

INTERCOMPARISON OF RADON AND DECAY PRODUCT MEASUREMENTS IN AN UNDERGROUND MINE AND EPA RADON LABORATORY: A STUDY ORGANIZED BY THE IAEA INTERNATIONAL RADON METROLOGY PROGRAMME

G. Budd,* R. Hopper,* E. Braganza,* M. Ronca-Battista,* F. Steinhäusler,[†] and P. Stegner[‡]

Abstract—The International Atomic Energy Agency (IAEA) in Vienna and the European Union (EU) in Bruxelles formed the "International Radon Metrology Programme" (IRMP, scientific secretary: F. Steinhäusler, University of Salzburg, Austria). The IRMP is designed to assess and foster the improvement of radon and decay product measurements that are made around the world. Within the framework of the IRMP, the U.S. Environmental Protection Agency Radiation and Indoor Environments National Laboratory (EPA) in Las Vegas, Nevada, organized jointly with the U.S. Bureau of Mines an international intercomparison exercise at a former uranium mine (Twilight Mine, Colorado) and the EPA Radon Laboratory. The main objective of this exercise was to compare radon and radon decay product instruments under both well-controlled as well as widely fluctuating exposure conditions. The laboratory exposures occurred under relatively steady radon and decay product conditions, with a moderate equilibrium ratio, while the conditions in the mine fluctuated greatly and the equilibrium ratio was low. An additional purpose of the exercise was to provide a forum for manufacturers and measurement organizations worldwide to exchange information and plan improvements in their operations and calibration programs. Altogether 19 organizations from seven countries intercomparing 32 different radon and radon decay product instruments participated in this exercise. This paper summarizes the results from the analysis of the experimental data obtained in the Bureau of Mines Twilight Mine in July of 1994, as well as the results from the EPA Radon laboratory in August of 1994.

Health Phys. 75(5):465-474; 1998

Key words: calibration; decay products; radon; instrumentation

INTRODUCTION

WITHIN THE framework of the International Atomic Energy Agency (IAEA)-European Union (EU) International Radon Metrology Programme (IRMP) an international exercise to evaluate radon and radon decay product measurement instruments was organized jointly with the U.S. Environmental Protection Agency Radiation and Indoor Environments National Laboratory (EPA) in Las Vegas, Nevada, and the former U.S. Bureau of Mines (BOM). The primary objective of the exercise was to compare the performance of radon and radon decay product measurement instruments from around the world under both laboratory and field exposure conditions. The results of this intercomparison has produced data useful to those performing radon and radon decay product measurements or using the results of such measurements in their work.

A total of 19 organizations from seven countries participated in the exercise with 32 types of radon and radon decay product measurement instruments. The exercise was conducted in both controlled laboratory and uncontrolled underground mine environments. The laboratory exposures were conducted within an environmental radon chamber at the EPA's Radon Laboratory in Las Vegas. These exposures were designed to compare the instruments under very stable, controlled environmental conditions at relatively low concentrations of radon and radon decay products. The field exposures were conducted at a former underground uranium mine in Colorado maintained by the BOM. These exposures were designed to compare the instruments under a fluctuating and uncontrolled environment. A description of the participants and equipment used at the EPA Laboratory appears in Table 1; Table 2 presents the participants and equipment used at the BOM underground mine. The work described in this report builds upon earlier intercomparison exercises conducted in a variety of locations and environments and should be considered in their context

* U.S. Environmental Protection Agency Radiation and Indoor Environments National Laboratory, Las Vegas, NV 89193-8517; [†] International Radon Metrology Programme, Institute of Physics and Biophysics, University of Salzburg, A-5020 Salzburg, Austria; [‡] International Atomic Energy Agency, Wagramerstrasse 5, P.O. Box 100, A-1400 Vienna, Austria.

For correspondence or reprints contact: G. J. Budd, Center for Indoor Environments, EPA Radiation and Indoor Environments National Laboratory, PO Box 98517, Las Vegas, NV 89193-8517.

(Manuscript received 28 December 1997; revised manuscript received 21 April 1998, accepted 14 June 1998)

0017-9078/98/\$3.00/0

Copyright © 1998 Health Physics Society

Table 1. Participants in the IAEA International Intercomparison Exercise at the EPA Las Vegas Radon Laboratory.

Code	Participant	Country	Measurement	Equipment description
Algade 2	Algade Boîte Postale 46 87250 Bessines-sur-Gartempe	France	Integrated radon decay products	Site Alpha Dosimeter, incorporating LR-115 for gross alpha detection and a continuous AC driven pump
Algade 3	Algade Boîte Postale 46 87250 Bessines-sur-Gartempe	France	Integrated radon decay products	Personal Alpha Dosimeter, incorporating LR-115 and a battery operated pump
Algade 4	Algade Boîte Postale 46 87250 Bessines-sur-Gartempe	France	Grab radon decay product	MEAP III unit incorporating a rotary pump, cellulose filter, and silicon junction detector. Unit uses Rollé method.
ASpectra1	Alpha Spectra 210 Clayton Street Denver, CO	USA	Quasi-integrated radon	7.6 cm open face charcoal adsorber
ASpectra2	Alpha Spectra	USA	Quasi-integrated radon	7.6 cm diffusion barrier charcoal adsorber
ASpectra3	Alpha Spectra	USA	Integrated radon	Alpha Film using filtered CR-39 film
BRI	Building Research Institute Warsaw	Poland	Quasi-integrated radon	Pico-Rad charcoal vial analyzed via Packard Tri-Carb 1900TR liquid scintillation analyzer
CLRP1	Central Laboratory for Radiological Protection Warsaw	Poland	Integrated radon	Electret ion chambers manufactured by Rad-Elec, LSTB configuration
CLRP2	Central Laboratory for Radiological Protection Warsaw	Poland	Quasi-integrated radon	Pico-Rad charcoal vial analyzed via Packard 1900TR liquid scintillation analyzer
CLRP3	Central Laboratory for Radiological Protection Warsaw	Poland	Integrated radon	Filtered CR-39 alpha track detector assembled and analyzed by participant
Genitron	Genitron Instruments Heerstrasse 149 Frankfurt D-60488	Germany	Integrated radon	Genitron AlphaGuard pulsed ion chamber with digital signal processing
Honey	Sun Nuclear 425-A Pineda Ct. Melbourne, FL 32940	USA	Integrated radon	Sun Nuclear 1023 incorporating a diffused junction detector within a filtered chamber
INCT	Institute of Nuclear Chemistry and Technology Warsaw	Poland	Grab radon decay products	RGR-30 Radon Mining Monitor incorporating a pump, fiberglass filters, and a surface barrier detector using the Markov method. Assembled by participant.
INP	Institute of Nuclear Physics Cracow	Poland	Integrated radon	Filtered CR-39 alpha track detector assembled and analyzed by participant
MAB	Munchener Apparatebau für Electronische Gerate Mehlbeerenstrasse 2 Taufkirchen 82024	Germany	Grab radon	LLMS 500 unit incorporating a beta detector, pump, and filter. Manufactured by participant.
MAIB	Medical Academy in Bialystock	Poland	Quasi-integrated radon	Pico-Rad charcoal vial analyzed via Packard Tri-Carb 1900TR liquid scintillation analyzer
NIH	National Institute of Hygiene Warsaw	Poland	Quasi-integrated radon	Pico-Rad charcoal vial analyzed via Packard 1900CA Tri-Carb liquid scintillation analyzer. Manufactured by Packard Instruments, 800 Research Parkway, Meriden, CT 06450.
Rad-Elec	Rad-Elec Frederick, MD	USA	Integrated radon	Short-term electret and "S" chamber
TN	Thompson and Nielsen	Canada	Grab radon decay product	TN-IR-21; silicon junction detector; Rollé analysis
UOZ	University of Zagreb	Croatia	Integrated radon	Participant-assembled LR-115 device

(George and Tu 1988; Knutson 1988; OECD 1980, 1986; Pearson 1989).

MATERIALS AND METHODS

Environmental Protection Agency Radon Laboratory

The EPA's Radiation and Indoor Environments National Laboratory in Las Vegas, Nevada, maintains and

operates three environmental radon chambers. These chambers range in volume from 10.45 m³ to 22.58 m³ and are walk-in units with multiple sampling ports. This configuration allows a variety of instruments to be exposed simultaneously by either being placed within a chamber or by drawing chamber air through sampling ports. Each chamber has measurement and feedback mechanisms for the regulation of temperature, humidity, and radon concen-

Table 2. Participants in the IAEA International Intercomparison Exercise at the BOM Twilight Mine.

Code	Participant	Country	Measurement	Equipment description
AECB	Atomic Energy Control Board	Canada	Grab radon decay product	Pump and filter; analysis via Modified Tsivoglou method
Algade 1	Algade	France	Integrated radon	BARASOL active monitor using silicon junction detector
Algade 2	Algade	France	Description in Table 1	
Algade 3	Algade	France	Description in Table 1	
Algade 4	Algade	France	Description in Table 1	
ASpectra1	Alpha Spectra	USA	Description in Table 1	
ASpectra2	Alpha Spectra	USA	Description in Table 1	
ASpectra3	Alpha Spectra	USA	Description in Table 1	
BRI	Building Research Institute	Poland	Description in Table 1	
CLRP1	Central Laboratory for Radiological Protection	Poland	Description in Table 1	
CLRP3	Central Laboratory for Radiological Protection	Poland	Description in Table 1	
CMI1	Laboratory of Radiometry Central Mining Institute 40-166 Katowice, Gwarkow1	Poland	Quasi-integrated radon	Charcoal absorber analyzed via liquid scintillation
CMI2	Laboratory of Radiometry Central Mining Institute	Poland	Integrated radon decay product	ALFA-31 unit incorporating a pump, TLD and filter assembled by participant
CMI3	Laboratory of Radiometry Central Mining Institute	Poland	Grab radon decay product	RGR-13 active unit; Markow analysis
EPA1	U.S. EPA Radiation and Indoor Environments National Laboratory, Las Vegas, NV	USA	Integrated radon	NITON RAD-7 active monitor electrostatically collects alpha emitters on a solid state detector
EPA2	U.S. EPA	USA	Integrated radon decay products	Scintrex Working Level Monitor using solid state detector
Genitron	Genitron Instruments	Germany	Description in Table 1	
Ghana	Republic of Ghana	Ghana	Integrated radon	Participant-assembled LR-115 device
INCT	Institute of Nuclear Chemistry and Technology	Poland	Description in Table 1	
INP	Institute of Nuclear Physics	Poland	Description in Table 1	
MAB	Munchener Apparatebau für Electronische Gerate	Germany	Description in Table 1	
MAIB	Medical Academy in Bialystock	Poland	Description in Table 1	
NIH	National Institute of Hygiene	Poland	Description in Table 1	
NIOM1	Nofer Institute of Occupational Medicine	Poland	Integrated radon	Participant-assembled LR-115 device
NIOM2	Nofer Institute of Occupational Medicine	Poland	Integrated radon decay product	Participant-assembled LR-115 device
NIOM3	Nofer Institute of Occupational Medicine	Poland	Grab radon decay product	RGR-30 active unit incorporating a semiconductor alpha detector; Markov analysis
NIOM4	Nofer Institute of Occupational Medicine	Poland	Grab radon decay product	RGR-13 active unit incorporating a semiconductor alpha detector; Modified Tsivoglou analysis
TN	Thompson and Nielsen	Canada	Description in Table 1	
UOZ	University of Zagreb	Croatia	Description in Table 1	

trations. In addition, condensation nuclei in the form of minerals from tap water[§] can be incorporated into the atmosphere and used to change and stabilize concentrations of radon decay products. All exposures for this intercomparison exercise were conducted in Chamber A which has an internal volume of 18.41 m³. A photograph of the chamber appears in Fig. 1.

Temperature is controlled in the chamber by a suspended metal plate through which either refrigerant or hot gas is flowed. Humidity is controlled using a humidifier and a dehumidifier. Both temperature and relative

humidity are measured continuously^{||} and adjusted as necessary by a computerized monitoring system. A continuous recirculation system provides a steady flow of air allowing for uniform mixing within the chamber.

Radon is produced by radioactive decay in radium sources[¶] that are calibrated against National Institute of Standards and Technology (NIST) standards. These

^{||} Digital Humit-temp 2200D, manufactured by Phys-chem Scientific Corp., 36 West 20th Street, New York, NY 10011.

[¶] Pylon RN-1025, manufactured by Pylon Electronic Development Co. Ltd., 147 Colonnade Road, Ottawa, Ontario, K2E 769, Canada.

[§] Atomizer custom-made by the EPA.

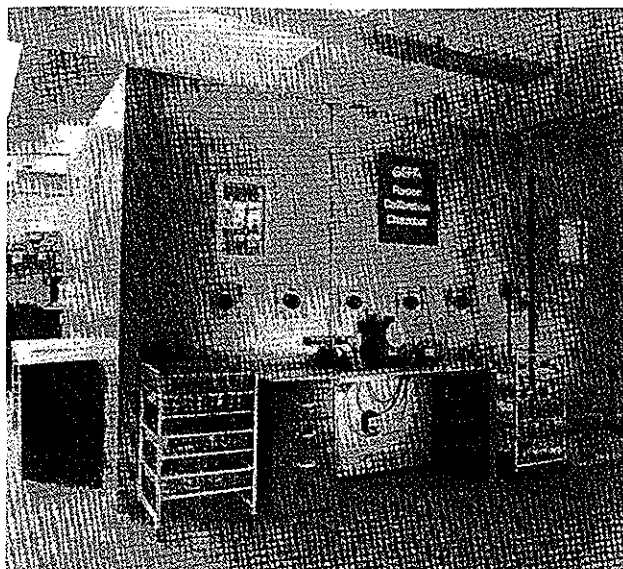


Fig. 1. EPA environmental radon chamber "A."

sources are integrated into the air flow mechanisms of each chamber. A constant volume (300 L h^{-1}) of dry air is passed through the sources to flush radon from them. The radon-laden air is divided into two streams and passed through mass flow controllers. There are two mass flow controllers: one controls air into the chamber and the other controls flow into a vent stack. Radon that does not go into the chamber is vented to the atmosphere. The mass flow controllers are regulated by a control box which in turn is controlled externally by the computer monitoring system. The computer adjusts the chamber and stack mass flow controllers via the control box to maintain the desired concentration of radon within the chamber. This is done by varying the ratio of radon-laden air going into the chamber and the stack.

The radon concentration during the intercomparison study varied from 605 to 832 Bq m^{-3} , the relative humidity varied from 45 to 51%, and the temperature varied from 20 to 23°C . The radon decay product concentration during the intercomparison varied from 1.25 to $2.08 \mu\text{J m}^{-3}$ (0.06 to 0.10 WL), with an equilibrium factor ranging from 37 to 50%, with a mean of 46%.

Radon measurement system used by the EPA

The primary radon measurement system used by the EPA Radon Laboratory to determine radon concentration within the chambers are 0.36 L scintillation cells* [also termed Lucas cells (Lucas 1957)]. The cells are evacuated, filled with ambient chamber air, and then analyzed using 7.62-cm-diameter photomultiplier tubes. This primary measurement system is traceable by two separate

* Lucas Scintillation Cells Model RA 1000-1, manufactured by Rocky Mountain Scientific Glass Blowing Company, 4990 E. Ashbury Avenue, Denver, CO 80222.

performance tests to the primary ^{222}Rn measurement system used by NIST (U.S. EPA 1996).

First, NIST provides the EPA Radon Laboratory with multiple spherical glass sample ampules each containing an activity of ^{222}Rn gas known only to NIST. The radon contained within the ampules is produced at NIST from their ^{226}Ra Standard Reference Material (SRM) and is directly relatable to the NIST primary measurement system (Colle et al. 1990). The EPA staff dilutes each radon sample ampule with a known quantity of air, and then using a transfer manifold system transfers the diluted radon sample to a series of EPA scintillation cells (Lucas and Markun 1988). The cells are analyzed to determine the activity of radon within the sample ampules and the results are reported to NIST. Reports of traceability are issued from NIST to the EPA for each of the sample ampules and serve to assess any bias between the two laboratories.

A second performance test requires EPA to fill a series of EPA scintillation cells with ^{222}Rn gas from the EPA's Radon Laboratory standard radon emanation flasks. These flasks contain a known activity of ^{226}Ra solution obtained from NIST and are used to generate ^{222}Rn gas for the routine calibration of the EPA scintillation cells. In this test the cells are filled with radon at the EPA Radon Laboratory, analyzed using the EPA cell counting system, and then shipped to NIST for analysis. NIST transfers the contents of the cells to a pulse ionization chamber to determine the activity of radon within each cell. The results of the EPA and the NIST measurements are compared and reports of traceability are issued from NIST to the EPA for each of the EPA scintillation cells. This intercomparison produces a second estimate of bias between the two laboratories and allows the EPA Radon Laboratory to validate the performance of its scintillation cell calibration system.

As a result of these performance tests, estimates of bias between -0.98 and 2.37% (measured as the percent difference from NIST) have been reported by NIST (U.S. DOC 1996). The bias between the two laboratories is tracked over time by EPA and decisions regarding changes in procedures or equipment at the Radon Laboratory are made with the goal of maintaining the low bias between NIST and the EPA.

In addition to the NIST performance tests, routine internal checks of bias and precision are made at the EPA Radon Laboratory using scintillation cells filled with radon from the standard radon emanation flasks. The scintillation cell counting system is routinely checked using a check source which consists of a scintillation cell containing a small amount of ^{226}Ra and producing a known count rate. These procedures are used to assess any changes in the calibration factors of the scintillation cells and associated counters.

The continuous radon measurement system for each chamber consists of two 1.4-L flow-through scintillation cells coated with zinc sulfide,** and counted on 12.7-

** Eberline Model SAC-R5 cells, manufactured by Eberline Instruments, 504 Airport Road, Santa Fe, NM 87505.

cm-diameter photomultiplier tubes. The continuous flow-through cells are calibrated using the smaller 0.36-L scintillation cells filled with air from the chamber of interest. The two continuous flow-through cells operate independently of each other and provide hourly results of the radon concentration within a chamber. The average of the two continuous radon monitor results serve as the reference value for the EPA Radon Laboratory during the period of interest.

The one-sigma uncertainty in the grab radon concentration using the 0.36 L scintillation cells is 3.6%. This uncertainty includes the *maximum* bias uncertainty reported by NIST for the measurements made by EPA of the NIST-produced radon gas ampules (U.S. DOC 1996), the precision counting uncertainty of the cells, the *maximum* precision uncertainty of the calibration factors for each cell, and bias and precision uncertainties inherent in the transfer procedure of the NIST-produced radon gas. An additional factor accounting for precision uncertainty in the 1.4-L flow-through cells produces an uncertainty value of 5.1% in the EPA Radon Laboratory's reference value for integrated radon concentration.

Radon decay product measurement system at the EPA Radon Laboratory

The primary radon decay product measurement method used by the EPA Radon Laboratory consists of radon decay product grab samples collected on filters with a diameter of 25 mm and a pore size of 0.8 microns. The samples are collected with a probe consisting of a metal tube 0.635 cm in diameter and 1.2 m in length with a metal screen and screw assembly at the collection end to hold the filter. The probe is attached with rubber tubing to a pump which is attached to a calibrated dry gas meter. Chamber air is drawn through the filter for 5 min at a rate of approximately 480 L h^{-1} . The filters are removed from the probe assembly and placed on zinc sulfide scintillation disks for counting on photomultiplier tubes^{††} using a modified Tsivoglou (Thomas 1972) counting scheme. The efficiency of the photomultiplier tubes is verified prior to sample counting using ^{241}Am sources purchased from NIST.

A minimum of two radon decay product grab samples are drawn and counted simultaneously. The average result of the radon decay product grab measurements are used to calibrate continuous radon decay product monitors, which consist of two working level monitors (WLM-30).^{‡‡} Several sets of grab samples are made throughout the day and corrections are applied each time to the average of the two continuous radon decay product monitors. The corrected average of the two continuous monitors during the time period of interest serves as the EPA Radon Laboratory reference value.

^{††} Ludlum Model 182, Ludlum Measurements, Inc., 501 Oak Street, Sweetwater, TX 79556.

^{‡‡} Scintrex Corporation, WLM-30/IBM PC Interface Operator's Manual, Revision 8.02, 222 Snidercraft Road, Concord, ON L4K 1B5, Canada.

The one-sigma uncertainty in the EPA Radon Laboratory reference value for the grab radon decay product concentration is 3.7%. This uncertainty includes the precision uncertainty of the multiple grab measurements and the overall uncertainty in the ^{241}Am source, as reported by NIST. An additional factor to account for the precision uncertainty in the continuous radon decay product monitors produces an uncertainty value of 4.5% in the integrated radon decay product concentration. It must be noted that there is no available standard for radon decay product concentration, and calibration processes would produce additional sources of uncertainty estimates.

U.S. Bureau of Mines facility (BOM)

The BOM Twilight Mine is a previously-operated uranium-vanadium mine, which was acquired by the BOM for the purpose of conducting research. The ore consists of a well-sorted, fine-grained, highly fractured sandstone with abundant carbonaceous material (Cooper and Holub 1990). The ventilation system in the mine consists of two primary fans located at the main exhaust portal and a secondary fan located in the north loop. Diesel pollutants were introduced into the environment via a small diesel engine located near the measurements performed on the third day (July 14), which caused an increase in particulates for the attachment of radon decay products. Radon concentration ranged from 5,110 to $15,535 \text{ Bq m}^{-3}$ and radon decay product concentration varied from about 3.54 to $74.32 \mu\text{J m}^{-3}$, as measured hourly. The mean equilibrium factor was 14.2%. Temperature varied from about 16 to 17°C and the relative humidity varied from about 16 to 22%.

Radon measurement system used by the U.S. Bureau of Mines

The primary radon measurement system used by the U.S. BOM consists of a set of 0.5-L scintillation cells^{¶¶} [also termed Lucas cells (Lucas 1957)], filled with mine air and analyzed for radon concentration using photomultiplier tubes. This primary measurement system has been intercompared with a variety of international and national programs, including the U.S. Department of Energy Environmental Measurements Laboratory intercomparison program (U.S. DOC 1990). The uncertainty associated with the U.S. BOM reference value reported here is limited to that calculated from their counting uncertainty; additional sources of uncertainty were not factored into the U.S. BOM uncertainty estimate of 1% associated with their reference values for integrated radon measurements. The reference values for integrated radon concentrations as reported by the U.S. BOM were obtained using the average of the two U.S. BOM flow-through scintillation cells to produce an average radon concentration each hour. The reference value for each period of exposure for each device is calculated as the integrated average of the hourly flow-through cell results.

The U.S. BOM provided the reference values for those measurements that began after July 11; the U.S. EPA provided the reference value for measurements that began prior to July 11 (see Table 4). The reference values for integrated radon concentrations at the U.S. BOM as measured by the EPA consist of the mean between two collocated Niton RAD-7^{§§} continuous radon monitors calibrated by EPA. These active monitors electrostatically collect charged alpha emitters on a solid-state detector capable of spectroscopic analysis. The uncertainty in the EPA reference value of 4.0% is an estimate propagated from the precision uncertainty exhibited between the two side-by-side units, the estimated uncertainty in the grab radon measurements used to calibrate the continuous monitors, and the calibration uncertainty (as described in the section on the EPA measurement system).

Radon decay product measurement system used by the EPA at the U.S. Bureau of Mines

The method and equipment used by the U.S. EPA to measure radon decay products in the mine is identical to the method used in the U.S. EPA Laboratory, as described previously.

The U.S. EPA produced the reference values for the grab radon decay product measurements at the mine, which were calculated from the mean of two or three multiple simultaneous grab measurements. The one-sigma uncertainty in the reference value for the grab radon decay product concentration is 2.5%. This uncertainty includes the precision uncertainty of the multiple grab measurements and the overall uncertainty in the ²⁴¹Am source, as reported by the NIST. This uncertainty is reported with the reference values in Table 8.

Radon decay product measurement system used by the U.S. Bureau of Mines

The continuous radon decay product monitors used in the mine consisted of two U.S. BOM-constructed units described in previous reports (U.S. BOM 1977; U.S. Patent Office 1980). These units incorporate a differential flow regulator to hold the air flow constant and a mass flow transducer or a volumetric flow transducer to measure the flow rate. The average of the two continuous monitors during the time period of interest serves as the reference value for integrated radon decay product concentration. Complete uncertainty estimates are not available; a 1% uncertainty due to calibration uncertainty (U.S. DOE 1986) alone is used in this report.

PARTICIPANTS

Table 1 presents the participants and their measurement methods used at the U.S. EPA Radon Laboratory; Table 2 presents those used at the U.S. BOM Twilight Mine. The methods ranged from simple and inexpensive to sophisticated and expensive. Many of the participants used commercially-available equipment, while other par-

ticipants employed components assembled themselves. A recent review describes the same or similar equipment used in this exercise (George 1996).

Participants measured both radon and radon decay product concentrations with integrated methods. In addition, participants measured radon decay products using grab sampling methods consisting of a sample taken over less than 0.1 h. Since this intercomparison exercise was conducted to facilitate the estimation of bias uncertainties for radon and radon decay product concentrations among different facilities, participants were not informed of the reference equilibrium factor. Grab radon sampling methods were not used by any of the participants. Participants were free to take part in one or more measurement methods during the exercise.

RESULTS

The results are shown in Tables 3 through 8 in terms of the participant's reported results and number of measurements (with one sample standard deviation), the U.S. EPA or U.S. BOM reference value (with the estimated uncertainty for the calculated average value during the measurement interval), and the performance ratio. The performance ratio is defined as the participant's result divided by the reference value, with an associated uncertainty calculated from the uncertainty of both values.

The organization providing the reference value is the U.S. EPA at the EPA laboratory (Tables 3, 5 and 7), as well as for the mine grab radon decay product values (Table 8) and for some mine integrated radon measurements (Table 4); the U.S. BOM provided the reference values for the integrated radon decay product concentrations (Table 6) and some of the integrated radon measurements at the mine (Table 4).

DISCUSSION

Integrated radon measurements at the U.S. EPA Laboratory

Participants integrated radon concentration measurements (see Table 3) made with continuous monitors were based on at least three multiple measurements. Reference values vary depending upon the individual start and stop times. For example, measurements with the continuous monitors manufactured by Honeywell instruments (now manufactured by Sun Nuclear^{¶¶}) were performed with three different devices of the same type. Genitron Corporation,^{¶¶} performed measurements with five AlphaGuard pulsed ion chamber devices. The average of the EPA hourly measurements during the measurement period was used as the reference value, as described earlier. The continuous monitors performed well, with the Genitron instrument producing a perfor-

^{¶¶} Sun Nuclear Corporation, 415 Pineda Court, Melbourne, FL 32940.

^{¶¶} Genitron Instruments, Heerstrasse 149 D60488, Frankfurt, Germany.

^{§§} Niton Corporation, Bedford, MA.

Table 3. Participants' integrated radon results from measurements in the EPA Laboratory. OC = open face charcoal collector; DB = diffusion barrier charcoal collector; LS = analyzed via liquid scintillation counting; SS = solid-state detector type; P = passive; A = active (pump); CR-39 and LR-115 = alpha track detectors; EIC = electret ion chambers; PC = pulse counting method.

Participant	Method	<i>N</i>	Result $\pm S_p$, Bq m ⁻³	Reference $\pm S_R$, Bq m ⁻³	Performance ratio $\pm S_{PR}^a$
Honeywell	SS,P	3	658 \pm 19	717 \pm 37	0.92 \pm 0.06
Genitron	PC,A	5	698 \pm 9	717 \pm 37	0.97 \pm 0.05
ASpectra1	OC,P	10	663 \pm 56	720 \pm 37	0.92 \pm 0.10
ASpectra2	DB,P	10	1,009 \pm 116	711 \pm 36	1.42 \pm 0.13
BRI	DB,P,LS	4	638 \pm 66	720 \pm 37	0.87 \pm 0.12
CLRP2	DB,P,LS	4	760 \pm 26	720 \pm 37	1.06 \pm 0.06
NIH	DB,P,LS	4	776 \pm 38	720 \pm 37	1.08 \pm 0.07
MAIB	LS	6	764 \pm 31	720 \pm 37	1.06 \pm 0.07
RadElec	EIC,P	4	662 \pm 11	711 \pm 36	0.93 \pm 0.05
CLRP1	EIC,P	2	739 \pm 38	733 \pm 37	1.01 \pm 0.07
ASpectra3	CR-39,P	15	480 \pm 172	733 \pm 37	0.66 \pm 0.36
CLRP3	CR-39,P	4	655 \pm 93	733 \pm 37	0.89 \pm 0.15
INP	CR-39,P	5	1,030 \pm 150	733 \pm 37	1.41 \pm 0.27
UOZ	LR-115,P	3	392 \pm 23	733 \pm 37	0.54 \pm 0.08

^a $S_{PR}^2 = S_p^2 + S_R^2$, where S_p = sample standard deviation of participant, and S_R = estimated standard deviation of the reference value \cong 5.1%. N = number of simultaneous measurements.

Table 4. Participants' integrated radon results from measurements in the BOM Twilight Mine. OC = open face charcoal collector; DB = diffusion barrier charcoal collector; A = active (pump); P = passive; CR-39 and LR-115 = alpha track detector; EIC = electret ion chamber; PC = pulse counting method; LS = analyzed via liquid scintillation counting; SS = solid state detector type.

Participant	Method	<i>N</i>	Result $\pm S_p$, Bq m ⁻³	Reference $\pm S_R$, Bq m ⁻³	Performance ratio $\pm S_{PR}^a$
Algade1	SS,A	1 ^b	8,917	8,673 \pm 87 ^c	1.03 ^d
Genitron	PC,A	5	6,751 \pm 107	8,468 \pm 85 ^c	0.80 \pm 0.01
Honey	SS,A	3	8,160 \pm 271	8,468 \pm 85 ^c	0.96 \pm 0.03
ASpectra1	OC,P	10	9,461 \pm 1543	8,673 \pm 87 ^c	1.09 \pm 0.18
ASpectra2	DB,P	10	13,107 \pm 2105	8,390 \pm 336 ^c	1.56 \pm 0.26
BRI	DB,P,LS	6	7,479 \pm 221	8,592 \pm 86 ^c	0.87 \pm 0.15
CMII	DB,P,LS	5	7,821 \pm 289	8,390 \pm 336 ^c	0.93 \pm 0.05
MAIB	DB,P,LS	10	8,749 \pm 318	8,592 \pm 86 ^c	1.02 \pm 0.04
NIH	DB,P,LS	4	9,245 \pm 424	8,592 \pm 86 ^c	1.08 \pm 0.05
CLRP1	EIC,P	2	8,329 \pm 908	9,109 \pm 91 ^c	0.91 \pm 0.10
ASpectra3	CR-39,P	15	9,779 \pm 1137	8,390 \pm 336 ^c	1.17 \pm 0.14
CLRP3	CR-39,P	5	8,616 \pm 1426	8,390 \pm 336 ^c	1.03 \pm 0.18
INP	CR-39,P	5	10,800 \pm 1500	8,390 \pm 336 ^c	1.29 \pm 0.19
NIOM1	LR-115,P	30	16,800 \pm 5401	8,390 \pm 336 ^c	2.00 \pm 0.65
UOZ	LR-115,P	4	7,586 \pm 820	8,390 \pm 336 ^c	0.90 \pm 0.10
Ghana	LR-115,P	4	11,866 \pm 1286	8,528 \pm 85 ^c	1.39 \pm 0.15

^a $S_{PR}^2 = S_p^2 + S_R^2$, where S_p = sample standard duration of participant's data, and S_R = estimated standard deviation of the EPA reference value (4.0%) or the BOM reference value (1% due to counting uncertainty alone; other uncertainty estimates are not available).

^b Participant reported only one value with no associated uncertainty.

^c BOM reference value uncertainty of 1% is based on the uncertainty due to counting statistics only; complete uncertainty estimates were not available.

^d Propagated uncertainty values not possible; see b, above.

^e EPA reference values represent a propagated uncertainty of 4.0% based on estimates of precision, bias, and calibration uncertainty.

Table 5. Participants' integrated radon decay product results from measurements in the EPA Radon Laboratory. A = active (pump); LR-115 = alpha track detector.

Participant	Method	<i>N</i>	Result $\pm S_p$, μ J m ⁻³	Reference $\pm S_R$, μ J m ⁻³	Ratio $\pm S_{PR}^a$
Algade 3	A,LR-115	5	1.58 \pm 0.31	1.58 \pm 0.07	1.00 \pm 0.20
Algade 2	A,LR-115	4	2.45 \pm 0.21	1.75 \pm 0.08	1.41 \pm 0.09

^a $S_{PR}^2 = S_p^2 + S_R^2$, where S_p = sample standard deviation of participant's data, and S_R = estimated standard deviation of the EPA reference value \cong 4.5%. N = number of simultaneous measurements.

Table 6. Participants' integrated radon decay product results from measurements made in the BOM Twilight Mine. A = active (pump); P = passive; LR-115 = alpha track detector; SS = surface barrier detector; TLD = thermoluminescent detector.

Participant	Method	N	Result $\pm S_p$, $\mu\text{J m}^{-3}$	Reference $\pm S_p$, ^a $\mu\text{J m}^{-3}$	Ratio $\pm S_{PR}$ ^b
Algade2	A,LR-115	8	9.59 \pm 1.23	8.74	1.10 \pm 0.14
Algade3	A,LR-115	10	8.05 \pm 1.27	8.74	0.92 \pm 0.15
CMI2	A,TLD	12	7.03 \pm 1.29	9.51	0.74 \pm 0.14
EPA2	A,SS	3	7.63 \pm 0.46	7.18	1.06 \pm 0.06
NIOM2	P,LR-115	30	23.58 \pm 7.53	7.11	3.32 \pm 1.06

^a The 1% uncertainty for the BOM reference value is based only on calibration uncertainty (U.S. DOE 1986); a complete uncertainty estimate is not available.

^b $S_{PR}^2 = S_p^2 + S_R^2$, where S_p = sample standard deviation of participant's data, and S_R = estimated uncertainty in BOM reference value due to calibration uncertainty (1%). N = number of simultaneous measurements.

mance ratio of 0.97 ± 0.05 , with a low precision uncertainty of 1.3%. The Honeywell device's results produced a performance ratio of 0.92 ± 0.06 , with a precision uncertainty of 2.9% relative standard deviation. These continuous monitors operate quite differently, with the Genitron model housing a sophisticated pulse counting ionization chamber and the Sun Nuclear Model 1023 consisting of a filtered chamber enclosing a diffused junction photo diode with relatively low sensitivity.

Five participants used activated charcoal adsorbers, of both the open-face and diffusion-barrier type. The open-face and diffusion barrier models that used a surface area of approximately 7.6 cm in diameter were analyzed for gamma emissions with Na(I) systems. The three types that used a smaller mass of charcoal each incorporated diffusion-barriers on their surfaces and were analyzed for alpha and beta emissions using liquid scintillation technology. Results were generally good, with a performance ratio between 0.92 and 1.06, except for the Alpha Spectra^{##} diffusion barrier model, which produced a performance ratio of 1.42 ± 0.13 . Precision estimates were calculated from between four and 10 replicate, side-by-side measurements, with replicate results all exhibiting a relative standard deviation less than 11.5%.

Two organizations used electret ion chambers, including a primary U.S. manufacturer and a Polish organization that used devices manufactured by the same U.S. organization. Both produced results close to the reference value with low precision uncertainties. The Polish organization's performance ratio was 1.01 ± 0.07 , with a precision uncertainty estimated from only two devices to be a 5.1% relative standard deviation. The results from the electret manufacturer produced a performance ratio of 0.93 ± 0.05 , with a precision uncertainty estimated from four devices to be a 1.7% relative standard deviation.

Alpha-track devices were used by four participants, including two Polish, one Croatian, and one U.S. organization. Results ranged from a low performance ratio of 0.54 ± 0.08 from the University of Zagreb using a LR-115 detector assembled and analyzed by the partici-

pant, to a high performance ratio of 1.41 ± 0.27 from the Institute of Nuclear Physics in Poland, using a CR-39 detector assembled and analyzed by that organization. Precision uncertainties, calculated from sets of replicates ranging from three to 15 detectors, ranged from 8.4 to 35.8% relative standard deviation.

Integrated radon measurements at the U.S. BOM Twilight Mine

Table 4 presents the integrated radon measurements performed at the Twilight Mine. Participants using continuous monitors include Algade1, Genitron, and Honeywell, with performance ratios ranging from 0.80 to 1.03. Genitron performed much better in the EPA laboratory environment (see Table 3) than in the mine, while Honeywell performed about the same, and Algade1 did not perform measurements for comparison in the laboratory environment.

Six participants performed integrated measurements with charcoal adsorbing devices in the Twilight Mine, deploying from 4 to 10 devices simultaneously. The performance ratios range from 0.87 ± 0.15 from the Polish BRI organization using the Packard Pico-Rad charcoal vial analyzed via liquid scintillation to 1.56 ± 0.26 from the ASpectra2 diffusion barrier device. The results for charcoal adsorbing devices were generally consistent between the EPA laboratory and the mine environment.

One participant employed two electret ion chambers in the mine environment, producing a performance ratio of 0.91 ± 0.10 . This participant, the Polish CLRPI group, also used two electret ion chambers in the EPA laboratory environment, producing a performance ratio of 1.01 ± 0.07 in the laboratory.

Alpha-track devices were used by six participants in the mine, deploying between 4 and 30 devices simultaneously. Their performance ratios ranged from 0.90 ± 0.10 from UOZ in Croatia using LR-115, to 2.00 ± 0.65 from the Polish NIOM1 group also using LR-115. Participants using alpha-track devices produced performance ratios spanning a broad range in both the mine and the EPA laboratory environment.

^{##} Alpha Spectra, Inc., 210 Clayton Street, Denver, CO 80206.

Table 7. Participants' grab radon decay product results from measurements in the EPA Laboratory. S = silicon alpha detector; B = beta detector.

Participant	Method	N	Performance ratio $\pm S_R^a$
Algade4	S	11	1.26 \pm 0.05
TN	S	12	1.09 \pm 0.04
INCT	S	12	0.67 \pm 0.02
MAB	B	12	1.04 \pm 0.04

^a S_R = estimated standard deviation of the EPA reference value \approx 3.7%.
N = number of measurements.

Table 8. Summary of the participants' grab radon decay product results from measurements in the BOM Twilight Mine. SS = solid state detector; Mk. = Markov method; Tsi. = Modified Tsivoglou method; B = beta detection method.

Participant	Method	N	Performance ratio $\pm S_R^a$
MAB	B	10	0.86 \pm 0.02
AECB	Tsi.	11	0.98 \pm 0.03
TN	SS	11	0.95 \pm 0.02
Algade 4	SS	11	1.13 \pm 0.03
CM13	Tsi.	11	0.74 \pm 0.02
INCT	Mk.	11	0.66 \pm 0.02
NIOM3	Mk.	11	0.96 \pm 0.02
NIOM4	Tsi.	11	0.91 \pm 0.02

^a Uncertainty associated with the EPA reference value is 2.5% based on estimates of precision (between simultaneous grabs) and bias (uncertainty reported in check source). N = number of measurements.

Integrated radon decay product measurements at the U.S. EPA Laboratory

Table 5 presents the results from the integrated radon decay product measurements. There were two instruments that measured average radon decay product concentration in the EPA chamber over time periods ranging from 4 h to 4 d, both from the Algade company in France. Four Site Alpha Dosimeters exposed simultaneously for an integrated time period of 99 h produced a performance ratio of 1.41 ± 0.09 and a precision estimate of 8.5%. Five Personal Alpha Dosimeters exposed for four time periods ranging from 4 to 6 h in duration produced more accurate results, with an average performance ratio of 1.00 ± 0.20 but a greater precision estimate of 19.6%. The Algade dosimeters are constructed of unfiltered LR-115 material; the Personal Alpha Dosimeters incorporate a battery-operated pump and required recharging between each sampling period. The Site Alpha Dosimeter operated continuously from AC power.

Integrated radon decay product measurements at the U.S. BOM Twilight Mine

Table 6 presents the results of integrated radon decay product measurements made in the mine by five organizations. Performance ratios ranged from 0.74 ± 0.14 to 3.32 ± 1.06 . The French Algade firm used eight of its Algade2 devices in the mine to produce a performance ratio of 1.10 ± 0.14 . This same device type in the laboratory produced a performance ratio of 1.41 ± 0.09 . The Algade3 device performed relatively well in both

environments, with a performance ratio of 0.92 ± 0.15 in the mine and 1.00 ± 0.20 in the laboratory.

Short-term (grab) measurements of radon decay product concentrations at the U.S. EPA Laboratory

Four organizations from France, Canada, Poland, and Germany participated in measurements of short-term radon decay product concentrations (Table 7). The organization from Germany used an instrument incorporating a beta detector, pump, and filter, and produced a performance ratio of 1.04 from 12 sequential measurements. The remaining three groups used units incorporating solid-state alpha detectors, filters, and pumps, and produced performance ratios between 0.67 and 1.26.

Short-term (grab) measurements of radon decay product concentrations at the U.S. BOM Twilight Mine

Table 8 presents the results from the grab radon decay product measurements made in the mine. Eight organizations performed measurements over 3 d, with up to 11 total measurements performed. Average performance ratios ranged from 0.66 to 1.13. Results for the four participants who conducted measurements in both the mine and laboratory were generally consistent.

CONCLUSION

The continuous radon gas monitors performed well in the EPA laboratory both in terms of low uncertainties of precision and bias over the measurement period of 75 h. The continuous monitors in the mine environment performed similarly, with the exception of the Genitron instrument, which exhibited a low bias in the mine.

Of the six participants who measured radon concentrations in the EPA laboratory with charcoal collectors, four produced results within 8% of unity for the performance ratio. Of the remaining two participants, one produced results that gave a performance ratio of 0.87 ± 0.12 while the other was clearly biased with a performance ratio of 1.42 ± 0.13 . This latter manufacturer participated using two designs of charcoal collectors; the diffusion barrier produced the high bias of 1.42 ± 0.13 and the open-face configuration a less biased 0.92 ± 0.10 performance ratio. The results for charcoal adsorbers in the mine environment were consistent with those in the laboratory.

Only one of the four participants who used alpha-track detectors in the EPA laboratory produced results within 11% of unity for the performance ratio. In the mine, only two of the six alpha-track participants produced performance ratios within 10% of unity. (It must be noted that the major worldwide producer and laboratory for analyzing alpha-track detectors was not represented in this study.)

Both participants who employed electret ion chambers in the EPA laboratory performed well, with performance ratios within 10% of unity and relative standard deviations less than about 7%. The single participant who

deployed electret ion chambers in the mine also produced a performance ratio within 10% of unity.

The performance ratios for integrated radon decay product measurements in the EPA laboratory ranged from 1.00 ± 0.20 to 1.41 ± 0.09 with only two device types; the performance ratios in the mine ranged from 0.74 ± 0.14 to 3.32 ± 1.06 with five participants. Three of the five mine participants produced performance ratios within 10% of unity.

Of the four organizations that measured radon decay product concentrations in the EPA laboratory using grab methods, two produced average performance ratio results within 10% of unity, and the remaining two were within 30% of unity. In the mine, four of eight participants produced average performance ratios within 10% of unity; of the four remaining participants two were within 15%, and two were within 36%.

The overall conclusion that can be drawn is that more international intercomparison exercises are needed, as there are differences between calibrations even within the same organization. This and similar exercises are extremely useful to the radon measurement community, providing valuable data that can be used to improve calibrations and modernize equipment. Intercomparison exercises are necessary to ensure that published studies produce meaningful results that can be interpreted in terms of studies conducted using different types of equipment and by different organizations. This is especially important when making interpretations relevant to health effects, which is the ultimate objective of many radon and decay product measurements.

Acknowledgments—The IRMP would like to thank Robert Holub for his support and assistance, including the use of the U.S. Bureau of Mine's Twilight mine, without which this intercomparison would not have been possible.

REFERENCES

- Colle, R.; Hutchinson, J.; Unterweger, M. The NIST primary radon-222 measurement system. *J. Res. Natl. Inst. Standards Technol.* 95:155-165; 1990.
- Cooper, W. E.; Holub, R. F. International intercalibration and intercomparison program radon daughter measurements, exercise at the Twilight Mine, Uravan, CO. Denver, CO: U.S. Department of the Interior, Bureau of Mines. IC 9257; 1990.
- George, A. C. State-of-the-art instruments for measuring ra-

- don/thoron and their progeny in dwellings—A review. *Health Phys.* 70:451-463; 1996.
- George, A. C.; Tu, K. W. Intercomparison of radon progeny measurement methods and equipment in North America. *Radiat. Protect. Dosim.* 24:273-276; 1988.
- Knutson, E. The OECD/CEC radon measurements intercomparison. A further analysis of the results. *Radiat. Protect. Dosim.* 24:277-280; 1988.
- Lucas, H. F. Improved low-level alpha scintillation counter for radon. *Rev. Sci. Inst.* 28:680-683; 1957.
- Lucas, H. F.; Markun, F. Radon in air calibration procedure: A primary method. *Nucl. Sci. Eng.* 99:82-87; 1988.
- OECD Nuclear Energy Agency. International intercalibration and intercomparison of radon, thoron, and daughters measuring equipment. Paris, France: OECD; 1980.
- OECD Nuclear Energy Agency. International intercalibration and intercomparison of radon, thoron, and progeny measuring equipment. Report on Part 1, Radon measurement. Paris, France: OECD; 1986.
- Pearson, M. D. Evaluation of the performance characteristics of radon-daughter concentration measurements devices under controlled environmental conditions. Grand Junction, CO: U.S. DOE; Report UNC/GJ-44 (TMC); 1989.
- Thomas, J. W. Measurement of radon daughters in air. *Health Phys.* 23:783-789; 1972.
- U.S. Bureau of Mines. Continuous working level measurements using alpha or beta detectors. Denver, CO: U.S. BOM; RI 8237; 1977.
- U.S. Department of Commerce, National Institute of Standards and Technology. A calibration and quality assurance program for environmental radon measurements. In: Special Issue on Radon Measurement Standards and Calibration. *J. Res. Natl. Inst. Stand. Technol.* 95:127-134; 1990.
- U.S. Department of Commerce, National Institute of Standards and Technology. Report of traceability for radon-222 for U.S. Environmental Protection Agency Office of Radiation and Indoor Air—Las Vegas. Gaithersburg, MD: U.S. DOC; 1996.
- U.S. Department of Energy, Grand Junction Projects Office, Technical Measurements Center. Interlaboratory Radon-Daughter Measurement Comparison Workshop: 9-12 September 1985. Washington, DC: U.S. DOE; GJ/TMC-25 UC-70A; 1986.
- U.S. EPA. Quality Assurance Project Plan for the Radon Measurement Traceability Project. Las Vegas, NV: Office of Radiation and Indoor Air, Radiation and Indoor Environments National Laboratory; 1996.
- U.S. Patent Office. Method of continuously determining radiation working level exposure. U.S. Pat. 4185199; 22 Jan 1980.



Abstr
work
sure
mad
and
studi
and
per
niqu
prox
worl
cent
staff
not
with
1977
their
orga
at o
worl
140
The
on a
the
to 2
depr
any
125
ing
1991
effe
tecti
tion
Hea
Key
radi

WH
isol

SE-

add

ceiv