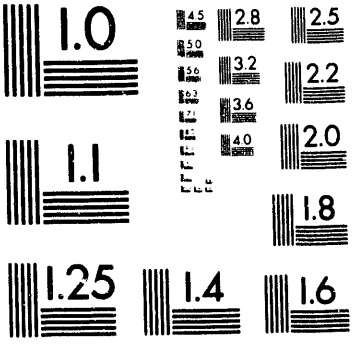
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1 of 1

EML-554

Intercomparison and Intercalibration of Passive/Active Radon and Active Radon Progeny Instruments and Methods in North America

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ABSTRACT

An intercomparison and intercalibration exercise for radon and radon progeny measurements made with active and passive instruments was held at EML from October 22-30, 1992. Twenty-five participants submitted 96 passive integrating devices, eight active devices for radon, and seven integrating devices for potential alpha energy concentration (PAEC). In addition, 40 grab samples for radon progeny analysis were taken by five groups that participated in person during the intercomparison. The results reported to EML indicate that the majority of the participants (70%) obtained mean results within 10% of the EML reference value. Although the instruments used in this exercise are based on different principles of collection and detection, they all appear reliable. However, in some instances there seemed to be some minor problems with quality control and calibration bias. Also, the large counting errors for the PAEC experienced by some of the participants can be minimized by using higher sampling air flow rates without sacrificing instrument portability.

INTRODUCTION

The quality of measurements for the assessment of the radiation exposure of the general public from radon and its progeny depends on the proper calibration and maintenance of the instruments and methods that are used to make them. It is recommended that instruments used in the field be evaluated periodically in a laboratory setting in which radon and radon progeny concentrations are well-defined and traceable to a nationally recognized standards laboratory.

Such exercises are part of a continuing effort sponsored by the International Intercomparison and Intercalibration Program (IIIP), sponsored by the International Atomic Energy Agency (IAEA) in cooperation with the Commission of European Communities (CEC). The Environmental Measurements Laboratory (EML), as the designated reference laboratory under IIIP for the intercomparison and intercalibration of the measurement equipment for radon, thoron and their progeny in North America, hosted such an exercise from October 22-30, 1992. A previous exercise dealing with active and passive instruments and methods for radon was conducted in April 1990 (George, 1991). In the present exercise, active instruments for measuring radon progeny were also included to serve those who measure the potential alpha energy concentration (PAEC) with grab sampling, integrating or continuous monitoring instruments and methods. The purpose of this exercise was to evaluate the present state and quality of radon and radon progeny measurements in the indoor environment.

The passive instruments for radon included integrating devices such as activated carbon collectors, electret/ionization chambers, nuclear track detectors, pulse ionization chambers, and continuous passive and active alpha scintillation monitors. The instruments for the measurement of the concentration of individual radon progeny or PAEC consisted of active devices only including those for grab sampling, continuous monitors, and integrating instruments. Radon progeny collected on filter paper were counted by solid state and scintillation counters or by registration of nuclear alpha tracks in solid-state materials. After exposure, most of the devices were returned to the participating laboratories for analysis. Twenty-five participants submitted 44 sets of instruments consisting of 96 passive integrating devices for radon, 8 active continuous devices for radon, 7 integrating devices for PAEC and 40 grab samples for individual measurements for radon progeny and PAEC.

EXPOSURE AND TEST FACILITY

For all tests we used the 20 m^3 radon test chamber, the designated calibration facility for radon, thoron and their progeny (George and Fisenne, 1992). During exposure, conditions were well controlled for temperature, relative humidity and radon concentration. However, because we included active devices for measuring radon progeny, we controlled the generation of wax particles as well to serve as the carrier aerosol. To accommodate the various instruments and exposure protocols, test durations ranged from 1 to 10 days. During exposure all instruments were placed inside the radon/radon progeny test chamber 0.5-1.0 m above the floor. Grab sampling for radon progeny was performed from the adjacent room by inserting sampling filter heads into the test chamber through sampling ports. Each participant provided his or her own sampling apparatus, including: pump and flowmeter, filter paper, filter holder and counting equipment. One of the participants used the protocol of the Rolle method for calculating PAEC (Rolle, 1972), and the remainder used the modified Tsivoglou method (Thomas, 1972) for the

calculation of the concentration of the individual radon progeny and PAEC. EML measured the reference radon progeny test atmosphere on an hourly basis using a precalibrated automated radon progeny sampling and counting system in conjunction with the modified Tsivoglou method. The PAEC was varied by manipulating the concentration of the carrier aerosol onto which radon progeny attach. The carrier aerosol was generated from a wax particle generator (Tu, 1982). Passive instruments were mailed to EML and were returned to the respective participants by mail after exposure. The analysis for most of the measurements made with mailable passive instruments was performed by the participants in their respective laboratories. They reported their results to EML within 2 weeks after the end of the exercise.

QUALITY ASSURANCE

The concentration of radon in the test chamber was measured continuously with two flow-through scintillation cell monitors (George, 1977; Thomas and Countess, 1979). The accuracy of the radon atmosphere in the test chamber was ascertained by intercalibration and intercomparison with radon samples that were analyzed by the EML pulse ionization chambers (Fisenne and Keller, 1985). The calibration of the EML pulse ionization chambers with the National Institute of Standards and Technology (NIST) standard radium sources is the basis of EML's traceability and quality assurance program. The total range of uncertainty (combined systematic and random errors) in the EML reference value is estimated to be approximately ±5% (Fisenne *et al.*, 1990). The concentration of radon progeny in the test chamber was measured by the standard EML grab sampling technique using the Least Squares method (Raabe and Wrenn, 1969). For hourly measurements of the PAEC we used the modified Tsivoglou method (Thomas, 1972). Both methods have been intercalibrated and intercompared on numerous occasions with several reference laboratories and they were found to be within ±5% of each other.

RADON OR RADON PROGENY INSTRUMENTS

The participating laboratories and the characteristics of their instruments and methods are listed in Tables 1 and 2. In Table 1 the characteristics of the passive and active radon instruments and methods are shown. In Table 2 the characteristics of active radon progeny instruments and methods are shown. The passive integrating devices for radon included: activated carbon collectors, electret/ionization chambers, nuclear track detectors, pulse ionization chambers and continuous scintillation cell type monitors. The active instruments for radon consisted of flow-through scintillation type monitors with sensitive cell volumes ranging from 0.1 - 3 L. The active instruments for radon progeny included: grab sampling, integrating and continuous monitoring instruments, which sampled airborne radon progeny in air on filters that were measured by solid-state and scintillation counters or by registration of nuclear tracks in solid-state materials. In grab sampling for PAEC, tests were conducted at two concentration levels using the wax aerosol generator to achieve the desired level. The particle concentration inside the test chamber was measured with a condensation nucleus counter. The accuracies of the PAEC measurements when available were calculated based on counting statistics alone, not taking account of other possible errors from filter paper characteristics, air leakage, sampling flow rate and counter efficiency. Most of the participants using passive instruments did not supply measurement statistics, depriving us of an additional tool to evaluate the intercomparison

results. In the future, we will stress to the participants the importance of the statistical information.

RESULTS AND DISCUSSION

The measurement results reported by the participating groups are listed in Tables 3 and 4, along with the results obtained by EML during the same exposure period. One participant did not submit measurement results because he discovered air leakage in his sampling system after the exercise was over. The EML values are used as the reference values against which all other measurements are compared. To maintain participant-result confidentiality, the reported values in the tables are listed randomly. For comparison purposes and easier evaluation, the results for radon measurements listed in Table 3 and Figure 1 were grouped into five device categories consisting of passive activated carbon, nuclear track, electret/ ionization chamber and passive and active continuous scintillation cell monitors. The range, the mean and standard deviation of the individual group data are compared with the reference mean value obtained by EML during the same time period. In most cases, no error uncertainties were reported for the individual measurements and therefore the error of the ratios in the last column of Table 3 could not be estimated. Table 3 and Figure 1 illustrate the precision and accuracy of individual participant measurement results.

The ratios from the mean values for the passive activated carbon collectors, nuclear alpha track detectors, electret ionization chambers and continuous passive and active commercial electronic instruments are 0.95, 0.90, 0.95, 0.96, and 1.12, respectively. Considering the overall uncertainty in the reference value of ±5%, all of the participants as groups did well on the average. However, the results from three participants using activated carbon collectors exhibited large deviations from the mean values. In the previous intercomparison (George, 1991), the activated carbon collector data showed very good precision.

Nuclear alpha track detectors, except for one case, performed reasonably well, indicating a substantial improvement since the last intercomparison (George, 1991). The mean ratios of the participant/reference ranged from 0.72-1.02, as compared to 0.69-1.80 reported in the previous exercise. However, it appears that some participants may still have problems with calibration bias and product quality control. One of the participants obtained high precision and accuracy indicating that there is no generic problem with nuclear track detector materials when good quality control is exercised. This participant claims that the proper design of the alpha track detector and processing of the nuclear film are the major factors in producing high precision data. The monitoring device cannot have areas of persistent charge as this produces artifacts that interfere with the accumulation of alpha tracks. The processing of the film must be conducted under strictly controlled conditions or calibration fluctuations will occur. Since the number of participants in this intercomparison was relatively high (11 participants), it appears that nuclear alpha track detectors are gaining wide acceptance for long-term radon exposure determination.

Electret ionization chambers, used by a total of four participants, showed good precision and accuracy. The detectors of one participant exhibited a large dispersion among the four measurements, with a standard deviation of 6.5%. The remainder of the detectors exhibited standard deviations that were about three times smaller, indicating good precision.

Most of the continuous passive electronic radon monitors gave slightly lower mean readings than the reference values, but they were still well within the range of possible biases experienced with commercial instruments. Only one instrument showed a possible calibration bias (15% low). Two participants with active electronic continuous radon monitors reported slightly high mean readings (12% high).

The measurement results for the individual radon progeny concentrations and PAEC are listed randomly in Table 4, and are illustrated in Figures 2 and 3. Listed in Table 4 are the results of the individual radon progeny, and the PAEC obtained with grab sampling measurements, followed by the integrating and continuous monitoring measurements of the PAEC. The results from three participating laboratories who sampled and analyzed for individual radon progeny at high particle concentration are also given in Table 4, and they are shown graphically in Figure 2. As mentioned earlier, one participant did not submit measurement results due to instrument malfunction. The last column in Table 4 gives the concentration of the reference radon progeny atmosphere. The reported ratios show very good agreement among the mean values of all three radon progeny. However, during sampling at low particle concentration, one participant did poorly by overestimating the concentration of ^{214}Pb. What is unusual about this discrepancy is that it is in the wrong direction from what would be expected if plateout was occurring. The overestimation of the PAEC in two cases (6% and 9%) occurred during low particle concentration, indicating that there is no effect from radon progeny plateout. The results of the grab sampling intercomparisons indicate that the participants used generally good techniques and properly calibrated equipment to measure the PAEC.

The results of integrating and continuous monitoring instruments for PAEC are summarized in Table 4, and are illustrated in Figure 3. The ratios of the participant's PAEC to that measured by EML ranged from 0.58 to 1.25, with 50% of the participants reporting results within 10% of the actual reference value. The lowest ratio reported is indicative of possible problems with flow rate, total volume of air sampled and counting efficiency. Most of the participants did not report measurement uncertainties.

CONCLUSIONS

The intercomparison results for passive and active radon instruments and active PAEC instruments used mainly in North America are generally satisfactory. More than 70% of the instruments used for measuring radon either actively or passively were within 10% of the reference value. Although the instruments are based on different principles of collection and detection, they appear to be reliable. However, in some instances there may be some minor problems with quality control and calibration bias.

Good agreement was found between the participants' grab sampling instruments and techniques and the EML reference values. The instruments used for radon progeny measurements appear to be properly calibrated for the intended use. Large counting errors experienced by some participants can be minimized by using larger sampling flow rates without sacrificing

instrument portability. The quality of the measurements made with integrating and continuous monitoring commercial instruments during this intercomparison exercise appears to be satisfactory. Plateout of radon progeny in the head of the sampling detector of some of these instruments does not seem to be a problem. However, in some cases deficiencies of inaccurate flow rate and inappropriate counting efficiency may still persist. In general, commercial instruments can be used with confidence for the assessment of the airborne PAEC once they are calibrated and routinely maintained and properly operated.

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TABLE 1

PASSIVE AND ACTIVE RADON INSTRUMENTS
USED BY PARTICIPATING GROUPS

Participant	Instrument/Method	Exposure Duration (days)
AECL-Low Radioact. Ottawa, Canada	PI, Electret/Ioniz. Chamber PC, Pylon AB-5	4 2
Altrac Berlin, Germany	PI, Nuclear Track Type A PI, Nuclear Track Type C/D PI, Nuclear Track Type B	3 8 8
Bowser/Morner Dayton, OH	PC, Pulse Ioniz. Chamber R-210F PI, Carbon Collector (OF)	2 2
Enviroserv, Inc. Morristown, NJ	PI, Carbon Collector (OF)	2
femto-TECH, Inc. Carlisle, OH	PC, Pulse Ioniz. Chamber (Model CRM-510)	2
Health/Welfare Ottawa, Canada	PI, Nuclear Track	8
Landauer, Inc. Glenwood, IL	PI, Nuclear Track	8
LBL Berkeley, CA	AC, Scintillation Cell (LBL Type)	2
National Inst. Radiat. Sciences Chiba, Japan	PI, Nuclear Track	7
NJ Dept. Environ. Protection Trenton, NJ	PI, Carbon Collector (OF)	3
New York Univ. Med. Center, New York, NY	PI, Nuclear Track PI, Nuclear Track	2 3
ORNL Oak Ridge, TN	PC, Pulse Ioniz. Chamber, R-210F	2

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

TABLE 1 (Cont'd)

Participant	Instrument/Method	Exposure Duration (days)
Penn. Dept. Env. Resources Harrisburg, PA	AC, Scintillation Cell RGM-3	2
	PI, Carbon Collector (DB)	6
	PI, Electret/Ioniz. Chamber	4
Rad Elec, Inc. Frederick, MD	PI, Electret/Ioniz. Chamber	4
	Short-Term Exposure, Long-Term Exposure	8
Radon Instrument Calibr. Services Bethlehem, PA	PC, Pylon AB-5	4
Radon QC Northbrook, IL	PI, Nuclear Track	7
RSSI Morton Grove, IL	PI, Nuclear Track	8
RTCA Irvington, NY	PI, Electret/Ioniz. Chamber	4
	PI, Carbon Collector (OF/DB)	4
St. Johns Univ. Collegeville, MN	PI, Nuclear Track	8
Teledyne Isotopes Westwood, NJ	PI, Carbon Collector (OF)	4
Univ./Pittsburgh Pittsburgh, PA	PI, Carbon Collector (DB)	7
USDOE-EML New York, NY	AC, Scintillation Cell (EML Type)	10
USEPA (NAREL) Montgomery, AL	PI, Carbon Collector (DB)	7
Wilkes Univ. Wilkes Barre, PA	PI, Carbon Collector (DB)	4
	PC, Pylon AB-5	1

TABLE 2

ACTIVE INSTRUMENTS AND METHODS USED BY PARTICIPANTS TO MEASURE THE PAEC

Participant	Method - Detection System	Counter Efficiency (%)	Flow Rate (L min^{-1})
Alpha Nuclear Mississauga, CAN	Cont. Alpha Silicon Detector	17.0	0.13
AECL Ottawa, Canada	Grab, Alpha Scintillation	48.0	4.08
Canadian Inst. Radiation Safety Toronto, Canada	Integrating Nuclear Track	-	-
COMRO Johannesburg, SA	Cont. Alpha Spectrometry	30.0	1.9
Enviroserv Morristown, NJ	Cont. Alpha Silicon Detector	17.0	0.13
Pylon Electronics Ottawa, Canada	Grab, Alpha Scintillation Cont. Alpha Scintillation Semi-Cont. Scintillation	48.0 13.5 13.5	8.6 0.5 4.0
Radon Instrument Calibration Serv. Bethlehem, PA	Cont. Alpha Solid State	15.0	1.0
Thomson/Nielsen Ottawa, Canada	Grab, Alpha Solid State	-	4.0
USEPA-NAREL Montgomery, AL	Grab, Alpha Scintillation	45.4	13.2
USDOE-EML New York, NY	Grab, Alpha Scintillation Cont. Alpha Solid State	49.3 19.0	12.4 20.0
Wilkes Univ. Wilkes Barre, PA	Integr. Alpha Scintillation Cont. Alpha Solid State	16.0 -	3.5 -

TABLE 3

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

Type of Instrument	Participant		Reference Mean±Error*	Participant/ Reference
	Range	Mean±σ		
Passive	1624-1813	1695± 84	1647±30	1.03
	1526-1660	1597± 65	1591±28	1.00
	1757-1872	1807± 50	1610±30	1.12
Activated Carbon	1262-1440	1332± 76	1672±30	0.80
	1691-1809	1746± 49	1684±30	1.04
	1539-1647	1614± 81	1691±30	0.95
	1047-1706	1295±294	1646±30	0.79
	1376-1650	1484±124	1698±30	0.87
	1536-1602	1572± 32	1647±30	0.95
Nuclear Track		1415	1680±30	0.84
		1526	1680±30	0.91
		1580	1576±28	1.00
	1308-1703	1586±111	1680±30	0.94
	1221-1480	1378±114	1680±30	0.82
	1425-1628	1506± 91	1680±30	0.90
	1590-1610	1600± 14	1576±28	1.02
	1630-1631	1630± 0	1684±30	0.97
	1259-1992	1495±341	1680±30	0.89
	1413-1657	1576± 94	1680±30	0.94
	1106-1234	1167± 57	1632±29	0.72
Electret/ Ionization Chamber	1480-1676	1587±102	1645±29	0.96
	1543-1613	1580± 35	1684±30	0.94
	1565-1654	1617± 37	1680±30	0.96
	1506-1554	1532± 25	1684±30	0.91
	1536-1602	1576± 48	1645±29	0.96
Continuous		1602	1627±29	0.99
		1654	1591±28	1.04
		1576	1591±28	0.99
		1410	1650±29	0.85
		1584	1680±30	0.94
		1421	1462±25	0.97
Active Continuous	1758-1772	1765± 10	1610±29	1.10
		2012	1769±32	1.14

*Counting Error

TABLE 4

SUMMARY OF RADON PROGENY INTERCOMPARISON MEASUREMENTS

Random Facility Number/ Sampling Method	Particle Concentr.	Ratio Facility/Reference ^{218}Po	Ratio Facility/Reference ^{214}Pb	Ratio Facility/Reference ^{214}Bi	PAEC	Reference PAEC (nJ m^3)
1-Grab	High*	0.65	1.09	1.34	0.96	
	"	0.92	0.93	1.11	0.97	
	"	1.27	1.05	0.69	1.03	
	"	1.10	1.14	0.79	1.05	
	Mean±σ	(0.99)±0.26	(1.05)±0.09	(0.98)±0.30	(1.00)±0.05	1861±23
1-Grab	Low**	0.81	1.08	1.03	0.95	
		1.02	1.07	0.76	0.99	
		1.15	1.05	0.62	1.02	
		0.94	0.94	1.07	0.97	
	Mean±σ	(0.98)±0.14	(1.03)±0.06	(0.87)±0.22	(0.98)±0.03	639±18
2-Grab	High*	1.22	1.07	0.86	0.99	
		0.93	0.96	1.12	0.99	
		0.84	0.99	1.07	0.97	
		0.91	0.99	1.07	0.99	
	Mean±σ	(0.98)±0.17	(1.00)±0.05	(1.03)±0.12	(0.99)±0.01	1861±23
2-Grab	Low**	0.94	1.08	1.00	1.01	
		0.80	0.97	1.31	0.96	
		0.96	1.05	0.98	1.01	
		0.98	1.03	0.96	0.99	
	Mean±σ	(0.92)±0.08	(1.03)±0.05	(1.06)±0.17	(0.99)±0.02	639±18
3-Grab	High*	0.98	0.87	1.47	0.94	
		0.90	0.88	1.18	0.95	
		1.04	1.04	0.92	1.01	
		1.17	1.16	0.83	1.08	
	Mean±σ	(1.02)±0.11	(0.99)±0.14	(1.10)±0.28	(0.99)±0.06	1861±18

TABLE 4 (Cont'd)

Random Facility Number/ Sampling Method	Particle Concentr.	Ratio Facility/Reference ^{218}Po	Ratio Facility/Reference ^{214}Pb	Ratio Facility/Reference ^{214}Bi	PAEC	Reference PAEC (nJ m^3)
3-Grab	Low**	0.75	1.30	1.20	1.05	
		1.12	1.74	0.53	1.25	
		0.92	0.91	1.39	0.98	
	Mean±σ	(0.93)±0.19	(1.32)±0.42	(1.04)±0.45	(1.09)±0.14	545±18
4-Grab	High*					1.06
						1.03
						1.06
	Mean±σ				(1.05)±0.02	2362±22
4-Grab	Low**				1.20	
					0.91	
					1.22	
					0.93	
	Mean±σ				(1.06)±0.17	639±18
5-(No data)						
6-Integr.					1.25	2254±25
7-Integr.					1.07	1281±15
8-Integr.					0.58	1165±14
9-Integr.					1.07	1910±21
10-Integr.					0.95	1745±20
11-Cont.					0.96	1560±18
12-Cont.					0.98	1581±18
13-Cont.					1.12	1997±22

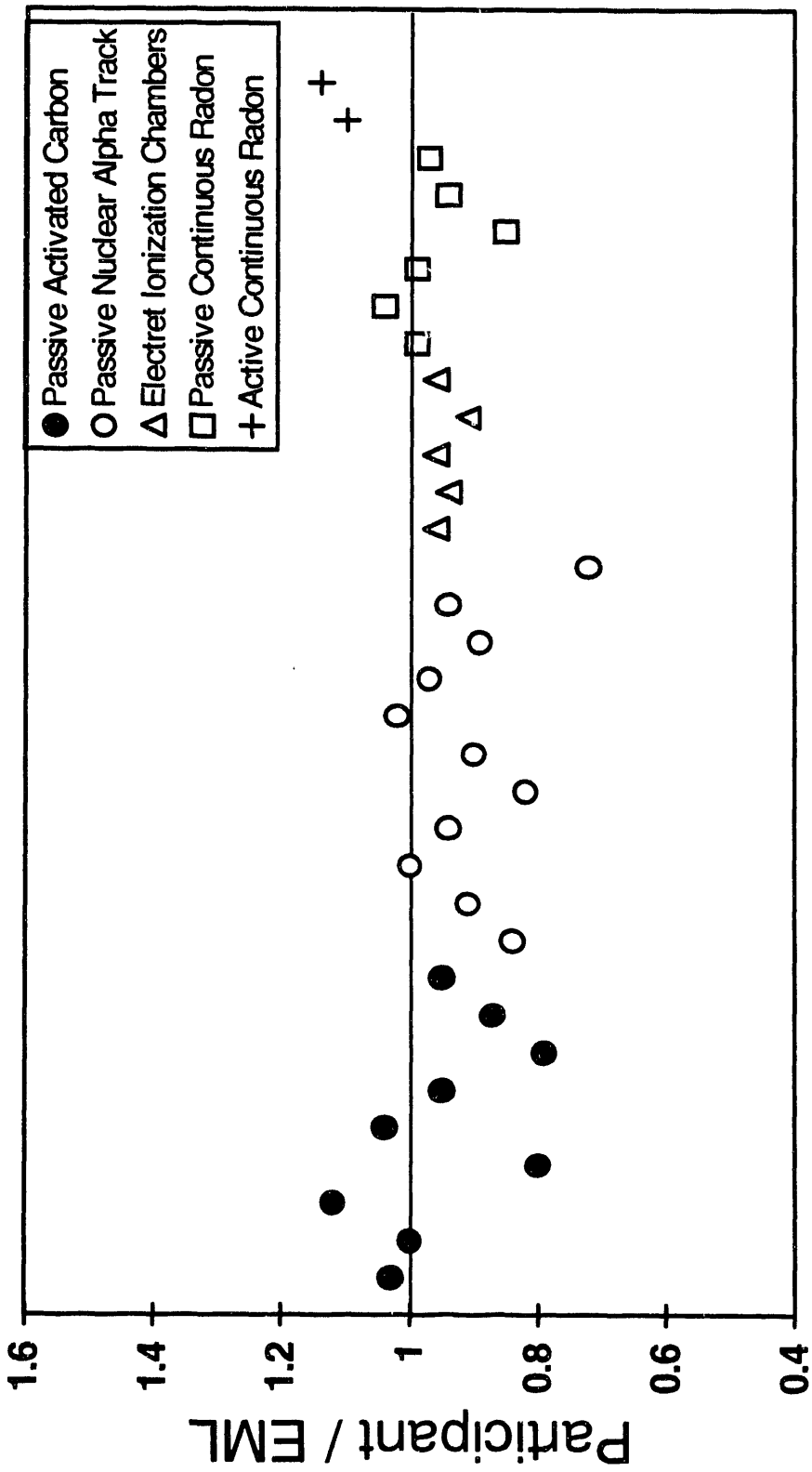

Figure 1. Intercomparison of passive and active integrating radon instruments.

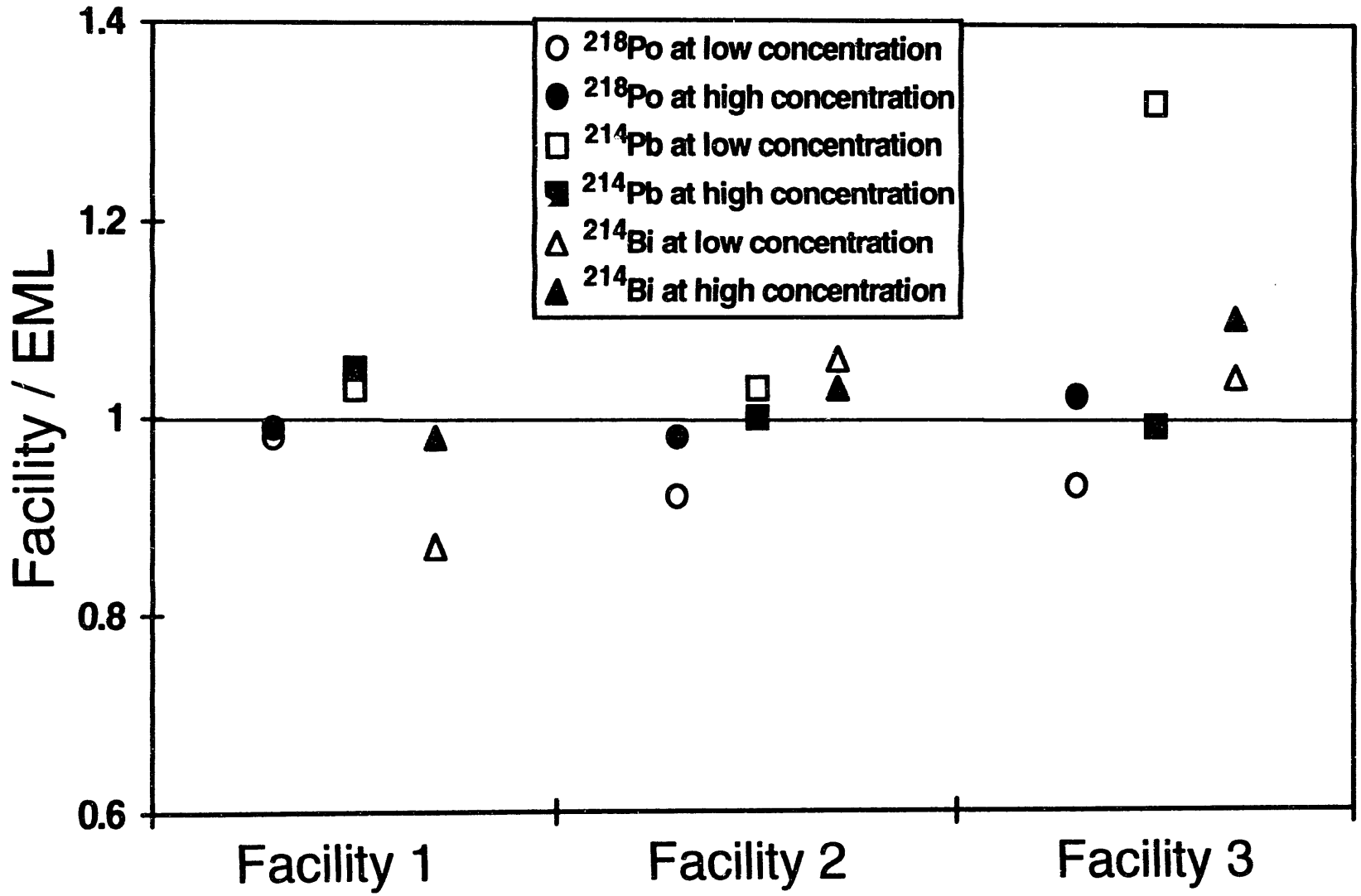

Figure 2. Intercomparison of individual radon progeny at low and high particle concentrations.

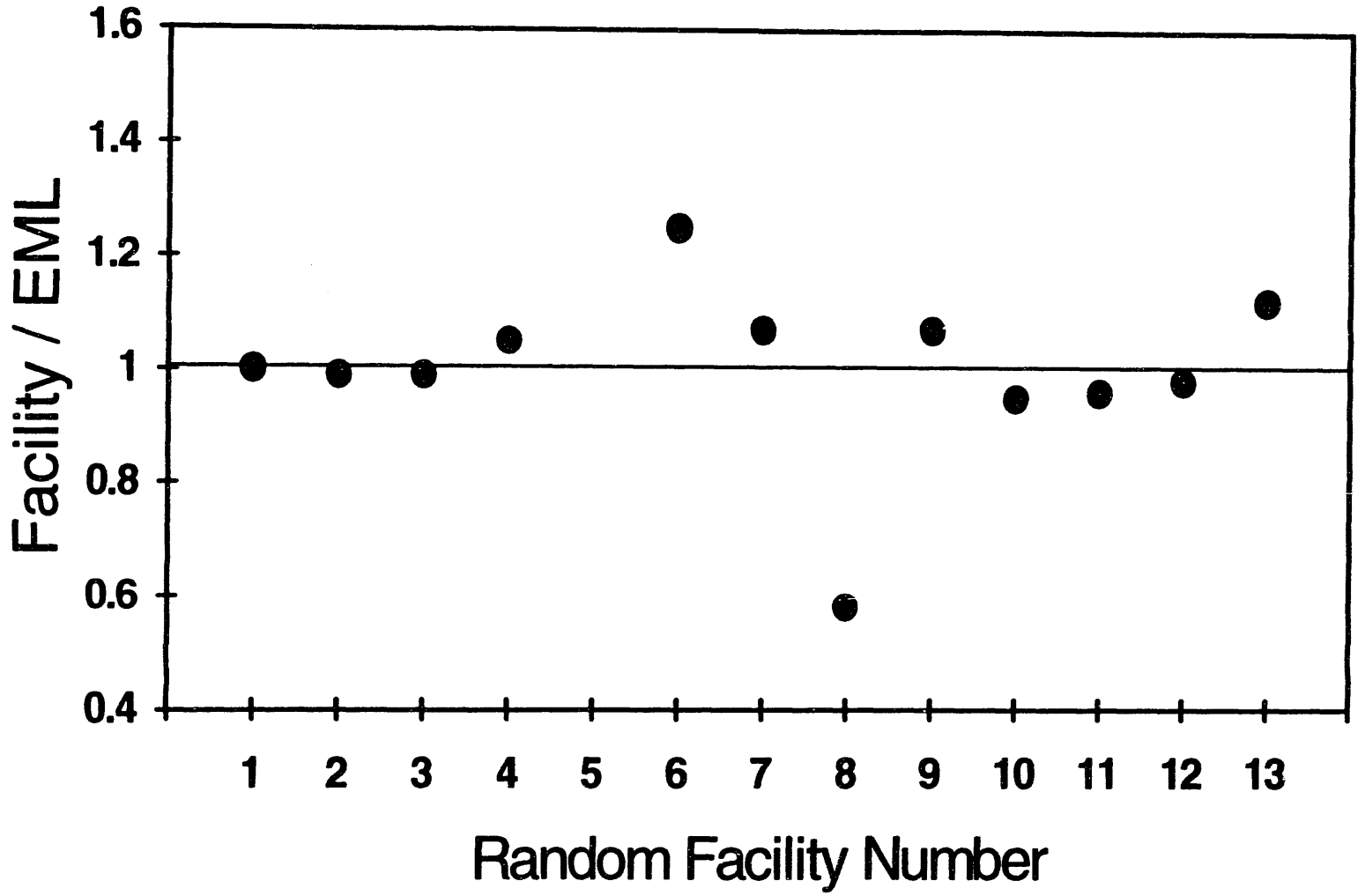

Figure 3. Intercomparison of grab, continuous, and integrating instruments for PAEC.

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