EML-566

Environmental Measurements Laboratory

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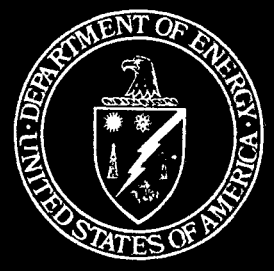
February 1995



DEPARTMENT OF ENERGY
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ABSTRACT

An intercomparison exercise for radon and radon progeny instruments and methods was held at the Environmental Measurements Laboratory (EML) from April 22-May 2, 1994. The exercise was conducted in the new EML radon test and calibration facility in which conditions of exposure are very well controlled. The detection systems of the intercompared instruments consisted of: 1) pulse ionization chambers, 2) electret ionization chambers, 3) scintillation detectors, 4) alpha particle spectrometers with silicon diodes, surface barrier or diffused junction detectors, 5) registration of nuclear tracks in solid-state materials, and 6) activated carbon collectors counted by gamma-ray spectrometry or by alpha- and beta- liquid scintillation counting. Twenty three private firms, government laboratories and universities participated with a 165 passive integrating devices consisting of: activated carbon collectors, nuclear alpha track detectors and electret ionization chambers, and 11 active and passive continuous radon monitors. Five portable integrating and continuous instruments were intercompared for radon progeny. Forty grab samples for radon progeny were taken by five groups that participated in person to test and evaluate their primary instruments and methods that measure individual radon progeny and the potential alpha energy concentration (PAEC) in indoor air. The results indicate that more than 80% of the measurements for radon performed with a variety of instruments, are within ±10% of the actual value. The majority of the instruments that measure individual radon progeny and the PAEC gave results that are in good agreement with the EML reference value. Radon progeny measurements made with continuous and integrating instruments are satisfactory with some room for improvement.

I. Introduction

To determine the condition and state of monitoring devices for radon and radon progeny measurements, EML conducts annual intercomparison exercises in an atmosphere similar to that of the indoor environment. These type of regional intercomparisons are recommended by the Coordinated Research Program (CRP) of the International Intercomparison and Intercalibration Program (IIIP), under the sponsorship of the International Atomic Energy Agency (IAEA) in cooperation with the Commission of European Communities (CEC). The IAEA initiated this project to encourage research aimed at generating high quality data on radon exposures in the human environment, and to provide guidance on the assessment and control of exposures to radon and its progeny.

With the exception of some instruments that provided results on site, all the others were returned to the participating groups for analysis. Upon analysis the results were sent to EML for comparison and evaluation. The evaluation of the different radon and radon progeny instruments is the subject of this report.

II. Exposure and Test Facility

For all of the tests carried out during the intercomparison, EML's new 30 M^3 radon and radon progeny test chamber was used (Knutson and George, in press). The new test chamber became operational in March 1994. During exposure, conditions of radon/progeny concentration, temperature, relative humidity and particle concentration were well controlled. Wax particles and burning candle aerosols were generated when needed to test active devices that measure radon progeny. Test durations ranged from 1 to 10 days and the concentration of radon was kept at about 1050 Bq m^{-3}. The gamma background inside the EML test chamber was constant at 0.24 $\mu Sv\ h^{-1}$ (24 $\mu R\ h^{-1}$). The instruments were placed inside the test chamber 0.5-1.5 m above the floor. Grab sampling by the participants for radon progeny was conducted from the adjacent

room by taking samples from inside the test chamber through sampling ports. For the analysis of the collected radioactivity on filters and the calculation of the concentration of the individual radon progeny and PAEC, the participants used the modified Tsivoglou method (Thomas, 1972) and least squares method (Raabe and Wrenn, 1969).

The concentration of radon and radon progeny in the test chamber was measured continuously by means of two radon and two PAEC monitors. During grab sampling for the PAEC, tests were conducted at different concentration levels by adjusting the wax aerosol concentration to achieve the desired PAEC level. The wax aerosol was generated by a TSI condensation aerosol generator. At other times, a lit candle provided very high concentrations of airborne particulates. The particle concentration inside the test chamber was monitored continuously with a condensation nucleus counter. The geometric mean diameters (GMD) of the airborne particles during the three aerosol conditions were: 1) candle particles, GMD = 80 nm and GSD = 2.4; 2) wax particles, GMD = 73-149 nm and GSD =1.6-2.12; and 3) in the absence of generated particles the ambient aerosols in the chamber consisted of particles with GMD = 85 nm and GSD = 2.3. Temperature and relative humidity (RH) were regulated and maintained within ±2% of the projected values.

III. Quality Assurance

The concentration of the radon in the test chamber was measured continuously with two flow through scintillation cell monitors which were calibrated in a known atmosphere of radon traceable to the National Institute of Standards and Technology (NIST) (Fisenne and Keller, 1985). The total range of uncertainty in the EML reference value is estimated to be < 5%. Radon progeny measurements made with EML instruments and methods (Thomas, 1972; Raabe and Wrenn, 1969) are accurate to within 3% at the concentration levels tested. Also both methods have been intercalibrated and intercompared on numerous occasions with several reference laboratories and were found to be within ±5% of each other.

IV. Radon and Radon Progeny Instruments

The instruments and methods for the radon measurements are listed in Table 1. The instruments used for the radon progeny measurements are listed in Table 2. The passive integrating devices for radon included: 1) several types of activated carbon collectors (size, shape, open faced or with diffusion barrier); 2) electret/ionization chambers (two different types); 3) nuclear track detectors (twelve different configurations; and 4) pulse/ionization chambers (two types). Active and passive scintillation cell monitors (four types) and solid-state detection monitors (two types) were also included. The active instruments or methods for radon progeny measurements included grab, integrating and continuous by sampling on filters that were counted by solid state and scintillation detectors or by registration of alpha tracks in solid-state materials. In grab sampling, the PAEC was varied by the addition of particles generated by the wax aerosol generator.

V. Results and Discussion

The results of the intercomparison of instruments and methods for radon and those for individual radon progeny and PAEC are listed in Tables 3 and 4 and are also shown in Figures 1-3, respectively. The EML values were used as the reference against which all other measurements were compared. The uncertainty of the radon measurements made by EML is between 2-3%. To maintain participant confidentiality, the reported values are shown randomly. For comparison purposes, the different types of radon instruments were grouped separately into four categories 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 group data are compared with the mean value obtained by EML during the same test period. Since all of the individual measurements for radon had error estimates, the error propagation of the ratios (participant/EML) were calculated and are listed in the last column of Table 3 and are illustrated in Figure 1.

The measured mean ratios and their SDs for the different instrument categories are: activated carbon collectors = 1.04 ± 0.10; nuclear alpha track = 0.96 ± 0.09; electret/ionization chambers = 0.97 ± 0.03; and continuous active and passive commercial instruments = 1.01 ± 0.05. Among the activated carbon collectors, those with diffusion barriers produced the best data. Only one participant's collectors overresponded by 20%, probably due to a bias in the calibration. Open-faced collectors in general overresponded by 10%, suggesting slight sensitivity to air movement in the radon chamber. Environmental Protection Agency (EPA) studies indicate that air movement affects only some open-faced activated carbon when the air velocity in the test chamber is between 10-20 linear feet per minute (LFM) (Gray, 1987, 1988). The air circulation in the EML test chamber during the intercomparison was about 10 M^3 min^{-1}, translating into an air velocity of about 1 LFM. To obtain accurate results, open-faced collectors should be exposed under conditions of minimal air movement or the conditions of exposure must be documented so that the proper correction factor can be applied.

The mean ratio of the 12 sets of nuclear alpha track detectors was 4% lower than the reference value, indicating that generally very good results can be obtained. Except for three participants who seemed to have a 15-20% low calibration bias, the remainder of participants did exceptionally well. The range of the mean ratios was 0.81-1.10 as compared to 0.69-1.80 in a similar exercise in 1991 and to 0.72-1.02 in 1993 (George, 1991; George and Tu, 1993). It appears that good measurement quality control (QC) is exercised by the users of nuclear track detectors. Since the number of participants is large (12 groups), it seems that nuclear alpha track detectors are becoming one of the most commonly used long-term exposure monitors for radon.

The number of participants using short-term and long-term electret/ionization chambers increased to nine as compared to four during the 1993 exercise. The mean ratio of the participant/reference value was 0.97 ± 0.03, indicating high precision and accuracy. The radon monitors were supplied from two sources but the analysis was performed by several participants. The high precision obtained with these monitors is reflected in the exercise of good QC by the manufacturer of the electrets.

Continuous active and passive commercial radon instruments performed very well. The mean ratio and the SD of the different instruments was 1.01 ± 0.05, indicating the use of calibrated and properly maintained instruments. A substantial improvement was noted from the 1993 intercomparison in which both positive and negative calibration biases were observed.

The measurement results for the individual radon progeny concentrations and PAEC obtained by the four participants are listed randomly in Table 4 and are shown in Figures 2 and 3. Table 4 shows the ratios of the individual radon progeny and PAEC measured by the four participants to that of the EML reference value using simultaneous grab sampling. The last column lists the concentration of the reference radon progeny atmosphere. The accuracy of the PAEC measurements were calculated from counting statistics alone. Except for participant No. 4, the data indicate that the ^{218}Po ratio values are in very good agreement for both low- and high-radon progeny concentrations; participant No. 4 was biased towards low measurements. The data for ^{214}Pb and ^{214}Bi are mixed, with participant No. 4 being closer in agreement with the EML reference value. The PAEC ratios for participants 1-3 are in very good agreement with each other, indicating very close agreement with the EML reference value. Participant No. 4 showed the PAEC ratio to be low (0.86-0.90), indicating a low bias in calibration possibly as a result of using inappropriate air volume or counter efficiency.

The results from continuous and integrating instruments that measure the PAEC shown in Figure 3 indicate ratios ranging from 0.66-1.05 with a mean and SD of 0.89 ± 0.14. About 70% of the instruments gave results within ±20% of the reference value. The instruments with low readings are suspected to measure less radon progeny due to either plateout effects or to inappropriate air volume and counting efficiency. A similar trend was observed by EPA in its radon measurement proficiency testing program that began in 1986. This discrepancy results from several contributing factors such as: the collection of radon progeny under different conditions of radioactive equilibrium; the stability of air pumps and the accuracy of the air volume sampled; the integrity of the filter media and the filter holders; and the counting geometry and efficiency of the detector system.

Conclusions

The state of the art instruments and methods for active and passive radon measurements and methods in North America and Canada are in good standing. Although the instruments are based on different principles of collection and detection, more than 80% of the measurements for radon were within 10% of the EML reference value. The few outliers were from first time participants using activated carbon collectors and nuclear alpha track detectors with product QC problems and calibration bias. Commercial electronic instruments for radon performed well, indicating that they were calibrated and maintained by both the manufacturer and the user.

The majority of the participants that used grab sampling for the measurement of the concentrations of individual radon progeny and PAEC show very good agreement with the EML reference values. One participant who reported lower values by 15% needs to examine the sources of the problem. The quality of the PAEC measurements made with continuous and integrating commercial instruments is satisfactory. However, there is an indication that some instruments have a calibration bias with the possible causes being plateout of the radon progeny, an inappropriate total volume of sampled air, and improper counter efficiency.

Overall, the intercomparison exercises demonstrate once again the state of instrumentation, and points out areas where improvements need to be made to ensure that accurate measurements are carried out for radon and radon progeny for the assessment of human exposure by inhalation.

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TABLE 1

PASSIVE AND ACTIVE RADON INSTRUMENTS USED BY PARTICIPANTS

Participant	Instrument/Method
AECL-Low Level Rad. Waste Ottawa, Canada	PI, electret/ionization chamber sensitive volume = 0.2 L
Air Chek Arden, NC	PI, carbon collector (DB), gamma count
Alpha Spectra Lakewood, CO	PI, carbon collector (DB), gamma count
Bowser/Morner Dayton, OH	PI, carbon collector (OF), gamma count
Enviroserv Inc. Morristown, NJ	PI, carbon collector, (OF), gamma count
femto-TECH Inc. Carisle, OH	PC, pulse ionization chamber (CRM 510) sensitive volume = 0.2 L
Gemini Research Timonium, MD	PI, nuclear track (NYU type) AC, scintillation cell (Certifier II)
Health/Welfare of Canada Ottawa, Ontario, Canada	PI, electret/ionization chamber sensitive volume = 0.2 L
Landauer Inc. Glenwood, IL	PI, nuclear track
Niton Corporation Bedford, MA	PI, carbon collector, liquid scintill. AC, solid state alpha spectrometry sensitive volume = 0.6 L
Northeastern Laboratory Winslow, Maine	PI, carbon collector, liquid scintill.
NY State Department of Health Albany, NY	PI, carbon collector (OF), gamma count

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

TABLE 1 (Cont'd)

Participant	Instrument/Method
Pennsylvania DER Harrisburg, PA	AC, scintillation cell (RGM-3) sensitive volume = 3.2 L AC, solid state alpha spectrometry sensitive volume = 0.6 L PI, carbon collector (DB) gamma count PI, electret/ionization chamber sensitive volume = 0.2 L
Pylon Electronics Inc. Ottawa, Ontario, Canada	PC, scintillation cell (Pylon AB-5) sensitive volume = 0.27 L
Rad Elec Frederick, MD	PI, electret/ionization chamber sensitive volume = 0.2 L
Radon Envir. Monitoring Northbrook, IL	PI, carbon collector, liquid scintill. PI, nuclear track
Radon QC Lakewood, CO	AC, pulse ioniz. chamber (Alphaguard) sensitive volume = 0.554 L
RSSI Morton Grove, IL	PI, nuclear track
RTCA Irvington, NY	PI, carbon collector (OF) gamma count
Safety Analysis Unit Chiba, Japan	PI, nuclear track
St. Johns University Collegesville, MN	PI, nuclear track (type LAE, LCE, LEE, LRE and PCE)
Ukranian Research Center Ukraine	PI, nuclear track
Wilkes Barre University Wilkes Barre, PA	PI, electret/ionization chamber PI, carbon collector (DB), gamma count AC, scintillation cell AB-5 PC, scintillation cell AB-5

TABLE 2

ACTIVE INSTRUMENTS AND METHODS FOR MEASURING PAEC

Participant	Method-Detection System	Counting Efficiency (%)	Flow Rate (L min^{-1})
EML (Alpha Prism) New York, NY	Cont. Alpha Silicon Detector	17.0	0.13
AECL Ottawa, Canada	Grab, Alpha Scintillation	47.0	11.70
Canad. Inst. of Rad. Safety Saskatoon.	Integrating Nuclear Track	-	1.00
Bowser/Morner Dayton, OH	Grab, Alpha Scintillation	48.3	11.20
Enviroserv (A-Prism) Morristown, NJ	Cont. Alpha Silicon	17.0	0.12
Gemini Research Timonium, MD	Cont. 500 mm^2 Silicon	32.0	0.25
PA DER Harrisburg, PA	Cont. Alpha Silicon Detector	17.0	0.12
Pylon Electronics Ottawa, Canada	Grab, Alpha Scintillation Cont. Solid State WLx	48.5 -	9.10 1.00
USEPA, NAREL Montgomery, AL	Grab, Alpha Scintillation	47.1	17.20
Wilkes University Wilkes Barre, PA	Cont. Solid-State Detector	-	0.15

TABLE 3

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

Instrument	Participant Range	Participant Mean ± σ	Reference Mean ± Error	Participant/ Reference
Activated carbon	1110-1158	1123 ± 80	1054 ± 20	1.06 ± .034*
	1055-1095	1073 ± 17	1042 ± 20	1.03 ± .040
	1029-1066	1048 ± 18	1042 ± 20	1.00 ± .001*
	1221-1387	1284 ± 72	1062 ± 20	1.20 ± .024
	1092-1132	1113 ± 17	1051 ± 20	1.06 ± .025
	1040-1110	1073 ± 28	1051 ± 20	1.02 ± .040
	1084-1232	1169 ± 63	1042 ± 20	1.12 ± .120
	1043-1162	1108 ± 55	1056 ± 20	1.05 ± .070
	1128-1254	1208 ± 57	1042 ± 20	1.16 ± .042
	762- 951	861 ± 67	1054 ± 20	0.83 ± .035*
	881-1021	955 ± 61	1057 ± 20	0.90 ± .033*
	733-1043	925 ± 130	1062 ± 20	0.87 ± .033*
	1017-1158	1080 ± 62	1050 ± 20	1.03 ± .042
	1095-1173	1142 ± 34	1053 ± 20	1.08 ± .043
	1217-1272	1246 ± 25	1053 ± 20	1.18 ± .050
Nuclear track	1018-1180	1102 ± 68	1062 ± 20	1.04 ± .070
	781- 936	856 ± 64	1051 ± 20	0.81 ± .170
	920-1080	1000 ± 68	1051 ± 20	0.95 ± .068
	970-1030	1040 ± 54	1051 ± 20	0.99 ± .050
	810- 962	853 ± 73	1051 ± 20	0.81 ± .050
	973-1018	989 ± 21	1051 ± 20	0.97 ± .068
	1018-1185	1095 ± 73	1051 ± 20	1.04 ± .055
	928-1201	1088 ± 115	1051 ± 20	1.04 ± .110
	971-1042	1007 ± 34	1051 ± 20	0.96 ± .037
	1060-1240	1158 ± 94	1051 ± 20	1.10 ± .08
	1008-1037	1023 ± 15	1051 ± 20	0.97 ± .042
	1068-1254	1179 ± 88	1051 ± 20	1.12 ± .087
	834- 948	892 ± 56	1051 ± 20	0.85 ± .055

TABLE 3 (Cont'd)

Instrument	Participant Range	Participant Mean ± σ	Reference Mean ± Error	Participant/ Reference
Electret/	895-1010	964 ± 56	1051 ± 20	0.92 ± .050
ionization	980-1097	1045 ± 59	1042 ± 20	1.00 ± .059
chamber	998-1084	1034 ± 43	1042 ± 20	0.99 ± .050
	974-1002	976 ± 27	1051 ± 20	0.93 ± .049
	1018-1069	1047 ± 21	1051 ± 20	1.00 ± .053
	992-1073	1036 ± 34	1052 ± 20	0.98 ± .050
	1036-1055	1045 ± 9	1051 ± 20	0.99 ± .051
	921-1092	1020 ± 72	1050 ± 20	0.97 ± .050
	951-1058	1025 ± 50	1055 ± 20	0.97 ± .044
Continuous+		1106 ± 19	1055 ± 20	1.05 ± .026
Passive		1086 ± 41	1055 ± 20	1.03 ± .043
and Active		1084 ± 19	1054 ± 20	1.03 ± .026
		1143 ± 19	1054 ± 20	1.08 ± .027
	1132-1184	1158 ± 37	1053 ± 20	1.10 ± .040
		995 ± 50	1057 ± 20	0.94 ± .050
	932-1110	1025 ± 70	1062 ± 20	0.97 ± .068
		1017 ± 37	1048 ± 20	0.97 ± .040
		1006 ± 40	1050 ± 20	0.96 ± .042
		1055 ± 37	1055 ± 20	1.00 ± .039
		1029 ± 52	1055 ± 20	0.98 ± .052

*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 Random No. and Sampling Method	Cond. Nucl. Particles (cm^{-3})	Ratio Participant/Reference				Reference PAEC ($nJ\ m^{-3}$)
		^{218}Po	^{214}Pb	^{214}Bi	PAEC	
1-1 Grab	3.3 K	0.88	0.99	1.10	0.94	500 ± 19
1-2	3.2 K	0.96	1.19	0.79	1.01	553 ± 12
1-3	3.0 K	0.93	1.12	0.94	0.9	535 ± 12
1-4	2.8 K	1.01	1.48	0.67	1.06	473 ± 23
	Mean σ_g	(.95±.05)	(1.2±.20)	(.88±.19)	(1.00±.05)	(515 ± 35)
1-5 Grab	16.0 K	0.94	1.00	1.05	0.99	1416 ± 17
1-6	30.0 K	0.86	1.00	0.92	0.94	1824 ± 21
1-7	16.0 K	1.12	1.06	0.88	1.02	1734 ± 21
	Mean σ_g	(.97±.13)	(1.02±.03)	(.95±.09)	(.98±.04)	(1658 ± 21)
2-1 Grab	3.3 K	1.06	0.96	0.88	1.00	500 ± 19
2-2	3.2 K	1.00	0.86	1.10	0.96	553 ± 12
2-3	3.0 K	1.03	1.23	0.83	1.03	535 ± 12
2-4	2.8 K	0.77	0.97	1.38	0.98	473 ± 23
	Mean σ_g	(.97±.13)	(1.00±.15)	(1.04±.25)	(.99±.03)	(515 ± 35)
2-5 Grab	16.0 K	0.97	0.98	1.00	0.98	1416 ± 17
2-6	30.0 K	0.91	1.05	0.90	0.98	1824 ± 21
2-7	16.0 K	1.27	1.13	0.74	1.03	1734 ± 21
	Mean σ_g	(1.05±.19)	(1.05±.07)	(.88±.130	(1.00±.03)	(1658 ± 21)
3-1 Grab	3.3 K	1.00	0.96	1.01	1.00	500 ± 19
3-2	3.2 K	0.95	1.00	1.21	0.98	553 ± 12
3-3	3.0 K	1.13	1.17	0.72	1.01	535 ± 12
3-4	2.8 K	1.22	1.33	0.68	1.10	473 ± 23
	Mean σ_g	(1.07±.12)	(1.11±.17)	(.91±.25)	(1.02±.05)	(515 ± 35)
3-5	16.0 K	0.98	1.05	0.99	1.01	1416 ± 17
3-6	30.0 K	0.99	1.06	0.93	1.01	1824 ± 21
3-7	16.0 K	1.00	1.00	0.98	1.00	1734 ± 21
	Mean σ_g	(.99±.01)	(1.03±.03)	(.97±.05)	(1.01±.01)	(1658 ± 21)
4-1 Grab	3.3 K	0.68	0.64	1.56	0.79	500 ± 19
4-2	3.2 K	0.61	0.53	1.52	0.73	553 ± 12
4-3	3.0 K	0.93	1.32	0.63	0.97	535 ± 12
4-4	2.8 K	0.87	1.22	0.75	0.95	473 ± 23
	Mean σ_g	(.77±.15)	(.92±.40)	(1.12±.49)	(.86±.12)	(515 ± 35)

TABLE 4 (Contd.)

Participant Random No. and Sampling Method	Cond. Nucl. Particles cm^{-3}	Ratio Participant/Reference ^{218}Po	^{214}Pb	^{214}Bi	PAEC	Reference PAEC ($nJ\ m^{-3}$)
4-5	16.0 K	0.86	0.92	0.94	0.91	1416 ± 17
4-6	30.0 K	0.67	0.83	1.12	0.87	1824 ± 21
4-7	16.0 K	1.03	0.98	0.77	0.93	1734 ± 21
	Mean σ_g	(.85±.13)	(.91±.08)	(.94±.18)	(.90±.03)	(1658 ± 21)
5 Continuous					0.76	1121 ± 25
6 "					1.05	575 ± 15
7 "					0.77	585 ± 15
8 "					0.66	585 ± 15
9 "					1.00	816 ± 20
10 "					0.89	830 ± 20
11 "					0.80	1647 ± 40
12 "					1.02	825 ± 20
13 "					1.05	825 ± 20
14 Integrating					0.88	824 ± 20

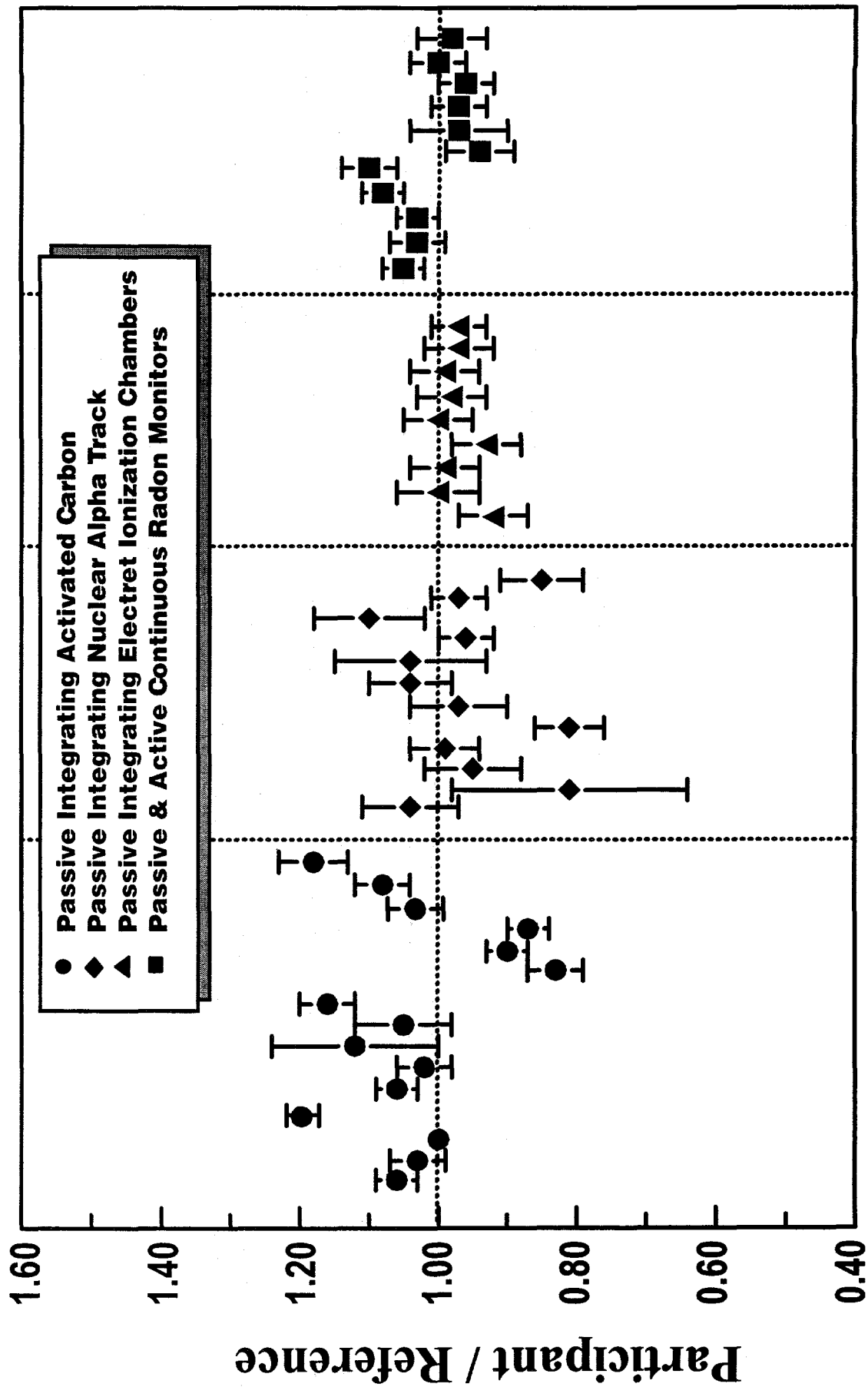

Figure 1. Intercomparison of active and passive integrating instruments.

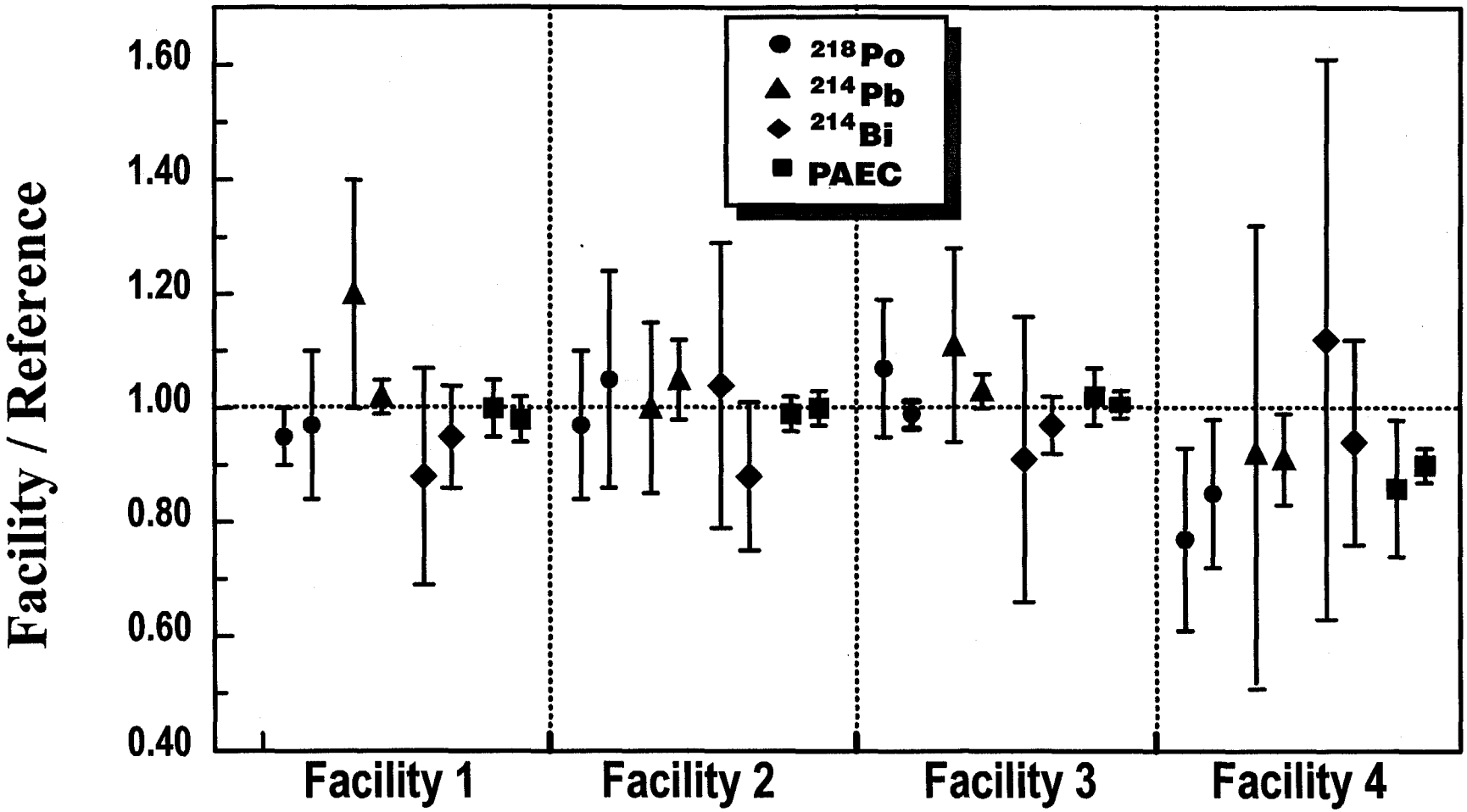

Figure 2. Intercomparison of individual radon progeny and PAEC.

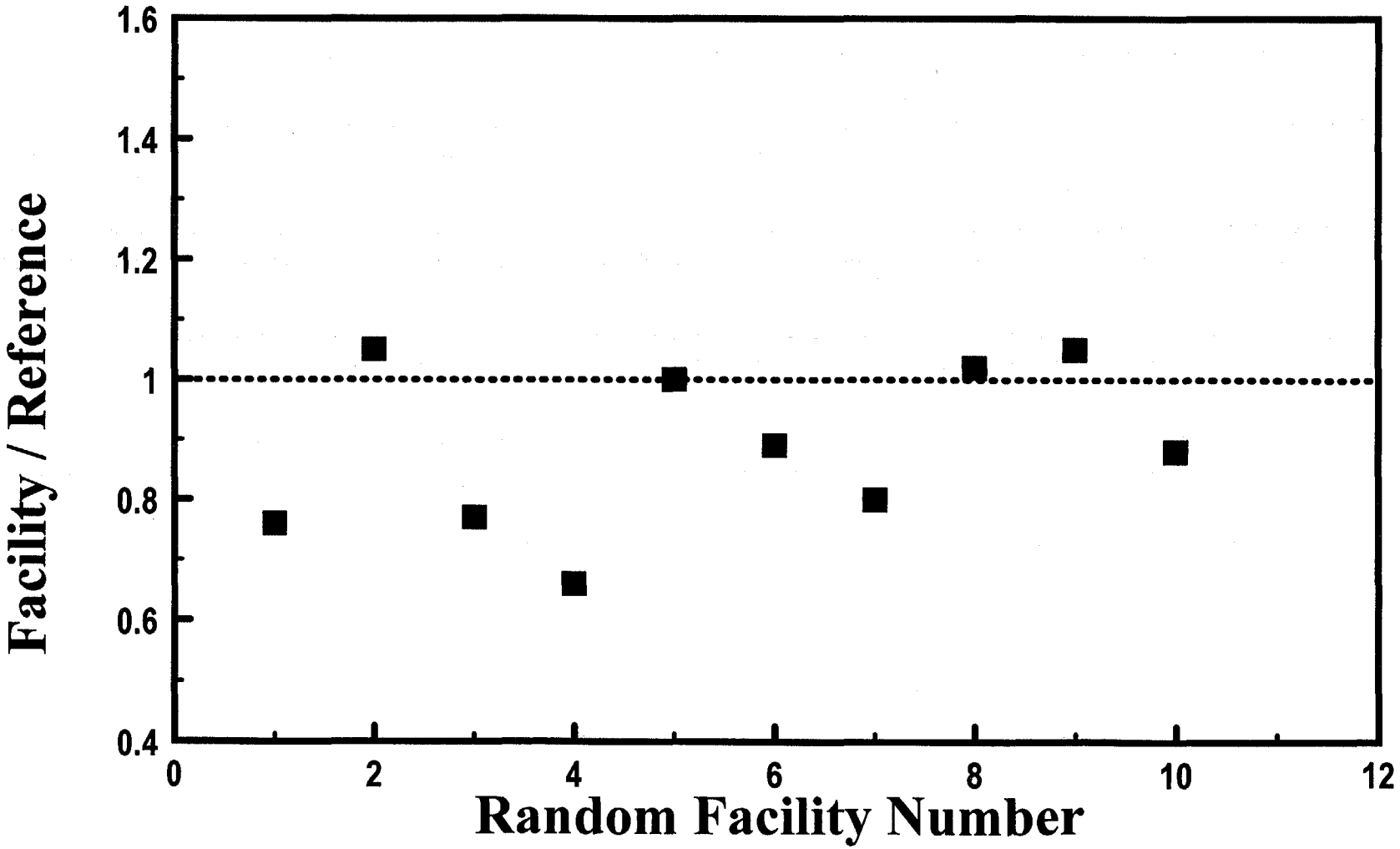

Figure 3. Intercomparison of continuous and integrating PAEC instruments.