

RESEARCH ARTICLE International intercomparison of Radon 222 activity concentration calibration facilities

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Abstract

The Coalition of International Radon Associations (COIRA) organised an inter-comparison of Rn-222 (radon) activity concentrations reported by calibration laboratories. A set of three AlphaGUARDs were used as transfer reference instruments against which to compare reported Rn-222 activity concentrations. Rn-222 activity concentration calibration facilities (sometimes termed chambers) from seven countries (Australia, US, Czech Republic, Spain, England, Sweden, Canada), and three continents participated in this project. The objective of the study was to provide information useful to calibration chamber operators and public health officials in the improvement of measurement and control systems, the maintenance of performance standards for measurements, and regulatory requirements for calibrations. This work builds upon and expands previous interlaboratory comparisons and provides data for estimating and using calibration uncertainty values as part of overall field error estimates and limits. A simple proportional difference between laboratories is presented here, calculated as the average for N hours of each hour's difference between the laboratory's concentration and the average of the three reference instruments. This percent difference ranged from less than 0.5% to just less than 8%. This work demonstrates that the ANSI/AARST standards limit of 8% for the estimated unexpanded (one sigma) individual calibration estimated uncertainty for continuous radon monitor calibration facilities in the US is achievable. However, given the few standards regarding calibration of Rn-222 activity concentration measurement instruments that themselves are often used to calibrate secondary and tertiary Rn-222 calibration facilities, there is a great need for continued interlaboratory comparisons to harmonise and document the calibration of Rn-222 activity concentrations.

Keywords: radon; radon chamber; radon reference levels; proficiency test; radon calibration; interlaboratory comparison test

he Consortium of International Radon Associations (COIRA) was founded in April 2015 by a group of international radon measurement and mitigation associations who saw benefits in establishing a collaborative community for sharing technical and policy information and collaborative projects. One of the first projects of COIRA was to continue the practice of assessing and publishing the degree of consistency of reference Rn-222 activity concentrations in air reported by calibration facilities from our respective countries. Such projects are vital to assist the calibration facilities improve their understanding of their own measurement and control systems as well as their various mechanisms for generating and controlling the reference radon activity concentration in air. In addition, regulatory programs for measuring radon activity concentration in air and mitigation assume that there is some limit on measurement

uncertainty, and such combined field uncertainty of radon measurements has as its minimum the initial calibration uncertainty associated with the estimated uncertainty associated with the standard reference Rn-222 activity concentration.

In the US, the commercial radon measurement industry obtains calibrations from a network of secondary and tertiary radon calibration facilities. The three secondary facilities in the US (all participants in this study) regularly compare Rn-222 activity concentrations using scintillation cells with the US primary authority, the US EPA National Analytical Radiation Environmental Laboratory. In most cases, US tertiary calibration facilities are used for in-house manufacturer research and development and interlaboratory comparisons with US secondary calibration facilities for a traceable chain of authority and documentation for their radon activity

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concentration in air measurement and reporting systems. These chambers and radon measurement and mitigation providers in the US participate in a voluntary accreditation program (the AARST National Radon Proficiency Program (1)), which includes policies to adhere to the ANSI/AARST standards for measurement performance (2) and quality assurance procedures for different measurement method categories (3). These standards provide guidance that, when followed, theoretically limits the resulting field uncertainty to 25% of known Rn-222 activity concentrations in air for a 48 h duration with any measurement method, based on a sequence of assumptions that are tested in periodic performance tests (1) required as a condition for continued certification. An irreducible component of estimated field error is the initial calibration error (estimated and reported as uncertainty), and the ANSI/AARST standard for Rn-222 measurement quality assurance practices requires a one sigma (unexpanded) uncertainty limit of 8% for the estimated calibration uncertainty. One method for estimating calibration uncertainty was applied to generate estimates and is used as a model for such estimates in other calibration facilities (4).

This study builds upon important previous work (5), which included 15 calibration facilities from European countries. The MetroRADON project evaluated the consistency of reference Rn-222 activity concentrations in air of 400, 1,000, and 6,000 Bq.m⁻³ as reported by the calibration facilities and as measured by the transfer instrument. The results showed a good consistency among all participants and a standard uncertainty of about 3% for the lower concentration range. The conclusions of the MetroRADON program included emphasis on the need for the international radon measurement community to perform this type of interlaboratory comparison on a regular basis, and for standards addressing interlaboratory comparison protocols.

The COIRA designed this study to include three continuous radon instruments together in a case with shortterm alpha track detectors as a Reference Instrument Package that was shipped from calibration facility to calibration facility. Although this study provides a wealth of data, this analysis is limited to the simple presentation of the relative performance of each calibration facility, using the hourly mean of the three Rn-222 activity concentrations as the hourly benchmark, and the mean over all the hours of the exposure as the percent difference from the benchmark. The AlphaGUARD DF2000 was chosen as the transfer instrument because of the demonstrated and published sensitive response, stability, proven ruggedness, and generous loan policy of the manufacturer, and does not imply that other devices could not be used in future. The same device model was selected for all three reference devices, but our recommendation for

future projects would be to repeat this project with other types/models of radon instruments in addition to this model.

Methods

The COIRA Interlaboratory Comparison Committee invited calibration facilities around the world to participate. Each facility was sent a copy of the scope of work, an agreement, and a link to a questionnaire regarding calibration facilities' descriptions. The information obtained included chamber specifications, signed agreements regarding the study, and dates for exposure at their facilities. In total, 14 commercial, national and research calibration facilities from seven countries (Australia, US, Czech Republic, Spain, England, Sweden, Canada), and three continents participated in this project, with several facilities including multiple concentration exposures and chambers. The participating calibration facilities and the specifications provided are listed in Table 1. Information was provided by the facility and reported as provided, including images of several participating facilities in Fig. 1.

The Reference Instrument Package included three AlphaGUARD DF2000 radon instruments from Bertin GmbH and a set of short-term Rapidos[®] alpha track detectors from Radonova Laboratories (AB) Sweden for use as transit detectors, as shown in Fig. 2. Although it would have been extremely surprising to identify exposure to high radon concentrations during shipping, and even more unexpected if this had any effect on the calibration facility exposures, the transit detectors were included to identify any significant radon exposures of the Reference Instrument Package other than in the calibration facilities' intended exposures.

The calibration facilities were scheduled by continent so that the Reference Instrument Package was shipped between all the participating calibration facilities in one country before travelling to the next, and then to the next continent. The calibration facility's operating costs during the exposure, and shipping costs to the next facility, were paid by each facility, so that the cost of the project was shared among participants. Additional administrative costs were paid by participating national and international radon associations. The AlphaGUARD reference devices and protective shipping box were generously provided by Bertin GmbH, and Radonova Laboratories AB provided the Rapidos[®] alpha track detectors.

Each calibration facility received the Reference Instrument Package with instructions to verify and note the satisfactory conditions of the equipment and the date/ time, seal the alpha track detectors that were exposed during that segment of the journey, and ship them to Radonova for analysis. The instructions for exposing the three transfer reference AlphaGUARDs included:

Table 1. Participating calil	pration facilit	y specificatic	Suc						
Calibration Facility Chamber (Country)	Number of chambers	ISO 17025 accredited	Volume [m ⁻³]	Temperature range [C]	Relative Humidity range [%]	Typical pressure/ adjustable	Range of stable radon concentration [Bq m ^{.3}]	Radon source	Chamber's reference instrument
Environment Protection Authority South Australia (Australia)	_	or	4. 1.	ambient – ~25	30-90	Ambient; not adjustable	0-12,000	²²⁶ Ra in HCI from NIST	AlphaGUARD PQ2000Pro
The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) (Australia)	-	°Z	16.47	16–28	30–80	Atmospheric, not adjustable	200-8,000	Uranium ore	Atmos 12 DPX
Kansas State University (KSU) Radon Chamber at Engineering Extension (United States)	-	õ	0.57	15-26	15–75	Atmospheric – no	370-5,900	Pylon 1025 No A-344, solid metal, sealed flow through source for radon chamber.	2 Pylon AB 5 and 2 scintillation cells with Ludlum Counters
National Institute for Nuclear, Biological and Chemical (NBC) Protection (Czech Republic)	-	yes	0	15–25	20-90	970 hPa, Not adjustable.	150-1,000,000	The main sources are: 1) special (36.8 mg ²²⁶ Ra); 2) Pylon RN-1,025; 3) CMI RF 2000	Scintillation cells (1 L) FASA and NP420L; AlphaGUARD DF2000 and PQ2000
National Radiation Protection Institute (Czech Republic)	2	Yes	0.15-48	12-45	25–92	Pressure cannot be adjusted	100-100,000	Different ²²⁶ Ra/ ²²² Rn sources type flow pass through	Scintillation cells, RAD 7 and AlphaGUARD
TCS Industries Inc. (United States)	-	ou	_	16–29	25–80	7.6 mm water gage	200–2,200	Pylon 1025 flow through cell	Pylon AB5 and 300A cells compared with the USEPA + Pylon 3150A Ra-226 doped Cell
University of Cantabria (Spain)	_	٥N	54	10–25	40–60	950 hPa not adjustable	100-100,000	The soil under the laboratory	AlphaGUARD PQ2000 and ATMOS12
UKHealth Service Agency (UKHSA) (England)	2	No	43	Ambient 18–30	30–50	Ambient atmospheric	200-9,000	3 x Pylon dry passive emanation source, 1 x CMI flow through source	Atmos I2 DPX + AlphaGUARD PQ2000
Swedish Radiation Safety Authority (Swedish)	2	No	#1:11 m ³ #2:27 m ³	17–23	Not controlled	Ambient atmospheric	150-20,000	²²⁶ Ra flow through source	AlphaGUARD DF2000 and PQ2000 Pro
Radiation Safety Institute of Canada (Canada)	-	° Z	10.7	20-30	20-80	960 mbar	150-500,000	Pylon RN-1025 gas flow through 87 kBq source and Pylon Rn-1025 gas flow through 4.07 MBq source	AlphaGUARD DF2000 and Pylon 300A Lucas Cells
Pylon Electronics Incorporated (Canada)	-	ou	39	20–25	15-40	100 to 102 kPa	250-15,750	6 sources (Pylon model RN-1025 – flow-through sources)	3 × Pylon AB-5 monitors (averaged)
Bowser-Morner, Inc. (United States)	2	о Х	10.7	#l: I5–30 # 2: 20–22 C	: #l: 20–80, # 2: 49–51	: 980–990 mbar (hPa)	#1:150–2,220, #2,880–960	Pylon flow-through sources, Model 1025, Chamber 1 238 and 500 kBq, Chamber 2 555 kBq	Custom monitoring system,



Fig. 1. Images from participating radon chambers.



Fig. 2. Reference instrument package.

- Exposure Period: 2–5 days; with the goal of at least 48 h after a minimum 4-h equilibration period,
- Exposure activity concentration: 200 Bq·m⁻³ to 40 kBq·m⁻³,

- If the calibration facilities included chamber control of temperature or relative humidity, exposures were conducted in an environment of 20°C (18–22° C) and 35% RH (30–40% RH), and measured at least hourly,
- If there were no controls for temperature or relative humidity, these were recorded at hourly intervals and maintained as constant as possible during the duration of the exposure.

At the end of the exposure period, calibration facilities sent their recorded hourly values for the radon activity concentration, temperature, and relative humidity from the chamber's (own reference) instrumentation, and the AlphaGUARD output data files (*.upf2) to the project committee via email. Furthermore, and as described in the project standard operating procedures, the calibration facility opened a set of three new Rapidos[®] alpha track detectors, noted date/time, and placed these in the Reference Instrument Package, which were then included in the shipment to the next calibration facility.

After all the participating calibration facilities had completed their measurements, the devices were shipped to a member of the project committee for additional exposures and analysis, where the devices were compared again in a very low concentration (outdoor) radon environment.

After all the data were received from the calibration facilities, the project committee prepared the data for analysis and interlaboratory comparison and created a private online link to individual folders containing all materials and data provided by each facility. This allowed the officials from each facility to review only their original and summarised data, verify assumptions about time zones, date/times, and most importantly, confirm the exclusion of initial hours, if any, during instrument equilibration in each chamber. All participating calibration facilities were invited to two online meetings where interim results and progress reports were presented, and calibration facilities were able to ask questions or identify any clarifications regarding their data or its interpretation. All original data, assumptions, and summary statistics were available and reviewed by each calibration facility and were consistent with the interpretations of the project committee.

This project was designed to provide transparency and consistency for the participants, so included written procedures and chain of custody documentation. Upon receipt at the calibration facility, the chamber staff was responsible for the handling and data acquisition from each instrument, following the procedures and documenting the exposure on forms which were photographed and sent via email, with original paper custody forms in the shipping container, to the COIRA Committee after completion of the chamber exposures and shipping. Data and treatment protocols (6) were available, reviewed, and consistent between all participants.

In addition to standardising the procedures as much as possible, the project included measures to secure, track, and verify measurement data, as well as post study exposures to verify instrument stability and lack of accrued background during the duration of the interlaboratory comparison. These procedures and assessments included the following:

- The AlphaGUARDs were shipped with unlocked screen and keys, enabling participating facility operators to verify each instrument's settings before the exposure (e.g. 10-min diffusion mode, date, temperature, relative humidity and atmospheric pressure sensors operational, etc.),
- Alpha track detectors were used as transit blanks, with a set of three exposed during each segment of the journey. Results are as expected, with exposures in all cases less than 50 Bq·m⁻³.h⁻¹.
- Although the study extended to more than 2 years in duration and the radon reference instruments were not recalibrated during this time, the instrument model was selected in part to its advertised calibration stability for 5 years (7, 8). Furthermore, it should be stressed that the analyses included an evaluation of any change in the relative response of each reference instrument relative to the others in the Reference Instrument Package during the whole duration of the study. The initial calibration of each instrument was in accordance with the manufacturer's customary practices and provided

an assurance of an unexpanded (one sigma) estimated uncertainty of 3% as documented by interlaboratory comparisons with the primary Rn-222 activity concentration authority (9) of the German Federal Office for Radiation Protection (BfS).

- Each participating facility was provided a private online folder hosting all the chamber-specific information, with the open-source software script (6) used to analyse their data, and resulting summary statistics, enabling chamber operators to thoroughly review all material and assumptions, before any data analysis was conducted,
- Simple summary statistics were generated, with the summary files for each participating calibration facility saved in their private folders, enabling complete reproducibility of all derived statistics,
- At the end of the project, a fourth AlphaGUARD, provided by the US EPA and calibrated by Bowser Morner in the US to concentrations generated by the USEPA (10) was used as a comparison instrument for additional exposures in outdoor tented low concentrations, and higher indoor concentrations. These exposures (with the 4th transfer standard measurement result signified by instrument USEPA-BMI in the Figures and Table) were a final test of reference instrument stability and final comparison to another comparable instrument, calibrated against a different primary authority, as well as an evaluation of any background accrued in the transfer reference instruments during their journeys.

Results and discussion

Although this study produced a wealth of data, this initial analysis only compares the response of each chamber to the response of the three transfer reference instrument (equations 1-3), evaluates the results as a function of concentration, and recommends expanding future interlaboratory comparisons.

The mean of the three transfer reference instruments for each hour is represented by r_h , and individual transfer reference instruments results are r_i , as shown in Equation 1.

$$r_h = \frac{1}{3} \sum_{i=1}^{3} r_i \tag{1}$$

The proportional difference between the transfer reference instruments' hourly mean and the laboratories reported hourly mean is given by a defined *proportional differences* d_h shown in Equation 2, where l_h represents the Rn-222 activity concentration in air reported by the laboratory for each hour.

 d_h therefore represents the difference between the laboratory and the reference instrument package, as a

proportion of the reference instrument package's concentration, for each hour.

$$d_h = \left(\frac{l_h - r_h}{r_h}\right) \tag{2}$$

The statistic parameter D, given in Equation 3, represents the mean of the hourly proportional differences d_h , where N is the total number of hours of the exposure.

$$D = \frac{1}{N} \sum_{h=1}^{N} d_h \tag{3}$$

This simple comparison statistic of the parameter D is presented in Table 2 with pertinent characteristics of each exposure. The D parameter for each exposure is represented in Fig. 3.

The proportional difference (D) values for each exposure are shown in Table 2.

This analysis also included evaluating the relative responses between the three transfer reference instruments during the duration of the study and conducting final interlaboratory comparisons collocated with a fourth AlphaGUARD that had been calibrated at a commercial US facility (10). This fourth instrument was collocated with the three AlphaGUARDs in the Reference Instrument Package to provide more data at very low environments and is represented by USEPA-BMI in the Table and Figures. All the exposure data was evaluated to determine whether there was evidence of: 1) nonlinear instrument response to concentration, 2) a change over time in relative response between the individual reference instruments during the study, and 3) an effect due to background accrued during the study. All data is presented for further evaluation and with limitations described.

Figure 4 presents D as a function of the *mean* chamber concentration during the exposure as a representation of possible (non-linear) effect of concentration on the statistic D. As can be seen, the evidence does not indicate an effect of concentration on D, with a near zero slope. (slope with all values is 1.5E-7, and the slope without the highest concentration exposure is 1.6E-7.) Error bars represent the sample standard deviation of each D value.

This analysis included a basic assessment of whether individual transfer reference instruments response functions changed relative to one another over time. We compared individual transfer reference instrument responses relative to reported chamber concentrations for the entire duration of each exposure to obtain a statistic for the *individual* transfer reference instrument result. The Reference Instrument Package included AlphaGUARDs with serial numbers 105, 106, and 107, and each is represented in Fig. 5. This Figure presents individual statistics for each transfer reference instrument in an attempt to reveal any consistent change between exposures in the relative responses of each transfer reference instrument.

Table 2. Summary of results (proportional difference from the benchmark as represented by D) by chamber ID and in time sequence during the 2 years of the study, including the maximum and minimum of the concentration during each exposure

ID	N hours in exposure [h]	D [equation 3]	Standard deviation of D	Mean concentration [Bq m ⁻³]	Minimum [Bq m ⁻³]	Maximum [Bq m ⁻³]
D	72	-0.002	0.019	1247	1121	1314
С	72	-0.013	0.023	899	844	1003
J	118	0.066	0.017	4528	4400	4614
E	447	-0.009	0.043	1374	1048	1552
К	97	0.015	0.035	1284	1044	1415
R	96	-0.006	0.029	1574	1458	1643
F	67	0.016	0.043	1365	1110	1528
н	114	0.001	0.021	4696	4352	4992
Q	45	0.004	0.022	7888	7644	8116
Ν	92	0.014	0.019	5897	5589	6277
М	115	0.002	0.058	929	477	1005
L	108	0.026	0.088	181	138	256
0	41	0.042	0.020	10564	8277	11104
1	38	0.011	0.055	40679	2330	99973
G	90	0.033	0.040	1330	988	1451
Р	70	0.076	0.027	3882	2957	4980
В	65	0.038	0.031	2045	1881	2187
A	229	-0.062	0.028	3046	1429	4195
FF4	211	0.019	0.077	1338	75	2523



Fig. 3. Proportional difference D between chamber and reference instrument package with standard deviation of hourly D values as error bars.



Fig. 4. Mean proportional difference D as a function of mean Rn-222 activity concentration.

As can be seen in Fig. 5, while there may be a lack of homogeneity or other problems in one of the chambers (F), the average of the three reference AlphaGUARDs in that chamber (F) still resulted in a proportional difference D within the range of the other chambers, as shown in Table 2.

Additional evaluations of these data are recommended, including an assessment of bias between the three transfer reference instruments. However, for the purposes of establishing a stable benchmark against which to compare between laboratories, these data exhibited no significant change in relative reference instrument response during the



Fig. 5. D statistics for each individual transfer reference instrument.

study. (An additional conclusion is the recommendation that this stability and interlaboratory comparison procedures be conducted at the beginning as well as the conclusion of any future interlaboratory comparison project.)

After participant exposures were concluded, a fourth AlphaGUARD was incorporated into the project, loaned from the Tribal Air Monitoring Support Center (11) which offers within-calibration equipment loans for US federally recognized Tribal Nations. This identical DF2000 model was calibrated (independently under a contract by the US EPA) by one of the calibration facilities in the project but does provide some information on the comparability of the final responses of the three transfer reference instruments. Three sets of exposures were conducted in an outdoor environment (tent), within which all four AlphaGUARDs were collocated within an encasement of three layers of 0.1 mm plastic to eliminate the possibility of dust contamination. The results of these 2-day exposures are shown in Fig. 6.

The three transfer reference instruments performed well in the low concentration environment, demonstrating no consistent bias between instruments in the low Rn-222 activity concentration, as shown in Fig. 6.

Conclusions

Reference atmospheres are used to calibrate Rn-222 activity concentration measurement systems that establish the validity of hundreds of thousands of measurements and public and individual health decisions. This study, as well as future (and prior) interlaboratory comparisons are vital to develop a shared understanding of operations of stable Rn-222 activity concentration reference atmospheres and for the harmonisation of calibration measurement systems. Continued estimations of the consistency between different reference atmospheres provide data used to improve calibrations and to assure public health officials that mitigation decisions are based on measurement methods with limits on uncertainty that are both theoretically calculated and estimated from actual interlaboratory comparisons such as the one reported here. Protocols and quality assurance requirements in various jurisdictions in the US are based upon limiting field measurement error, a fundamental component of which is the original calibration uncertainty, as estimated by the calibrating authority and reported as calibration uncertainty. Two standards developed by ANSI/AARST and used as the basis for proficiency testing by the US radon measurement industry trade association use a value of 8% for the unexpanded (one sigma) upper limit on calibration uncertainty reported with exposures of 48 h or longer, estimated in accordance with industry practice (4). This upper bound of 8% was an estimated limit generated by the calibration facility operators serving on the MS-QA standard committee, but the results of this COIRA study indicate that this initial limit is reasonable, and perhaps can be reduced through further collaboration on calibration normalization. While the maximum D statistic from this study is not directly comparable to the maximum deviations measured in the MetroRADON interlaboratory comparison, the present interlaboratory comparison produced results which is not inconsistent with the MetroRADON study. The present study design included additional opportunities for interferences incurred during shipping between three continents, a much larger range of chamber sizes and configurations, and lasted for many more months.

Although this project does not evaluate individual calibration uncertainty estimates, it does provide an



Fig. 6. Low Rn-222 activity concentrations measured with the three reference instruments and a fourth collocated AlphaGUARD, May 2022.

upper estimate of the maximum deviation that one internationally recognized calibration laboratory could reasonably expect to differ from another internationally recognized calibration facility, thereby informing policy makers of the current harmonization status of Rn-222 activity concentration reference atmospheres.

Regulatory and business interests coincide in encouraging future and expanded interlaboratory comparisons. Because the results are presented anonymously and without identification, individual calibration facilities may choose to publish their results and use the information to improve accuracy or for other purposes unaffiliated with COIRA.

It is important to note that the choice of the transfer reference instrument model and the number of instruments used was not based on an exhaustive determination of the best equipment for this purpose but was opportunistic and limited to three devices due to the size of some of the calibration facilities participating in this study. More interlaboratory comparisons provide more opportunities for normalizing Rn-222 activity concentrations in air between calibration facilities, thereby enabling increased accuracy in decisions impacting individual and collective public health.

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