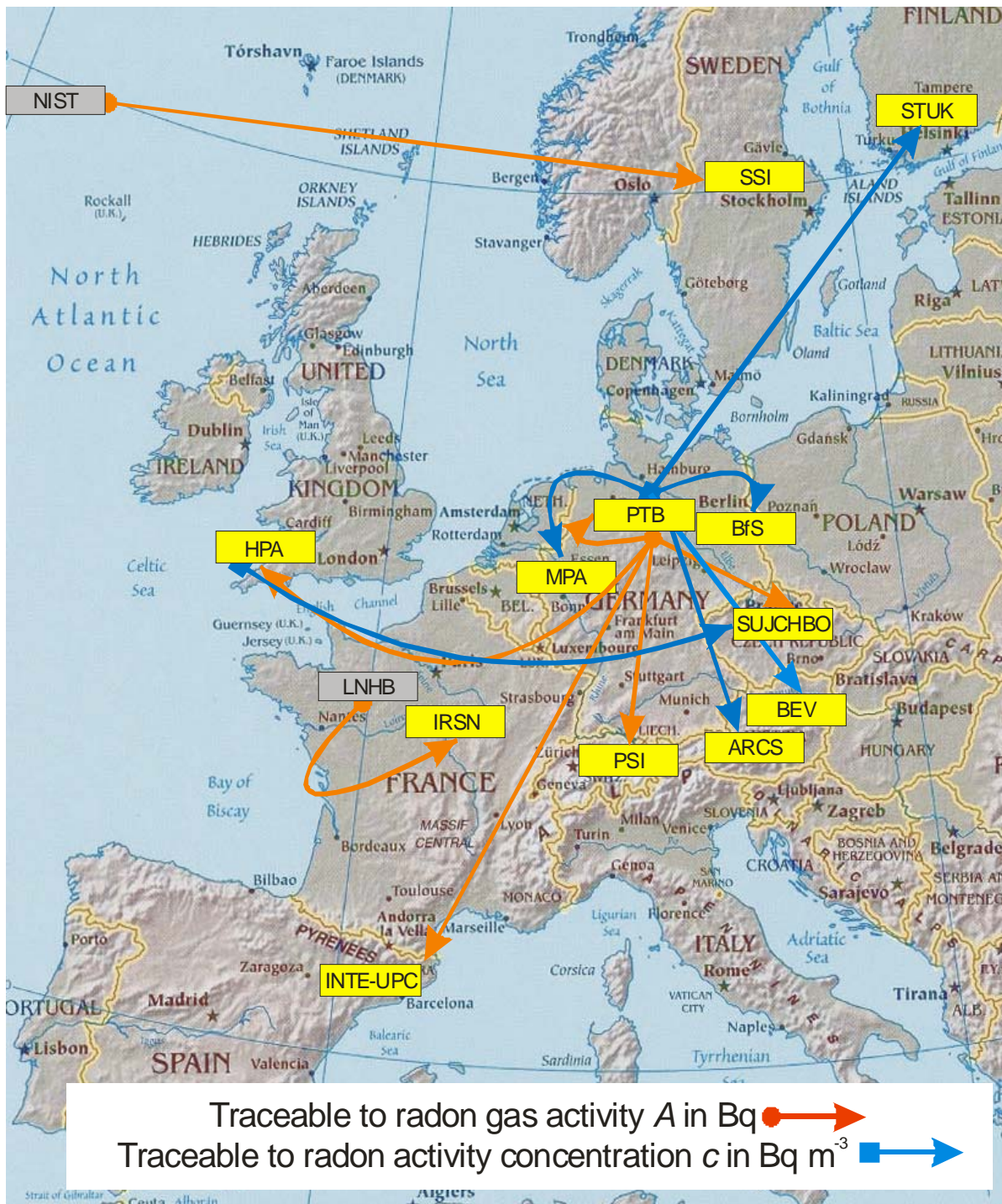


Figure 2: Traceability chains for the radon activity and the radon activity concentration of the participants. The arrows show the established transfer of the quantity, by sending from the institute at the starting point either a gas standard or a reference instrument to the institute at the ending point. Exception is SSI using a Ra-226 standard solution traceable to NIST as activity standard.

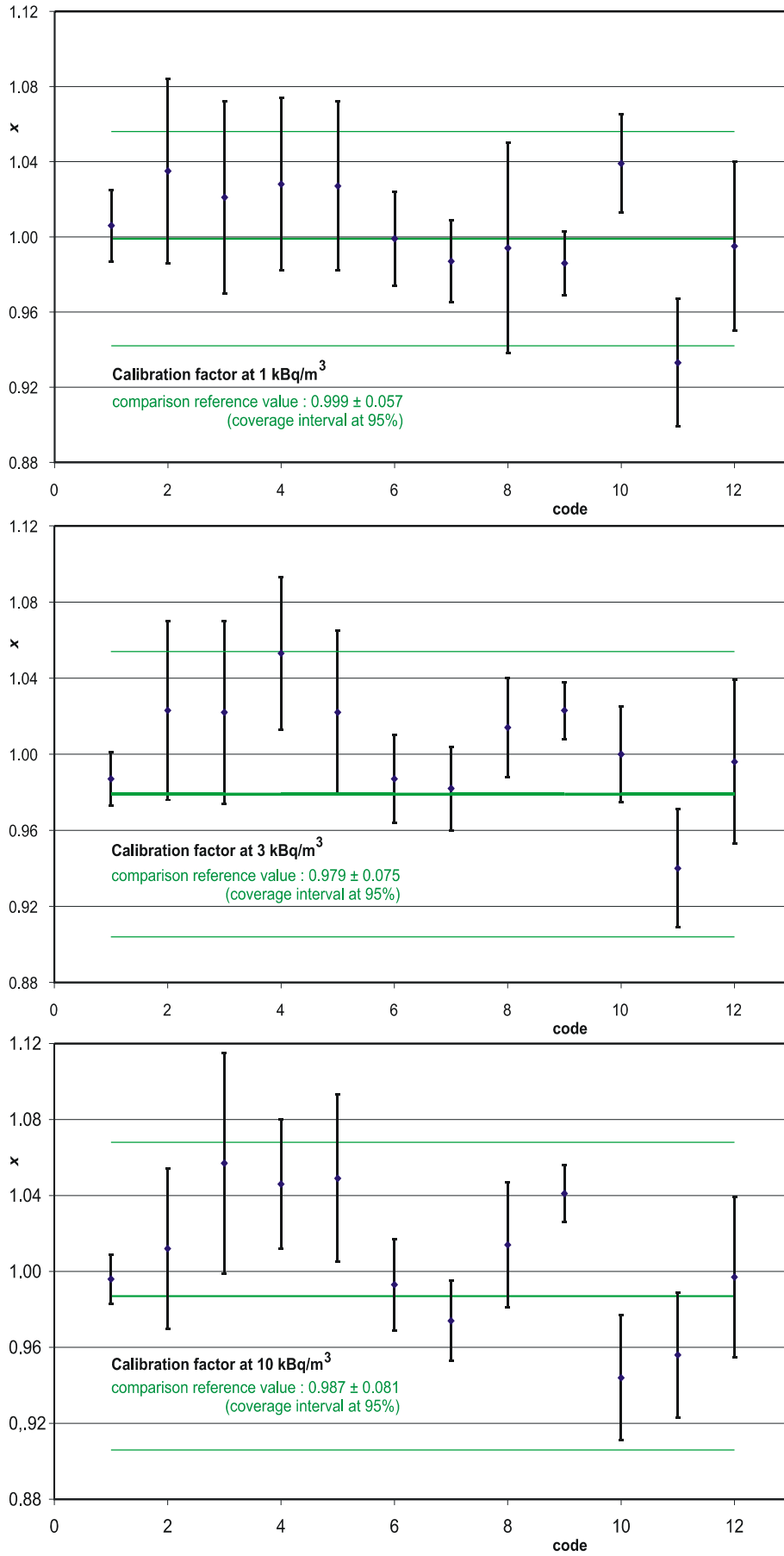


Figure 3: Calibration factors determined by the participants for 1 kBq/m³, 3 kBq/m³ and 10 kBq/m³ and the corresponding comparison reference value.

Table 1: Calibration factors obtained by the participants given as arithmetic mean, weighted mean and comparison reference value.

activity concentration	calibration factor k_c		
	arithmetic mean	weighted mean	comparison reference value
	with standard uncertainty	with standard uncertainty	with coverage interval at 95%
1 kBq m ⁻³	1.004 ± 0.028	0.998 ± 0.011	0.999 ± 0.057
3 kBq m ⁻³	1.004 ± 0.028	1.000 ± 0.009	0.979 ± 0.075
10 kBq m ⁻³	1.007 ± 0.035	1.011 ± 0.009	0.987 ± 0.081