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ABSTRACT

The results from the May 1995 Intercomparison of Active, Passive and Continuous Instruments for Radon and Radon Progeny Measurement conducted in the EML radon exposure and test facility are presented. Represented were 13 participants that measure radon with open faced and diffusion barrier activated carbon collectors, 10 with nuclear alpha track detectors, 9 with short-term and long-term electret/ionization chambers, and 13 with active and passive commercial electronic continuous monitors. For radon progeny, there were four participants that came in person to take part in the grab sampling methodology for measuring individual radon progeny and the potential alpha energy concentration (PAEC). There were 11 participants with continuous and integrating commercial electronic instruments that are used for measuring the PAEC.

The results indicate that all the tested instruments that measure radon fulfill their intended purpose. All instruments and methods used for grab sampling for radon progeny did very well. However, most of the continuous and integrating electronic instruments used for measuring the PAEC or working level appear to underestimate the potential risk from radon progeny when the concentration of particles onto which the radon progeny are attached is <5,000 cm^{-3}.

Table of Contents

INTRODUCTION

The fourth intercomparison of monitoring instruments for radon and radon progeny measurements was conducted at EML to determine the performance and suitability of these devices to assess human radiation exposure from radon and radon progeny (George et al., 1995). This intercomparison exercise is mandated by the U. S. Department of Energy (DOE), Office of Health and Environmental Research (OHER), and is recommended by the Co-ordinated Research Program (CRP) of the International Atomic Energy Agency (IAEA), in cooperation with the Commission of European Communities (CEC). In 1992, the International Radon Metrology Program (IRMP) was established to provide the scientific community and the users of these instruments with a network of reference calibration centers where they can obtain high quality assurance standards in the area of radon metrology. EML is the reference calibration facility in North America and as such provides support to participants from the U. S., Canada and South America. The success and usefulness of this program is indicated by the participation of researchers from Europe and Asia who are seeking a means to ensure consistency in radon measurements on a global scale.

This program is different from the U. S. Environmental Protection Agency (EPA) sponsored Radon Measurement Proficiency Program (RMP), and is separate from EML's sponsored National Radon Intercomparison Program (Fisenne, 1995). The purpose of this intercomparison exercise is to evaluate the performance of different types of instruments which are used to measure environmental radon and radon progeny. Previously, similar exercises were conducted by EML in 1990, 1992 and 1994 (George et al., 1995).

EXPOSURE AND TEST FACILITY

The intercomparison tests were conducted in EML's 30 m^3 radon, thoron and progeny test facility from April 28 through May 10, 1995. The chamber provides a well-controlled, airtight and uniform environment. It is the primary test facility at EML in which a large number and diverse types of monitoring instruments can be accommodated for calibration, evaluation and intercomparison purposes (EML Procedures Manual, in press). The test chamber is environmentally controlled for temperature and humidity. Monodispersed or polydispersed aerosols are generated to study radon and thoron progeny attachment and behavior, and to investigate instrument performance under different conditions of exposure. Also, particle size measurements are performed to develop techniques for the assessment of the health risk from inhalation of radon and thoron progeny.

The exercise extended over periods from 1 to 12 days in order to accommodate instruments with different exposure protocols and different sensitivity limits when used in field applications. In all, there were more than 30 participants (consisting of: U. S. government laboratories, universities and private firms, and several foreign government agencies and universities) that conduct radon and radon progeny measurements and research studies.

Temperature and humidity were controlled and ranged from 18-20°C and 31%-34% RH, respectively. The concentration of radon in the test chamber was maintained at about 940 Bq m^{-3}. During testing of active devices for radon progeny, concentrations ranging from 180 - 2,700 nJ m^{-3} were obtained by varying the concentration of particles generated from wax or a burning candle. The wax particles were generated by two TSI condensation aerosol generators Models 3470 and 3472, and the particle concentration was measured continuously with a condensation nuclei counter. The gamma background exposure inside the chamber was nearly constant at 0.09 μSv h^{-1}.

During testing, the instruments were placed inside the EML test chamber 0.5 - 1.5 m above the floor. Grab sampling for radon progeny was conducted from the adjacent room by taking samples from inside the test chamber through sampling ports. Analysis of the radon progeny activity inside the chamber was conducted using the Thomas method (Thomas, 1972), and the least squares method (Raabe and Wrenn, 1969). One participant used the Rolle method (Rolle, 1972). The concentrations of radon and radon progeny inside the test chamber were monitored continuously using a 3.0 L scintillation cell monitor (Eberline RGM3) and a quasi-continuous radon progeny monitor (Alpha Nuclear 770B), respectively. The particle size of the airborne radon progeny measured with a particle size analyzer ranged from 90 nm to 125 nm geometric mean diameter (GMD), corresponding to 100 nm to 200 nm activity median diameters (AMDs).

QUALITY ASSURANCE

The accuracy of the concentrations of radon inside the test chamber was determined by measuring it continuously with a flow through scintillation cell monitor that was calibrated in a known atmosphere of radon traceable to the National Institute of Standards and Technology (Fisenne and Keller, 1985). Also, calibration with a secondary standard was performed daily during the 12 days of the intercomparison. The total uncertainty in the EML value is 5.2%. Radon progeny measurements made with EML instruments and methods are accurate to within 3% at the concentration levels tested. Their accuracy was verified on numerous occasions during intercomparisons with several reference laboratories throughout the world.

RADON AND RADON PROGENY INSTRUMENTS

The instruments and methods used for radon measurements only are listed in Table 1. Those used for radon and thoron progeny are listed in Table 2. The passive integrating devices for radon included: 1) several types of open faced and diffusion barrier activated carbon collectors; 2) two types of electret/ionization chambers; 3) several types and different configurations of nuclear alpha track detectors; 4) pulse ionization chambers; and 5) scintillation cell monitors. The active instruments for radon included scintillation cell and solid-state detection monitors. The active instruments for measuring radon or thoron progeny included grab, integrating and continuous monitors by sampling on filters that are counted by solid-state and scintillation detectors or by registration of nuclear alpha tracks in solid-state materials.

RESULTS AND DISCUSSION

The results of the radon intercomparison measurements for instruments and methods are listed in Table 3 and are shown in Figure 1. The results for individual radon progeny and PAEC obtained with grab samples are listed in Table 4 and are shown in Figure 2. The results from continuous and integrating instruments for PAEC are also listed in Table 4 and are shown in Figure 3. The EML values were used as the reference against which all other measurements were compared. To maintain participant confidentiality, the reported values are listed randomly. In future intercomparisons, the performance of different instruments and their owner's identification will be revealed. For comparison purposes, the different types of radon instruments were grouped separately into four categories (Table 3) consisting of passive activated carbon collectors, nuclear alpha track detectors, electret/ionization chambers, and continuous active and passive electronic devices.

The range, the mean and standard deviation (SD) of the individual grouped data are compared with the mean reference value obtained by EML during the same test period. The ratios (participant/EML), and the associated propagated errors are listed in the last column of Table 3 and are shown in Figure 1. The mean ratios and their SDs for the different instruments categories are: activated carbon collectors = 1.02±0.07; nuclear alpha track detectors = 0.97±0.18; electret/ionization chambers = 0.99±0.14; and continuous active and passive electronic monitors = 0.98±0.04. More than 85% of the participants using activated carbon collectors obtained values that were within 10% of the reference value. When compared with the last intercomparison (George, 1995) with a mean ratio of 1.04±0.10, both open faced and diffusion barrier carbon collectors performed very well, indicating proper calibration with maintenance of good quality control procedures.

The mean ratio of the 10 sets of nuclear alpha track detectors is 3% lower than the reference value. However, the range of the ratios of the mean values is 0.69-1.25 as compared to 0.81-1.10 in the 1994 intercomparison (George, 1995).

The number of participants using short-term and long-term electret/ionization chambers was the same as in the 1994 intercomparison. However, two types of electret/ionization chambers were included for the first time. Most of the participants used the RAD elec type. The mean ratio and SD of the 9 participants is 0.99±0.14 as compared to 0.97±0.03 from the previous intercomparison. The higher SD is attributed to three participants that may have used inappropriate calibration factors or may have problems with quality control.

Continuous electronic active and passive radon instruments performed very well. The mean ratio and SD is 0.98±0.04 as compared to 1.01±0.05 from the 1994 intercomparison. It is encouraging to observe that continued maintenance and proper calibration resulted in reliable instruments and measurement results. Not long ago, some of these instruments were plagued with both positive and negative calibration biases.

The measurement results for the individual radon progeny concentrations and PAEC obtained by the four participants are listed in Table 4 and Figures 2 and 3. Table 4 lists the participant's individual radon progeny and PAEC ratios to that of the EML reference value during simultaneous grab sampling. The last column lists the concentration of the reference radon progeny atmosphere. The uncertainty of the PAEC measurements were calculated and reported based on counting statistics alone. The mean ^{218}Po values indicate good agreement with the EML reference value and the other participants. The mean ratios for both ^{214}Pb and ^{214}Bi are in good agreement with the reference value although the SDs for ^{214}Bi were large, ranging from 14%-22%. The mean ratios for PAECs were in very close agreement with EML's reference value and with each other, ranging from 0.97±0.04 - 1.01±0.04. The airborne radon progeny were collected on open faced filters. Even at low concentrations of condensation nuclei, all participants performed well. Also, the counting efficiencies and the air flow rates used by each participant were checked every day during the intercomparison and were found to be accurate.

The results from 10 continuous and 1 integrating PAEC instrument are listed at the bottom of Table 4 and are shown in Figure 3. The ratios ranged from 0.57-1.03 with a mean and SD of 0.80±0.16. By comparison, in the last intercomparison (George et al., 1995) the ratios ranged from 0.66-1.05 with a mean and SD of 0.89±0.14. In the present intercomparison, 55% of the instruments gave results within ±25% of the EML reference value. The intercomparison for PAEC measurements was conducted in an atmosphere where the concentration of condensation nuclei ranged from 2,000-200,000 particles cm^{-3}, but mostly it ranged between 2,000-5,000 particle cm^{-3}. Particle concentrations in residential buildings drop below 5,000 cm^{-3} at night when indoor activity ceases, therefore it was necessary to find out how some of the continuous and integrating PAEC instruments perform under such conditions. The exposure environment was too harsh for some instruments that seemed to experience radon progeny losses by plateout. Most of the commercial instruments are not calibrated at low concentrations of airborne particulates and the underestimation of the PAEC is an indication of radon progeny losses by plateout before they reach the collection filter.

CONCLUSIONS

The instruments and methods used by the participants in this intercomparisons for the measurement of radon were found to fulfill their intended purpose. All participants that were found to meet EPA's RMP performance standards used activated carbon collectors and continuous active and passive devises. Two participants with nuclear alpha track detectors and one with electret/ionization chambers failed. Those participants that were within 25-35% of the EML reference value were first time participants in the EML intercomparisons. Commercial electronic instruments used by some participants for radon measurements performed very well, indicating proper calibration and continuous maintenance by both the manufacturer and the user. However, most of the commercial electronic instruments for radon progeny did not do so well when tested in environments where the concentration of airborne particles is <5,000 cm^{-3}. It is believed that the filter detector assembly located inside the instrument is depleted of unattached radon progeny and of radon progeny that attach on small particles that plate out on surfaces upstream from the filter. The loss of radon progeny by plateout is more pronounced in instruments that operate with air flow rates of <0.2 L min^{-1}. Some of the underestimation of the PAEC is due to calibration bias. All participants that used grab sampling for radon progeny, which is considered their primary or standard method for measuring radon progeny, did very well indicating that the instruments are properly calibrated and maintained and that the operators are well trained in their use. In general, the intercomparison exercise demonstrated that active, passive, integrating, continuous or grab sampling instruments for radon are in good standing. Some participants that use instruments for measuring the PAEC need to address the question of plateout of radon progeny that causes the underestimation of radon progeny concentration levels.

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TABLE 1

PASSIVE AND ACTIVE RADON INSTRUMENTS USED BY PARTICIPANTS

Participant	Instrument/Method
AECL-Low Level Radioactive waste Ottawa, Ontario, Canada	PI, electret/ionization chamber PC, scintillation cell monitor
Air Check Arden, NC	PI, activated carbon collectors (DB) two types gamma counting
Bowser/Morner Dayton, OH	PI, activated carbon collectors (OF), gamma counting PC, pulse ionization chamber
Chile Nuclear Energy Commission Santiago, Chile	PI, Nuclear alpha track detectors PI, pulse ionization chamber
Enviroserv, Inc. Morristown, NJ	PI, activated carbon collectors (OF), gamma counting PC, solid-state with diffused junction photodiode.
Femto-TECH Inc. Carlisle, OH	PC, pulse ionization chamber (CRM-510)
Gemini Research Timonium, MD	PI, nuclear alpha track detectors (NYU type) AC, scintillation cell monitor
Harvard University Cambridge, MA	PI, activated carbon collectors (OF), gamma counting
Health and Welfare of Canada Ottawa, Ontario, Canada	PI, electret/ionization chamber
Hebrew University of Jerusalem Jerusalem, Israel	PI, activated carbon collectors-liquid scintillation counting
Institute of Nuclear Engineering Rio de Janeiro, Brazil	PI, nuclear alpha track detectors CR-39 and Makrofol
Institute of Nuclear Sciences Aomori, Profecture, Japan	PI, nuclear alpha track detector
Institute of Nuclear Sciences Vinca, Yugoslavia	PI, nuclear alpha track detectors
Landauer Inc. Glenwood, IL	PI, nuclear alpha track detectors
Nagoya University Nagoya, Japan	PI, nuclear alpha track detectors
National Institute of Env. Sciences Chiba, Japan	PI, nuclear alpha track detectors
New Jersey DEP Trenton, NJ	PI, activated carbon collectors (OF) gamma counting
New York University New York, NY	PI, nuclear alpha track detectors

TABLE 1 (Cont'd)

Participant	Instrument/Method
Pennsylvania DER Harrisburg, PA	AC, scintillation cell (RGM3) AC, scintillation cell (Certifier II) AC, solid-state alpha spectometry (RAD7) PI, activated carbon collectors (DB) gamma counting PI, electret/ionization chamber
Rad Elec. Inc. Frederick, MD	PI, electret/ionization chamber, short-term and long-term types
Radon Environmental Monitoring Northbrook, IL	PI, nuclear alpha track detectors PI, activated carbon collectors, liquid scint. counting
Radon Testing Corporation of America Irvington, NY	PI, activated carbon collectors (OF) gamma counting PI, electet/ionization chamber (new type)
Scintrex Ltd. Concord, Ontario, Canada	AC, solid-state (RG-30)
St. Johns University Collegeville, MN	PI, nuclear alpha track detectors
Teledyne Environmental Services Westwood, NJ	PI, activated carbon collectors (OF) gamma counting
Tokyo Metropolitan Isotopes Research Center, Tokyo	PI, nuclear alpha track detectors
Turkish Atomic Energy Authority Instanbul, Turkey	PI, nuclear alpha track detectors
U. S. EPA Montgomery, AL	PI, activated carbon collectors (DB) gamma counting PI, electret/ionization chambers
Wilkes Barre University Wilkes Barre, PA	PI, activated carbon collectors (DB) gamma counting PI, electret/ionization chamber AC, scintillation cell (RGM-3) PC, scintillation monitor (PRD)

AC = active continuous
DB = diffusion barrier
OF = open faced
PC = passive continuous
PI = passive integrating

TABLE 2

ACTIVE INSTRUMENTS AND METHODS FOR MEASURING PAEC

Participant	Method/Detection	Efficiency Counting (%)	Flow Rate ($L\ min^{-1}$)
Alpha Nuclear Mississauga, Canada	Continuous, alpha, silicon	17.0	0.13
AECL Ottawa, Canada	Grab sampling, alpha scintillation Continuous, solid-state	46.0 -	19.80 1.00
Bowser/Morner Dayton, OH	Grab sampling, alpha scintillation	48.0	11.40
Canadian Institute of Rad. Safety (CAIRS), Canada	Integrating, nuclear alpha track	-	0.80
Enviroserv Inc. Morristown, NJ	Continuous, alpha, silicon	17.0	0.13
Gemini research Timonium, MD	Continuous, alpha, silicon	32.0	0.25
Scintrex Ontario, Canada	Continuous, solid-state	-	1.00
Thomson & Nielsen Ottawa, Canada	Grab sampling, solid-state Continuous, solid-state	- -	7.50 1.00
EPA Montgomery, AL	Grab sampling, alpha scintillation	46.4	16.40
Wilkes Barre University Wilkes Barre, PA	Continuous, solid-state	-	0.15

TABLE 3

RESULTS OF THE RADON INTERCOMPARISON MEASUREMENTS
(Radon Concentration, Bq m^{-3})

Instrument	Participant		Reference Mean ± Error	Participant Reference
	Range	Mean ± σ_g		
Activated Collectors	936 - 969	949 ± 14	921 ± 10	1.03 ± .016
	969 - 977	974 ± 4	923 ± 10	1.05 ± .016
	932 - 955	943 ± 10	922 ± 10	1.02 ± .020
	807 - 858	844 ± 25	921 ± 10	0.92 ± .048
	951 - 1040	984 ± 40	924 ± 10	1.07 ± .024
	940 - 1051	1018 ± 55*	917 ± 10	1.11 ± .060
	840 - 1044	916 ± 77*	917 ± 10	1.00 ± .080
	814 - 855	836 ± 18	923 ± 10	0.91 ± .029
	999 - 1165	1053 ± 45*	927 ± 10	1.13 ± .040
	891 - 988	943 ± 40	924 ± 10	1.02 ± .040
	881 - 962	920 ± 33	932 ± 10	0.99 ± .036
	858 - 884	867 ± 15	922 ± 10	0.94 ± .090
	841 - 1094	977 ± 115	922 ± 10	1.06 ± .110
Nuclear Track	785 - 854	833 ± 31	924 ± 10	0.90 ± .320
	780 - 1130	965 ± 31	924 ± 10	1.04 ± .160
	679 - 806	735 ± 55	924 ± 10	0.80 ± .087
	947 - 971	956 ± 11	924 ± 10	1.03 ± .016
	932 - 952	950 ± 11	924 ± 10	1.03 ± .016
	1100 - 1290	1160 ± 88	924 ± 10	1.25 ± .090
	753 - 982	828 ± 74	924 ± 10	0.90 ± .079
	678 - 780	751 ± 22	924 ± 10	0.81 ± .050
	568 - 721	641 ± 54	924 ± 10	0.69 ± .049
	1036 - 1221	1147 ± 98	939 ± 10	1.22 ± .170
Electret/Ionization Chambers	839 - 897	864 ± 24	923 ± 10	0.94 ± .090
	884 - 1043	940 ± 70	924 ± 10	1.02 ± .100
	810 - 877	852 ± 32	924 ± 10	0.92 ± .035
	777 - 903	825 ± 54	923 ± 10	0.89 ± .097
	833 - 910	877 ± 33	923 ± 10	0.95 ± .037
	903 - 925	914 ± 12	924 ± 10	0.99 ± .016
	869 - 929	887 ± 28	922 ± 10	0.96 ± .040
	777 - 851	814 ± 36	929 ± 10	0.88 ± .040
	1033 - 1447	1276 ± 200	941 ± 10	1.35 ± .100

TABLE 3 (Cont' d)

Instrument	Participant Range	Participant Mean ± σ_g	Reference Mean ± Error	Participant Reference
Continuous Passive/Active		925 ± 25	919 ± 10	1.00 ± .020
		903 ± 24	919 ± 10	1.00 ± .020
		910 ± 56	922 ± 10	0.99 ± .060
		955 ± 48	922 ± 10	1.04 ± .040
		881 ± 26	922 ± 10	0.96 ± .030
		899 ± 33	922 ± 10	0.98 ± .040
		939 ± 49	911 ± 10	1.03 ± .070
		860 ± 40	922 ± 10	0.93 ± .040
		860 ± 28	922 ± 10	0.93 ± .020
		844	926 ± 10	0.91
		933 ± 7	923 ± 10	1.00 ± .013
		870 ± 33	929 ± 10	0.94 ± .030
		925 ± 7	928 ± 10	1.00 ± .013

* Liquid scintillation counting
± The error associated with the participant's average value is the total error of the measurement.

TABLE 4

RADON PROGENY INTERCOMPARISON MEASUREMENTS

Participant and Method	Cond. Nucl. Particles x 10^3 (cm^{-3})	Ratio Participant/Reference ^{218}Po	^{214}Pb	^{214}Bi	PAEC	Reference PAEC ($nJ\ m^{-3}$)
Grab						
1-1	5.6	0.93	0.98	1.05	0.97	693 ± 8
1-2	5.6	1.00	1.01	0.84	0.97	723 ± 10
1-3	5.6	1.20	1.06	0.68	1.01	720 ± 16
1-4	7.0	0.86	0.89	1.05	0.91	771 ± 11
1-5	12.0	1.17	1.00	0.86	1.01	936 ± 16
1-6	12.0	0.88	0.88	1.10	0.93	867 ± 11
Mean and σ_g		(1.00 ± .14)	(0.97 ± .07	(0.93 ± .16)	(0.97 ± .04)	(785 ± 96)
2-1	5.6	0.92	1.00	1.22	1.01	692 ± 8
2-2	5.6	1.03	0.98	1.05	1.01	723 ± 10
2-3	5.6	0.93	0.96	1.03	0.97	720 ± 16
2-4	7.0	0.96	0.90	1.21	0.98	771 ± 11
2-5	12.0	0.99	1.10	0.98	1.03	936 ± 16
2-6	12.0	1.14	1.18	0.87	1.08	867 ± 11
Mean and σ_g		(1.00 ± .08)	(1.02 ± .10)	(1.06 ± .14)	(1.01 ± .04)	(785 ± 96)
3-1	5.6	1.00	1.00	214Bi	1.05	692 ± 8
3-2	5.6	-	-	1.25	1.03	723 ± 10
3-3	5.6	1.00	1.13	-	1.06	720 ± 16
3-4	7.0	1.43	0.82	1.01	0.92	771 ± 11
3-5	12.0	0.93	1.02	1.32	1.03	936 ± 16
3-6	12.0	1.20	1.24	1.13	1.11	867 ± 11
3-7	3.0	1.01	1.15	0.78 0.81	1.02	558 ± 11
Mean and σ_g		(1.09 ± .19)	(1.06 ± .15)	(1.05 ± .22)	(1.03 ± .06)	(752 ± 11)
4-1	7.0	-	-	-	1.00	771 ± 11
4-2	12.0	-	-	-	0.93	936 ± 16
4-3	12.0	-	-	-	0.96	867 ± 11
4-4	3.0	-	-	-	1.04	558 ± 11
Mean and σ_g					(0.98 ± .05)	(783 ± 164)
Continuous or Integrating						
5					0.67	470 ± 10
6					0.68	498 ± 11
7					0.81	566 ± 10
8					0.88	566 ± 10
9					0.92	566 ± 10
10					0.96	566 ± 10
11					0.62	324 ± 8
12					0.98	444 ± 15
13					1.03	474 ± 15
14					0.57	428 ± 15
15					0.73	852 ± 11

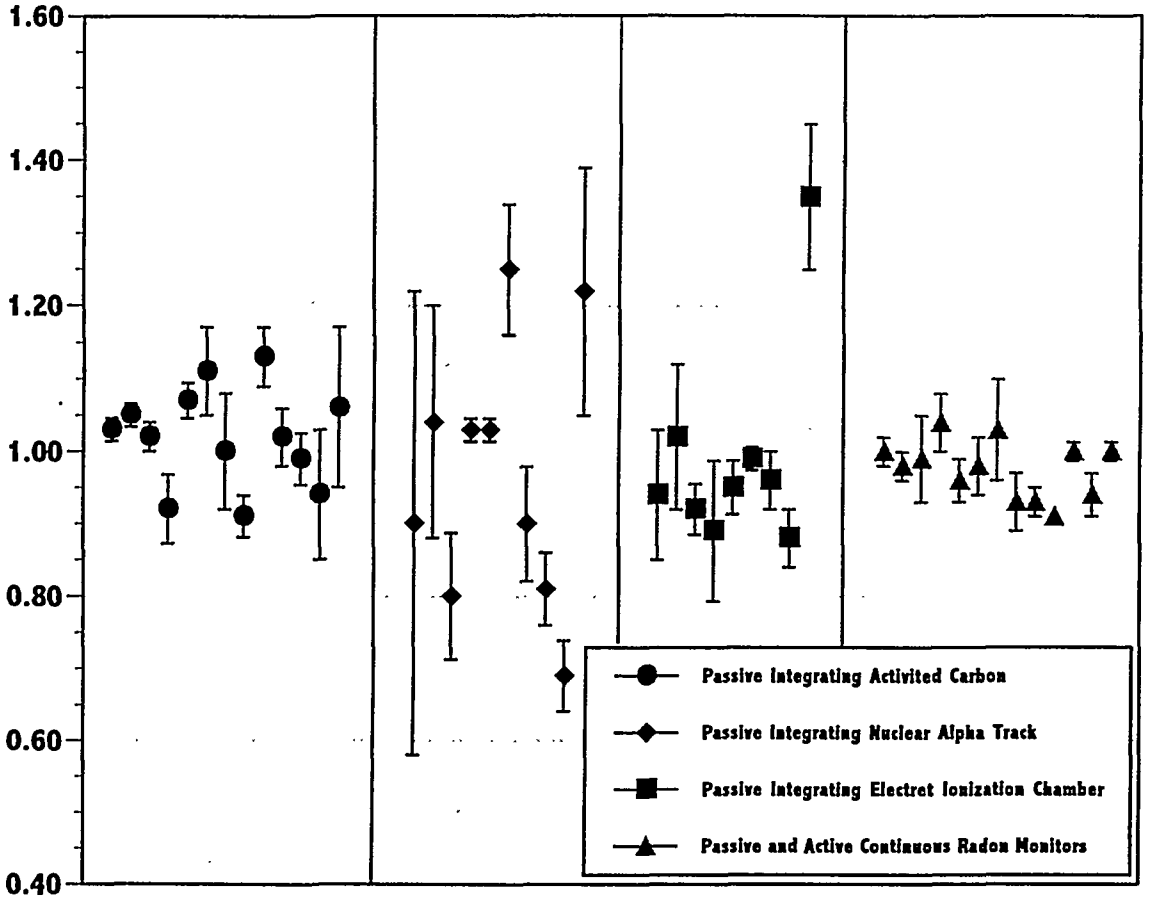


Figure 1. Intercomparison of active and passive radon integrating instruments.

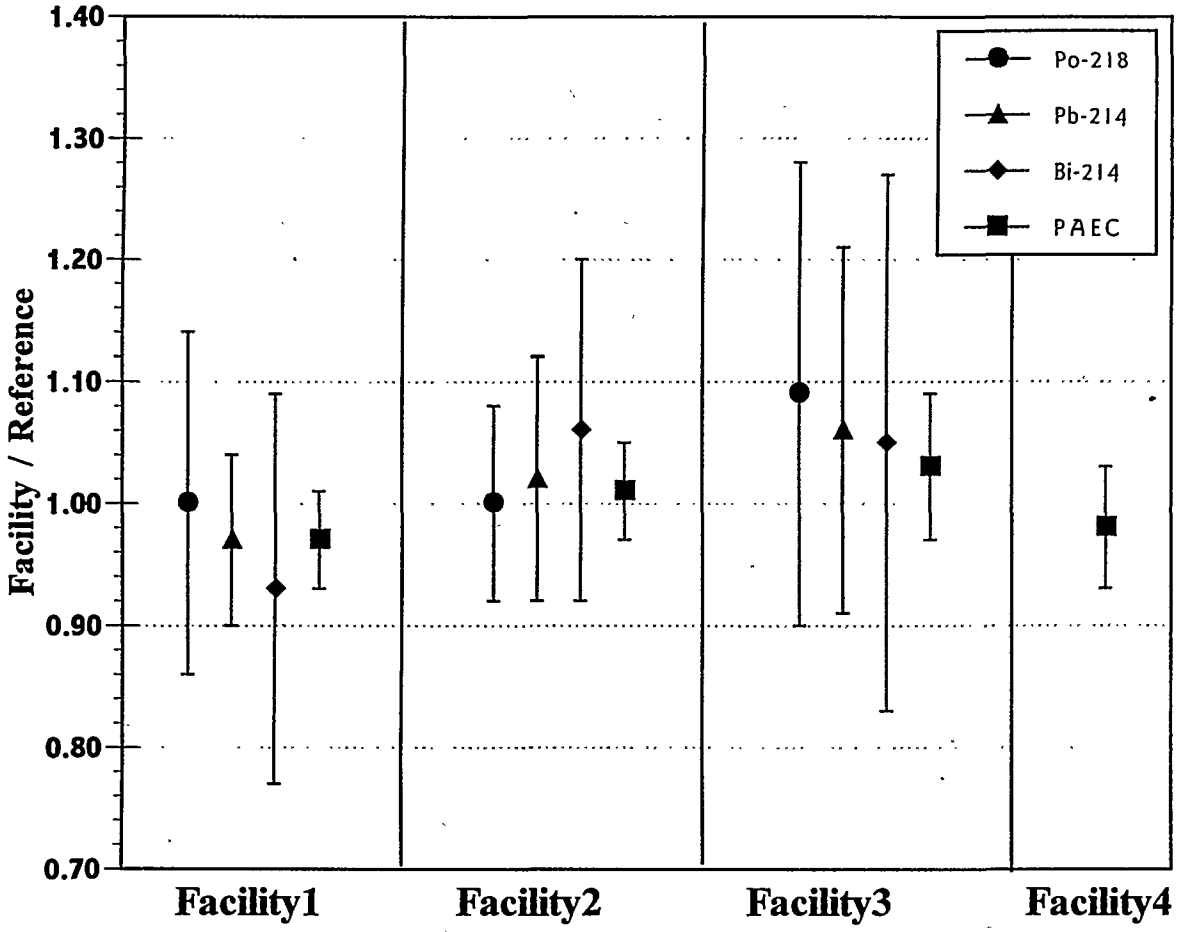


Figure 2. Intercomparison of individual radon progeny and PAEC.

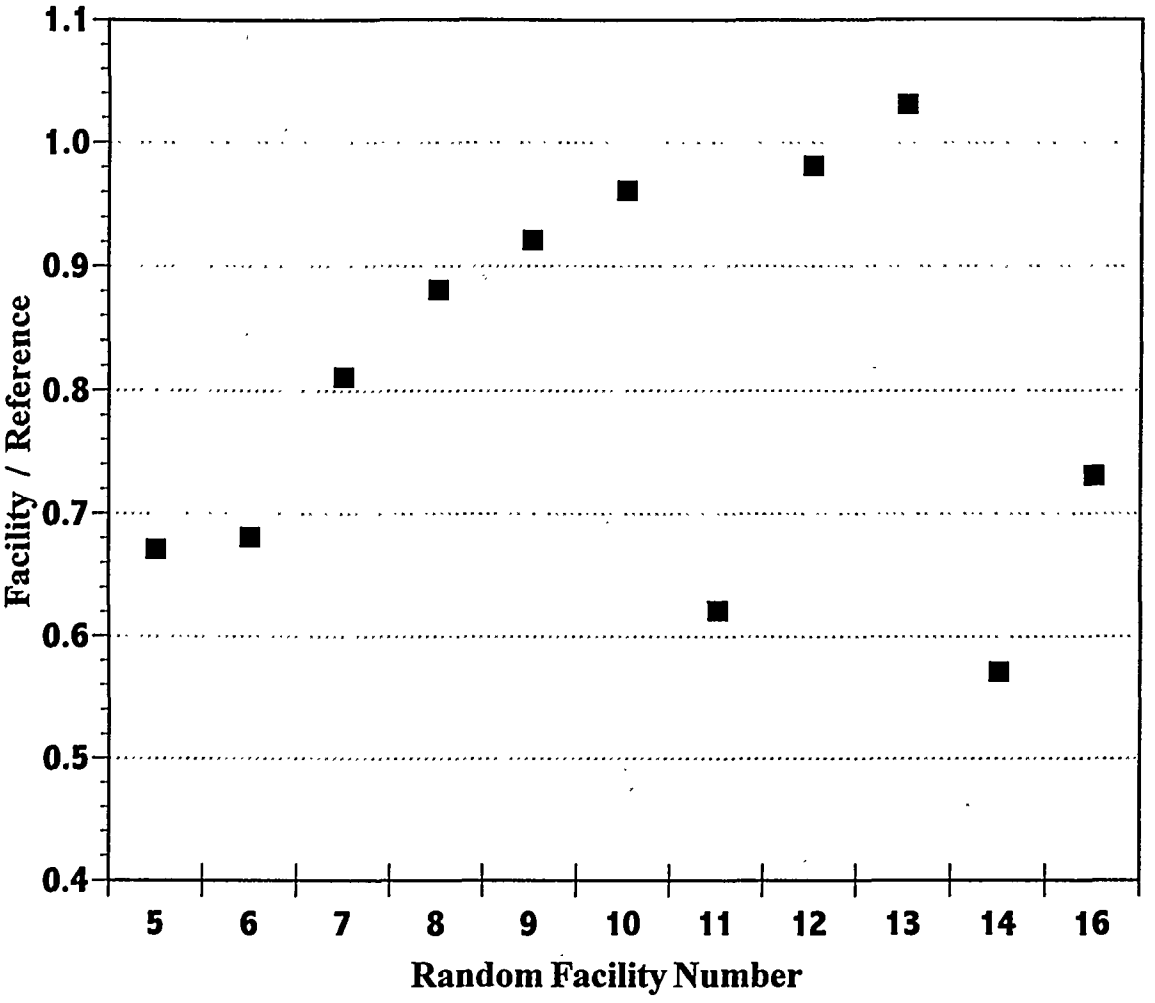
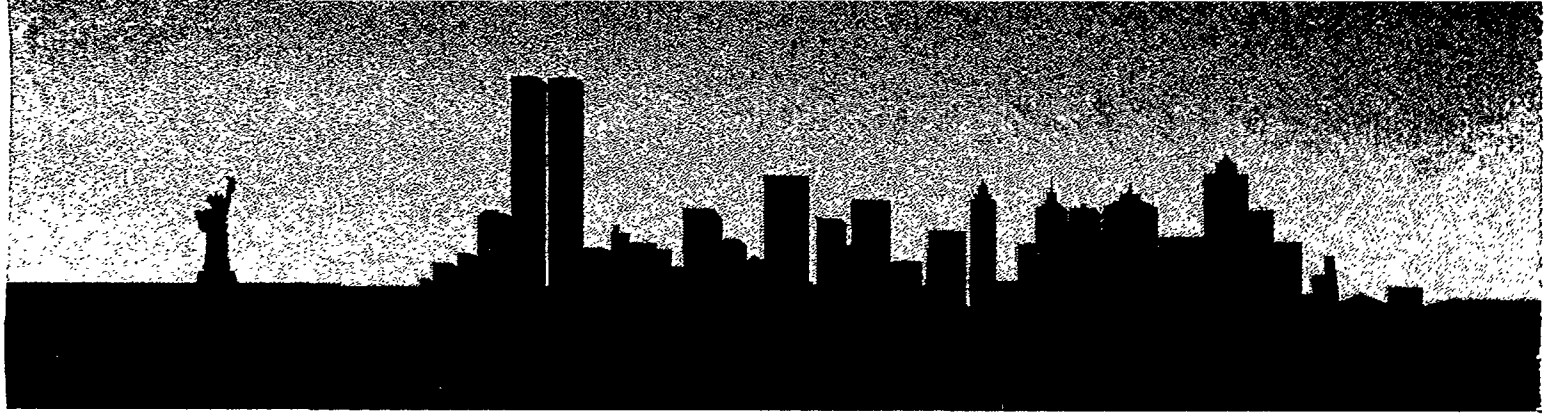


Figure 3. Intercomparison of continuous and integrating PAEC instruments.



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