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Australian Radiation Laboratory

OECD/NEA and CEC
Intercalibration and Intercomparison Programme for Radon

Radon Daughter Aerosol Workshop
ARL March 17-21 1986

by

Stephen B. Solomon, Earl O. Knutson[1]
and Robert F. Holub[2]

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Stephen B. Solomon, Earl O. Knutson(1)

and Robert F. Holub(2)

(1) Present Address: Environmental Measurements Laboratory
376 Hudson St, New York, N Y, 10014, USA

(2) Present Address: Geophysics Div, U S Bureau Of Mines,
Building 20, Denver Federal Centre, Denver, 80225, Co, USA

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ABSTRACT

The International Intercalibration and Intercomparison Programme for Radon, Thoron and Daughters Monitoring Equipment, abbreviated IIIP, is a programme initiated jointly by the Nuclear Energy Agency of the Organization for Economic Cooperation and Development and the Radiation Protection Research Programme of the Commission of European Communities. The broad purpose of the IIIP is to determine if the diverse calibration procedures for radon and radon daughters used in different countries are equivalent. The general approach was to select four regional reference laboratories and entrust them with the details of project definition and execution, and with the coordination of the regional intercomparison activities. The four selected laboratories are: Australian Radiation Laboratory (ARL), US Department of Energy, Environmental Measurements Laboratory (EML), US Department of Industry, Bureau of Mines (USBM), UK National Radiological Protection Board (NRPB). As one phase of this IIIP programme, an intercomparison of measurements of radon daughter activity concentration, "unattached" fraction and aerosol size distribution of radon daughters, was carried out by three of the reference laboratories (ARL, EML and USBM), over the period March 17 to 21, 1986, using the radon test chamber at the Australian Radiation Laboratory, Melbourne. The methodology, protocols and results for these measurements are described in detail in this report. The results of the intercomparisons by ARL, EML and USBM for the measurement of radon daughter activity concentrations, showed no systematic differences between the three laboratories, within the limits set by the counting statistics. In intercomparing methods for "unattached" fraction measurement, two of the reference laboratories (ARL and EML) used the single screen method for "unattached" fraction measurement. Good agreement was obtained between ARL and EML for the results of the intercomparison of the "unattached" fraction of potential alpha energy concentration (PAEC), and of "unattached" fraction of the individual radon daughters. Two of the reference laboratories (EML and USBM) also used graded screen devices for sizing these so-called "unattached" radon daughters. At the time of these measurements, the graded screen analysis methods available were of a preliminary nature. The ultrafine aerosol size distributions derived by EML and USBM show reasonable agreement in the position and magnitude of the derived peaks. The results of the measurements of the radon daughter aerosol size distributions between ARL, EML and USBM showed good agreement within the regions of overlap of the instrument response.

1.1 BACKGROUND

The recognition that radon and its short lived daughter products contribute more than one-half of the average annual radiation dose to an individual from natural radiation sources (UNSCEAR, 1988), has prompted an increased interest in radon in the environment. Many countries are engaged in, or have completed, large-scale surveys of ambient radon levels (UNSCEAR, 1988). These surveys have used a diverse range of measurement methods. The value of interlaboratory, inter-agency or international projects to intercompare measurement methods, has been recognized by a number of laboratories and organizations.

The International Intercomparison and Intercalibration Programme for radon, thoron and daughter products (abbreviated as IIIP), was initiated in November,1983 by the Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD) and the Commission of European Communities (CEC). Four laboratories were designated as regional radon reference facilities (Appendix 1). The IIIP was divided into Part I (radon) and Part II (radon daughters). Both Part I and Part II of the IIIP have been divided into two phases. The first phase involves measurements between the four reference laboratories, each representing a particular geographical region. The second phase involves measurements by the participating laboratories in each of the OECD-member countries of the defined geographical region, using the radon test facility of the corresponding reference laboratory. Part I of this programme, covering intercalibration and intercomparison of radon measurement equipment was successfully completed in 1986 (Knutson, 1986).

For the radon intercomparison programme (Phase 1 and Phase 2 of Part I), samples of radon gas were transferred from the reference test chambers to participating laboratories (Knutson, 1986; Solomon, 1983). For shipping times of the order of days, reductions in the activity of ^{222}Rn due to radioactive decay can be accurately adjusted for. The intercomparison of measurements of the shorter-lived radon daughters cannot be made in the same manner, and it is necessary to carry out side-by-side measurements of the same test atmosphere.

The first phase of the Part II programme for the intercomparison of radon daughter measurement equipment was commenced in 1983. The four laboratories designated as regional reference facilities embarked upon a programme to define, generate and measure controlled test atmospheres of radon daughters. A series of intercomparisons of radon daughter activity measurements was carried out by these three regional reference laboratories using the radon chambers at the National

Radiological Protection Board (NRPB) during October 1983 and at the Environmental Measurement Laboratory (EML) during May 1985 (Knutson, 1986).

The test atmospheres for the radon daughter measurements to be used in Part II of the IIIP were defined at a OECD/NEA and CEC International Radon Workshop, held in Paris during May 1985. The experimental conditions were defined in terms of the radon concentration, equilibrium ratio (F-factor), aerosol concentration and aerosol size. A representative activity median aerodynamic diameter of 100 nm was chosen. The purpose of the radon daughter aerosol workshop at ARL was to allow the reference laboratories to intercompare their measurements of radon daughter activity size distribution, and so ensure uniform conditions for the second phase of the radon daughter intercomparison programme.

During March 1986, three[1] of the four regional radon reference laboratories - the Australian Radiation Laboratory (ARL), the Environmental Measurement Laboratory and the U.S. Bureau of Mines (USBM), carried out measurements of radon daughter activity concentration, potential alpha energy concentration (PAEC), "unattached" fraction and activity size distributions for the radon daughters, using the ARL radon test chamber in Melbourne. This report describes the methodology and results of these measurements.

1.3 MEASUREMENT PROTOCOLS

In previous radon daughter measurement intercomparisons for Part II, Phase 1 of the IIIP (Knutson, 1986), the four regional reference laboratories carried out measurements of activity standards, flow standards and radon daughter activities using both filters and wire screens. The programme of measurements at ARL was divided into four sections;

(a) An intercomparison of standardised alpha particle sources. Although an intercomparison of alpha particle sources had been made previously, some problems were experienced with some of the sources in allowing for backscattered alpha particles and for system dead-times (Knutson, 1986). EML and USBM each provided an ^{241}Am alpha particle source. Three new ^{238}Pu alpha particle sources were provided by ARL. These sources had activities in the range 330 to 420 Bq, and were deposited on Kapton backings.

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[1] The NRPB was unable to participate in the Melbourne workshop. A set of radon daughter aerosol measurements were made by EML and NRPB using the EML radon facility in New York, USA during April 1986.

(b) An intercomparison of radon daughter activity concentration measurement.
(c) An intercomparison of measurements of the fraction of the so-called "unattached" or free radon daughters.
(d) An intercomparison of measurements of the size distribution of the radon daughter aerosol.

The last three sets of measurement were made using the radon test chambers at ARL, described in Appendix 2. A wide range of exposure conditions was used and these are summarized in Table 1. The radon and aerosol concentration in the chamber during the radon daughter measurements are shown in Figure 1. The commencement time for each test is also marked on this figure. The radon concentration in the chamber was continuously monitored using a flow-through scintillation cell. Grab samples were taken from the chamber at regular intervals using evacuated scintillation flasks (Lucas cells). A carnauba wax aerosol with a nominal diameter of 100 nm was used as the test aerosol (Tu, 1982). The size distribution of this aerosol was determined at the time of the test exposures using an electrical aerosol size analyzer (TSI Model 3030, abreviated as TSI EAA in Tables and Figures). The number concentration was measured using a condensation nuclei (CN) counter (Environment One Rich 200).

Over a period of three days, eleven sets of radon daughter measurements were carried out using test samples collected from the ARL chamber. Five sets of samples were collected by each laboratory using open-face filter holders inserted into the test chamber. Each laboratory then analyzed its respective sample to determine the individual radon daughter activity concentrations. The equipment used by each laboratory for these measurements is summarized in Tables 2 to 4. The equipment used by each laboratory for the two sets of measurements of "unattached" fraction is summarized in Table 5.

The equipment and analysis methods used by each laboratory for the four sets of determination of the radon daughter size distribution are summarized in Tables 6-8. The size ranges covered by the three sets of equipment were not the same. The EML device was designed to size both attached and "unattached" radon daughters and covers a size range of 2 to 1400 nanometers. The ARL and USBM devices were optimised for attached and "unattached" radon daughters respectively. The three laboratories carried four sets of concurrent determination of size distribution from the test chamber, two at high CN concentration, and two at low CN concentration.

1.4 RESULTS

1.4.1 Intercomparison of Efficiency of Activity Standards

All three laboratories used windowless detectors comprising photomultiplier tubes and zinc sulphide coated screens for the alpha counting. ARL used two identical detectors for the intercomparison. The detection efficiency of each laboratory's counting system and for each calibrated alpha reference source, as determined from the ratio of the measured count rate to the quoted source activity, is shown in Table 9. Dead-time corrections were negligible for all systems and for all five alpha sources used.

1.4.2 Intercomparison of Radon Daughter Activity Concentrations

The results for the three laboratories of the five concurrent measurements of radon daughter activity concentration and potential alpha energy concentration (PAEC) are shown in Table 10. The quoted uncertainties are one standard deviation derived from the counting statistics. Two of the laboratories (ARL and USBM) used the modified-Tsivoglou method (Thomas, 1972) for the analysis of the filter activity, while the third (EML) used a form of the Raabe-Wrenn least-squares analysis (Raabe and Wrenn, 1965). The results were derived from concurrent samples of 10 minutes duration.

1.4.3 Intercomparison of "Unattached" Fraction Measurements

Two of the laboratories used multi-screen devices (EML and USBM), designed to measure the particle size distributions of the "unattached" or free radon daughters. The ARL used a single wire screen device, designed to measure the fraction of radon daughters not attached to the ambient aerosol; the so-called "unattached" fraction. For the purposes of this intercomparison, the EML data (screen #1 only) were interpreted by the single screen technique (George, 1972). Two sets of measurements of these "unattached" fractions were made. The results obtained by ARL and EML for these two measurements are shown in Table 11.

1.4.4 Intercomparison of Activity Size Distributions Measurements

Two of the laboratories, ARL and EML, carried out measurements of activity size distributions, using a serial wire screen battery and a parallel tubular diffusion battery, respectively. The activities on the aerosols penetrating through each stage of the battery were analyzed for radon daughter concentrations and PAEC and the penetration was then stripped into particle size ranges. Measurements of the size distributions of particles less than 50 nm in diameter were made by EML and USBM using graded screen devices (Holub et al., 1987). An analysis technique for this multiple screen data, for both EML and USBM, is

currently being developed. For the four sets of particle size measurements, a preliminary analysis of the size distributions of particles less than 50 nm in diameter was carried out. The resultant size distributions for the four graded screen tests were then combined with the diffusion battery results, to produce an extended size distribution covering both "unattached" and attached radon daughters. These are shown in Figures 2. to 11. Although the results for the three laboratories used different size ranges and size widths, for these figures the fraction of activity, divided by the logarithm of the size width, has been plotted against the logarithm of the particle diameter, so that the particle size distributions for each test can be directly compared. The derived values for each test of the activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD) of radon daughter PAEC are shown in Table 12. Where the distribution was obviously bimodal, the AMAD and GSD for each peak were determined. The results are compared with median diameter and geometric standard deviation derived from the surface area distribution measured by the TSI electrical aerosol analyzer. In Table 13, the calculated values of the AMAD and GSD for each of the ^{218}Po (RaA) and ^{214}Pb (RaB) activity size distributions are given. As for the PAEC distribution, bimodal distributions have been analyzed for two sets of AMAD and GSD values.

1.5 DISCUSSION

The five activity standards, (three ARL sources, one each from EML and USBM) were intercompared using four counters (two ARL counters, one each EML and USBM). The data in Table 9 shows that, for the EML and USBM counters, the standard deviation of the mean efficiency for each detector, derived from all standards, was less than 0.5% and 0.8%, respectively. The corresponding results for the ARL counters show a small systematic difference of 3.4% between the standards with the thin Kapton backings and those with the thicker metal backings. The change to the geometric efficiency, due to an increase in backing material thickness of 0.6 mm, is sufficient to account for this difference in the ARL counter efficiencies. Both the EML and USBM counters were insensitive to small changes in the source to detector distance.

Of the five radon daughter measurements in Table 10, the USBM results in tests #1 and #2 were surprising. The USBM sampler was positioned away from the ARL and EML sample heads during these measurements. At the completion of test #2, the amount of air turbulence in the chamber was increased using an electric fan. The factor of two difference in the first two USBM results disappeared in the later tests #3, #5, and #7. It is possible that a lack of turbulence in the ARL

radon chamber may have produced spatial non-uniformity in the radon daughter concentration between the USBM and ARL/EML sampling positions. For the three later tests, the results show no significant systematic differences between the three laboratories, for radon daughter concentration and PAEC, within the limits set by the counting statistics. These results are consistent with the results of the previous radon daughter intercomparison, between ARL, EML, NRPB and USBM, at EML during May 1985 (Knutson, 1986).

For the attached aerosols, the derived PAEC particle size distributions (Figures 2 to 5), show good qualitative agreement between the ARL and EML results for particle sizes between 20 and 500 nm. There was good agreement between ARL, EML and the TSI electrostatic analyzer measurements in the derived values for geometric mean and standard deviation for the attached PAEC (Table 12). However, an analysis of the data to account for counting statistics effects on the deconvolution calculation, indicated very large uncertainties in the derived values for the geometric standard deviation.

For the individual ^{218}Po and ^{214}Pb particle size distributions over this size range, the qualitative agreement is still good. The derived values for geometric mean and standard deviation for each of these two radionuclides are shown in Table 13. The larger statistical uncertainties associated with the concentration measurements for these daughters leads to a wider variation in the derived values for the geometric means and standard deviations. Test #10 was not analyzed for either ^{218}Po or ^{214}Pb particle size distributions because the statistical uncertainties were too large to allow a sensible solution to the deconvolution calculation.

The results of the EML and ARL measurements of the "unattached" fraction of individual radon daughters and of PAEC, shown in Table 11, show that there is good agreement between the two laboratories. However, there was poor agreement between the three laboratories in the measurement of the size distribution of the "unattached" radon daughters (0.6 to 20 nm). The ARL diffusion battery was located outside the chamber and some losses of the nanometer sized particles would be expected in the connecting tube. Both the EML and USBM batteries were still under development for the sizing of nanometer sized particles at the time of these tests, and a full theoretical development of the analysis method is still in progress. The preliminary analysis of the results for EML and USBM multi-screen measurements of these "unattached" daughters does show promise.

REFERENCES

George,A.G.,Hinchcliffe,L. and Sladowski,R., 1975, "Size Distribution of Radon Daughter Particles in Uranium Mine Atmospheres", J American Industrial Hygiene Association, 36:484-490.

Holub, R.F. and Knutson, E.O., 1987, "Measuring Po-218 Diffusion Coefficient Spectra using Multiple Wire Screens", In: **Radon and its Decay Products**, Ed. Hopke, P.K., Am. Chem. Soc., Washington D.C., pp 311-356.

Holub, R.F., Knutson, E.O. and Solomon, S.B., 1987, "Radioactive Aerosol Measurement and Intercomparison as Part of the OECD/NEA Programme", Paper presented at the 4th. Int. Symp. on the Natural Radiation Environment, Lisbon, Portugal, December 1987.

Knutson, E.O. and Sinclair, D., 1978, "Experience in Sampling Urban Aerosols with the Sinclair Diffusion Battery and Nucleus Counter." In: Proceedings: Advances in Particle Sampling and Measurement, Ashville, N.C., U.S.A., EPA-600/7-79-065.

Knutson,E.O.,(Ed.),1986, "International Intercalibration and Intercomparison of Radon, Thoron and Daughter Measuring Equipment - Report on Part I, Radon Measurement." Nuclear Energy Agency of the Organization for Economic Cooperation and Development and the Commission of the European Communities, Paris.

Knutson, E.O., 1987, "The OECD/CEC Radon Measurement Intercomparison: A Further Analysis of the Results", Paper presented at the 4th Int. Symp. on the Natural Radiation Environment, Lisbon, Portugal, December 1987.

Nazaroff,W.W., 1984, "Optimising the Total-Alpha Three-Count Technique for Measuring Concentrations of Radon Progeny in Residences", Health Physics 46:395-405.

Nazaroff, W.W., Nero,A.V. and Revzan,K.L., 1982, "Alpha Spectroscopic Techniques for Field Measurement of Radon Daughters.", In: Natural Radiation Environment,Ed. by K.G. Vohra et al., Proc. of the 2nd Special Symposium on the Natural Radiation Environment, Bhabba Atomic Research Centre, Bombay, January 19-23, 1981; Wiley Eastern, New Delhi, pp 350-357.

Polito,M.D. and Negro, V.C., 1979, "SMART-1 and Clever Counter,

Environmental Data Acquisition Systems", IEEE Transactions on Nuclear Science, NS-26:765-769.

Raabe,O.G. and Wrenn,M.E., 1965, "Analysis of the Activity of Radon Daughter Samples by Weighted Least Squares", Health Physics 17:593-605.

Sinclair,D., 1972, "A Portable Diffusion Battery", J American Industrial Hygiene Association, 33:729-735.

Strong, J.C. and Tu, K.W., 1987, "Design and Calibration of a Multichannel Diffusion Battery for the Measurement of Radon Daughter Activity-size Distributions.", In: Proc. of the 1st Conference of the Aerosol Society, Aerosols: Their Generation, Behaviour and Applications, Loughborough, March 1987.

Thomas, J.W., 1972, "Measurements of Radon daughters in Air", Health Physics 23:783-790.

Twomey,S.,1975, "Comparison of Constrained Linear Inversion and an Iterative Non-linear Algorithm Applied to Indirect Estimation of Particle Size Distributions", J. Comput. Sci., 18:188.

Tu, Keng-Wu, 1982, "A Condensation Aerosol Generator System for Monodispersed Aerosols of Different Physiochemical Properties", J. Aerosol Sci. 13:363-371.

UNSCEAR, 1988, "Ionizing Radiation: Sources and Biological Effects"; United Nations Committee on the Effects of Atomic Radiation. 1988 Report to the General Assembly with Annexes. United Nations, New York.

Table 1

ARL Radon Chamber Exposure Conditions

Test	Date	Start Time	Type (1)	Temp C	%RH	[Aerosol] (2)	GMD(3) nm	GSD	[Rn] $Bq.m^{-3}$
#1	18 March	15:58	Filter	20.0	51.3	320000	121	1.81	1038
#2	19 March	08:55	Filter	21.0	50.5	1000	---	----	870
#3	19 March	11:25	Filter	20.2	50.6	40000	100	1.95	5325
#4	19 March	12:35	Battery	20.1	50.8	63000	103	1.78	5127
#5	19 March	14:47	Filter	20.7	51.4	78000	---	----	4772
#6	19 March	16:00	Battery	21.0	51.4	60000	---	----	4500
#7	20 March	9:45	Filter	20.0	50.9	5170	125	1.92	3280
#8	20 March	11:25	Battery	21.0	51.2	7360	129	2.18	3330
#9	20 March	12:45	Screen	21.0	51.6	7350	---	----	2935
#10	20 March	14:45	Battery	21.6	51.4	7340	116	1.91	16600
#11	20 March	16:25	Screen	21.6	51.3	4215	118	1.94	16000

(1) Measurement type; either open-faced filter holder, wire screen diffusion battery or single screen and filter combination.

(2) Condensation nuclei per cm3

(3) Geometric mean diameter (GMD) and geometric standard deviation (GSD) as determined from electrical aerosol size analyzer measurement of weighted surface area distribution.

Table 2

Equipment used by ARL for Radon Daughter Measurements at ARL

Gross alpha count method based on the modified-Tsivoglou technique.

Sampling :

1. Filter 47 mm Gelman AA membrane, 0.8 micron pore size.
2. Gelman aluminium open-face filter holder.
3. Brey carbon vane pump.
4. Dwyer rotameter, calibrated in situ against Precision wet-test meter.
5. Flow rate 4.85 $L.m^{-1}$, face velocity 6.5 $cm.s^{-1}$.

Filter Counting :

1. Windowless drawer assembly, ZnS screen, 50 mm diameter photomultiplier.
2. Portable scaler, Ludlum Model 2000.
3. Counting efficiency 0.44 cpm/dpm.
4. Background count-rate 2-3 cpm.

Calibration :

1. ^{238}Pu alpha source, electrodeposited on Kapton backing.
2. Source activity 334, 338 and 422 Bq (3 sources).
3. Active area diameter 7 mm.

Analysis method

1. Gross alpha counting analyzed using modified-Tsivoglou method (1).
2. 5 or 10 minute sample, count 2-5, 6-20, 21-30 minutes after sample.

References:

(1) Thomas,J.W., 1972, "Measurement of Radon Daughters in Air", Health Physics, 23:783-790.

Table 3

Equipment used by EML for Radon Daughter Measurements at ARL

Gross alpha count method based on Raabe-Wrenn weighted least squares technique

Sampling:

1. Filter paper : 25 mm, 0.8 micron pore size Millipore.
2. Flowrate of 3 $L.m^{-1}$, measured by flowmeter which was calibrated with standard wet or dry test meters.
3. Sampling time: 10 minutes (typical).

Filter Counting:

1. Detector system: ZnS disc in contact with filter in close proximity to a 5 cm bare photomultiplier tube (essentially windowless detector).
2. Counter efficiency: 0.48 cpm/dpm (Approximate- more exact value determined at time of each experiment).
3. Counter background: 0.1 cpm

Calibration:

1. ^{241}Am plated source, certified by US-NBS and EML.

Counting Procedure:

1. The gross alpha activity on the filter after sampling was counted in an alpha scintillation counter under the control of a microprocessor-based data logging system, SMART-1 (1). The data, which were transferred from the SMART-1 to a microcomputer at 1 minute intervals for about 40 minutes, were printed out for analysis or stored for future use.

Data Analysis:

1. The concentration of the individual radon decay products ^{218}Po, ^{214}Pb and ^{214}Bi, were determined from the counts per minute data using the Raabe-Wrenn least-squares analysis (2). The PAEC in mWL was calculated from these concentrations.

References:

(1) Polito,M.D. and Negro, V.C., 1979, "SMART-1 and Clever Counter, Environmental Data Acquisition Systems", IEEE Transactions on Nuclear Science, NS-26:765-769.
(2) Raabe,O.G. and Wrenn,M.E.,1965, "Analysis of the Activity of Radon Daughter Samples by Weighted Least Squares", Health Physics 17:593-605.

Table 4

Equipment used by USBM for Radon Daughter Measurements at ARL

Gross alpha count method based on the modified-Tsivoglou technique

Sampling :

1. Filter 47 mm Millipore type AA, 0.8 micron pore size.
2. Millipore type XX60 pump.
3. UG1 dry test meter on outlet of pump.

Filter Counting :

1. Ludlum model 182 Radon flask counter, with 7 cm diameter photomultiplier tube.
2. Filter placed on ZnS film on photomultiplier, held in contact using a weight.
3. Photomultiplier output fed to Ludlum model 2200 portable scaler.

Calibration :

1. ^{241}Am alpha source

Analysis Method :

1. Gross alpha counting analyzed using the modified-Tsivoglou method (1).

References :

(1) Thomas, J.W., 1972, "Measurements of Radon Daughters in Air", Health Physics 23:783-790.

Table 5

Summary of Wire Screen Devices for Unattached Fraction Measurement at ARL

	ARL	EML	USBM
Method	Screen and backing filter	Screen and separate filter	Three screens and backing filter
Activity analysis	Mod. Tsivoglou (See Table 2)	Raabe-Wrenn (See Table 3)	Mod. Tsivoglou (See Table 4)
Screens (a)	Stainless steel 1 screen 200 mesh	Stainless steel 3 screens #1 60 mesh #2 100 mesh #3 200 mesh	Steel or copper 3 or 4 screens (See Table 8)
Active area	40 mm dia.	39 mm dia.	20 mm dia.
Face velocity ($cm.s^{-1}$)	6.5	17.0	33.0
Collection effic. (b)	98%	-	-
Count effic. (c)	80%	-	-
Screen effic. (d)	78%	50% (60 mesh)	(e)

a) EML and USBM are developing multiple-screen methods for measuring properties of "unattached" radon daughters. However, the EML 60-mesh can be used separately to estimate the amount of "unattached" radon daughters (1).

(b) Collection efficiency is defined as the fraction of unattached radon daughters removed by the screen; for this exercise, the diffusion coefficient was assumed to be 0.05 $cm.s^{-1}$.

(c) Counting efficiency is defined as the count-rate from the screen, relative to the count rate for the same amount of activity deposited on a filter of the same face area.

(d) Screen efficiency is the product of the collection and the count efficiency.

(e) Multiple screens are designed for nanometer particle sizing. Screen efficiencies are particle size and mesh number dependent, and no single value can be specified.

References:

(1) George, A.C., 1972, "Measurements of Uncombined Fractions of Radon Daughters with Wire Screens", Health Physics 23:390-392.

Table 6

Equipment used by ARL for Radon Daughter Particle Size Distribution Measurements

1. Type :

- serial 6 stage wire screen diffusion battery

2. Characteristics of Battery :

- based on TSI Model 3040 diffusion battery
- computer sequenced valves on output ports of battery

Port	0	2	4	6	8	10
No./screens	0	3	10	21	36	55

3. Screens :

- 635 mesh stainless steel wire screens
- screen diameter 38 mm

4. Airflow :

- 4.0 $L.m^{-1}$
- single rotameter monitoring airflow through filter

5. Detection system :

- activity collected on Versapor supported membrane filter, 0.8 micron pore size, 5 mm diameter collection area.
- six solid state alpha detectors, 5 mm square, mounted in front of filter (one for each port).
- six charge sensitive preamplifiers/ amplifiers/ counters.

6. Counting regime :

- computer controlled counting for each port.
- three gross alpha counts (1).

7. Data stripping :

- Twomey nonlinear iterative algorithm (2) used in computer programme developed by EML (3).
- Number of sizes: 17
- Size range: 5 to 500 nm diameter

References

(1) Nazaroff,W.W., 1984, "Optimising the Total-Alpha Three-Count Technique for Measuring Concentrations of Radon Progeny in Residences", Health Physics 46:395-405.

(2) Twomey,S., 1975, "Comparison of Constrained Linear Inversion and an Iterative Nonlinear Algorithm Applied to the Indirect Estimation of Particle Size Distributions", J. Computational Physics, 18:188-200.

(3) Knutson,E.O., 1984, Private Communication.

Table 7

Equipment used by EML for Radon Daughter Particle Size Distribution Measurements

1. Type :

- parallel 5-stage tubular diffusion battery system

2. Specification of diffusion battery (1) :

Battery	Number of sections(2)	Total length of sections,cm	Equivalent length, m
1	1	0.34	50
2	2	0.95	138
3	5	8.08	1172
4	10	25.75	3735

3. Airflow :

- 3 $L.m^{-1}$, measured by flowmeter in each stage.

4. Radioactivity determination :

- same as radon daughter method.

5. Particle size determination :

-Twomey's nonlinear iterative algorithm (3) was used to calculate activity weighted particle size distribution at 19 sizes ranging from 2.8 to 1368 nm diameter.

References

(1) George,A.G.,Hinchcliffe,L. and Sladowski,R., 1975, "Size Distribution of Radon Daughter Particles in Uranium Mine Atmospheres", J. American Industrial Hygiene Assoc., 36:484-490.

(2) Sinclair,D., "A Portable Diffusion Battery", 1972, J American Industrial Hygiene Association, Nov., 33:729-733.

(3) Twomey,S., 1975, "Comparison of Constrained Linear Inversion and an Iterative Nonlinear Algorithm Applied to the Indirect Estimation of Particle Size Distributions", J. Computational Physics, 18:188-200.

Table 8

Equipment used by USBM for Radon Daughter Particle Size Distribution Measurements

1. Type :

- serial 3 or 4 stage graded wire screen diffusion device (1).

2. Characteristics of device :

- wire screens stacked in an extended filter holder.
- filter as last stage.
- screens stacked with smaller mesh numbers to front.
- particles with smaller diameters collected on low mesh screens.

3. Screens :

- 40,60,80,100,120,200,400,500,635 mesh per inch screens.

4. Airflow :

- Millipore type XX60 pump, UG1 dry test meter.
- 3.7 $L.m^{-1}$.

5. Detection System :

- ZnS film on 7 cm diameter photomultiplier tube.
- Ludlum model 2200 portable scaler.

6. Counting Method :

- Gross alpha counting of screens.
- analyzed using modified-Tsivoglou method.
- if conditions are steady then both front and back of screens are counted.

7. Data Stripping :

- Adapted Twomey nonlinear iterative algorithm.

Reference

(1) Holub, R.F. and Knutson, E.O., 1987, "Measuring Po-218 Diffusion Coefficient Spectra using Multiple Wire Screens", In: **Radon and its Decay Products**, Ed. Hopke, P.K., Am. Chem. Soc., ., Washington D.C., pp 311-356.

Table 9

Comparison of Counting Efficiencies using Calibrated Sources

Reference source	Activity Bq (1)	Detector Efficiency (cps/Bq) ARL #1	ARL #2	EML	USBM
ARL #1	422.5	0.425	0.422	0.488	0.494
ARL #2	334.4	0.423	0.421	0.484	0.485
ARL #3	338.1	0.426	0.432	0.490	0.490
EML #1	175.6	0.443	0.444	0.487	0.492
USBM #1	209.2	0.436	0.435	0.490	0.494
Mean	------	0.431	0.431	0.488	0.491
SD.	------	0.009	0.009	0.002	0.004

(1) Quoted value is activity of radionuclide deposited on backing material.

Table 10

Results of Radon Daughter Measurement Intercomparison at ARL

Test	Parameter	Activity Concentration $Bq.m^{-3}$ and PAEC, mWL (1) ARL	EML	USBM
#1	^{218}Po	944 ± 180	827 ± 104	2160 ± 275
	^{214}Pb	529 ± 32	560 ± 12	1486 ± 60
	^{214}Bi	395 ± 44	414 ± 20	987 ± 74
	PAEC	139 ± 3	142 ± 4	364 ± 4
#2	^{218}Po	179 ± 40	203 ± 30	551 ± 61
	^{214}Pb	16 ± 8	9 ± 3	92 ± 12
	^{214}Bi	13 ± 10	0.4 ± 5	62 ± 15
	PAEC	7.5 ± 0.6	7.0 ± 0.3	22 ± 1
#3	^{218}Po	3534 ± 227	3794 ± 217	3174 ± 228
	^{214}Pb	1143 ± 45	1155 ± 28	1057 ± 39
	^{214}Bi	424 ± 60	483 ± 45	563 ± 56
	PAEC	299 ± 4	314 ± 9	292 ± 3
#5	^{218}Po	3659 ± 292	3427 ± 243	3725 ± 537
	^{214}Pb	1859 ± 59	1806 ± 27	1886 ± 138
	^{214}Bi	1191 ± 78	1165 ± 44	1013 ± 167
	PAEC	478 ± 5	462 ± 9	466 ± 10
#7	^{218}Po	1052 ± 109	1055 ± 61	806 ± 110
	^{214}Pb	153 ± 20	194 ± 8	163 ± 20
	^{214}Bi	110 ± 27	50 ± 13	121 ± 27
	PAEC	62 ± 2	64 ± 2	57 ± 2

(1) Quoted uncertainties are 1 standard deviation from counting statistics

Table 11

Results of Unattached Fraction Measurements at ARL

Test #9		Aerosol concentration = 7350 CN cm^{-3}			
Lab.	Param. (1)	^{218}Po $Bq.m^{-3}$	^{214}Pb $Bq.m^{-3}$	^{214}Bi $Bq.m^{-3}$	PAEC mWL
ARL	Total	568 ± 94	139 ± 20	59 ± 27	40.3 ± 1.4
	Free	152 ± 41	3 ± 7	1 ± 10	4.1 ± 0.6
	%	27 ± 9	2 ± 5	2 ± 17	9.5 ± 0.7
EML	Filter	464 ± 68	112 ± 7	87 ± 10	37.3 ± 2.4
	#1 (60 mesh)	74 ± 14	0 ± 2	0 ± 2	1.7 ± 0.5
	#2 (100 mesh)	90 ± 15	0 ± 2	0 ± 2	1.9 ± 0.5
	#3 (200 mesh)	45 ± 13	0 ± 1	3 ± 2	1.6 ± 0.5
	Free	148 ± 28	0 ± 4	0 ± 4	3.4 ± 1.0
	%	32 ± 8	0 ± 4	0 ± 5	9.1 ± 2.7
USBM	Filter	378 ± 95	109 ± 19	71 ± 25	32.9 ± 2.1
	#1 (400 mesh)	59 ± 28	0 ± 5	0 ± 6	1.2 ± 0.4
	#2 (80 mesh)	31 ± 24	3 ± 4	0 ± 6	1.2 ± 0.4
	#3 (40 mesh)	41 ± 16	0	0	0.5

Test #11		Aerosol concentration = 4215 CN cm^{-3}			
Lab.	Param. (1)	^{218}Po $Bq.m^{-3}$	^{214}Pb $Bq.m^{-3}$	^{214}Bi $Bq.m^{-3}$	PAEC mWL
ARL	Total	3004 ± 206	706 ± 40	491 ± 53	230 ± 10
	Free	709 ± 78	36 ± 14	0 ± 19	23 ± 3
	%	24 ± 3	5.1 ± 2	0 ± 20	10 ± 2
EML	Filter	3044 ± 742	743 ± 21	481 ± 35	236 ± 7
	#1 (60 mesh)	345 ± 39	6.9 ± 4	4.8 ± 6	11 ± 1
	#2 (100 mesh)	463 ± 46	8.1 ± 5	0 ± 7	13 ± 2
	#3 (200 mesh)	542 ± 40	9.8 ± 4	0 ± 6	15 ± 1
	Free	690 ± 78	13.8 ± 8	9.6 ± 12	22 ± 2
	%	23 ± 6	1.9 ± 1.1	2.0 ± 2.5	9.3 ± 0.9
USBM	Filter	2704	744	385	218
	#1 (635 mesh)	44	13.0	15.5	5
	#2 (100 mesh)	326	15.2	14.4	10
	#3 (40 mesh)	218	6.7	1.7	7

(1) Total refers to total concentration of attached and unattached species. Free refers to concentration of "unattached " species. The derived unattached fraction, in percent, is listed in the row "%".

Table 12

Results of Calculation of Activity Median Aerodynamic Diameter (AMAD) and Geometric Standard Deviation (GSD) from PAEC Distribution Data.

Test	[Aerosol] $CN.cm^{-3}$	TSI (1) GMD nm	TSI (1) GSD	ARL AMAD(2) nm	ARL GSD	EML AMAD nm	EML GSD	USBM AMAD nm	USBM GSD
#4	63000	103	1.78	119 $^{+2}_{-9}$	1.8 $^{+0.1}_{-0.0}$	122	1.70	63	1.64
				---	---	13	1.26	0.6	1.00
#6	60000	---	----	122 $^{+46}_{-22}$	1.9 $^{+1.2}_{-0.4}$	113	1.81	---	----
				---	---	12	1.43	---	----
#8	7360	115	1.91	120	1.5	157	1.71	107	1.38
				---	---	2.6	1.27	1.4	1.30
#10	7340	129	2.16	117	1.6	148	1.91	130	1.00
				---	---	2.7	1.30	1.0	1.00

(1) Geometric mean diameter (GMD) and geometric standard deviation (GSD)of surface area distribution as determined from TSI electrical aerosol analyzer measurement.

(2) Quoted uncertainties for ARL values of AMAD and GSD are derived from Monte Carlo analysis of the effect of one standard deviation uncertainty in the Twomey deconvolution analysis.

Table 13

Results of Activity Median Aerodynamic Diameter (AMAD) and Geometric Standard Deviation (GSD) from ^{218}Po and ^{214}Pb Activity Concentration Distribution Data (1)

Test No.	Lab	^{218}Po AMAD nm	^{218}Po GSD	^{214}Pb AMAD nm	^{214}Pb GSD
#4	ARL	116 $^{+44}_{-21}$	1.8 $^{+1.0}_{-0.4}$	115 $^{+2}_{-9}$	2.0 $^{+0.2}_{-0.3}$
		3.4 $^{+2.7}_{-0.4}$	1.4 $^{+0.5}_{-0.1}$	4.3 $^{+1.4}_{-0.8}$	1.8 $^{+0.1}_{-0.2}$
	EML	133	1.7	131	1.6
		11	1.2	13	1.2
	USBM	45	1.3	72	1.9
		0.6	1.0	---	---
#6	ARL	255 $^{+148}_{-13}$	1.7 $^{+4.3}_{-0.7}$	97 $^{+50}_{-17}$	1.9 $^{+1.6}_{-0.7}$
		3.3 $^{+13}_{-0.4}$	1.5 $^{+0.5}_{-0.5}$	16 $^{+0}_{-12}$	1.1 $^{+2.0}_{-0.1}$
	EML	131	1.4	103	1.8
		5.6	1.6	---	---
	USBM	---	---	---	---
		---	---	---	---
#10	ARL	93 $^{+90}_{-46}$	1.7 $^{+2.0}_{-0.7}$	109 $^{+97}_{-25}$	1.5 $^{+0.8}_{-0.3}$
		9.0 $^{+7.1}_{-6.2}$	2.0 $^{+0.0}_{-0.9}$	14 $^{+2}_{-12}$	1.4 $^{+0.6}_{-0.4}$
	EML	141	1.5	149	2.2
		2.5	1.3	2.7	1.3
	USBM	130	1.0	129	1.3
		0.9	1.2	1.2	1.1

(1) Uncertainty ranges quoted for ARL measurements are derived from a Monte Carlo calculation of the effect of 1 standard deviation uncertainties in the Twomey deconvolution program.

Appendix 1

Designated Radon Reference Facilities for the International Intercomparison and Intercalibration Programme for Radon, Thoron and their Daughter Measurement

Europe (CEC)	: National Radiological Protection Board Didcot, United Kingdom (John Strong)
North America	: Environmental Measurement Laboratory New York, N.Y. , U.S.A (Earl Knutson)
	: U.S. Bureau of Mines Denver, Colorado, U.S.A. (Robert Holub)
Pacific region	: Australian Radiation Laboratory Melbourne, Australia (Stephen Solomon)

Appendix 2

The Australian Radiation Laboratory Radon Test Facility

A2.1 INTRODUCTION

The radon test chamber at the Australian Radiation Laboratory was developed to provide controlled atmospheres of radon and radon daughters for the evaluation and calibration of monitoring and measurement equipment. The chamber is described in terms of the control and monitoring instrumentation, and by the sampling facilities.

A2.2 PRODUCTION AND CONTROL OF TEST ATMOSPHERES

The radon test facility at ARL comprises two air-tight, copper-lined rooms of dimensions 1.65 x 1.75 x 2.75 m, with a total effective volume of 14.4 m^3. These chambers are connected, to form a closed loop, by 0.1 m diameter copper tubing and the air is circulated between the two chambers at 5 m^3 per minute. The air conditioning unit, the radon source and the aerosol generator are directly connected to chamber #1, while chamber #2 forms the main sampling volume. The main components of the environmental control system are;

(a) Temperature and humidity control. The temperature and humidity in both chambers are maintained at the standard values of 20 ± 1 C and 50 ± 5% RH respectively, using a closed cycle air conditioning unit mounted on the inside of the roof of chamber #1.

(b) Radon concentration. The radon concentration in the chambers is normally maintained at 3700 ± 100 $Bq.m^{-3}$ using a flow-through radon source. This source consists of a large piece of uranium ore in a sealed container, through which filtered room air is passed at 0.010 m^3 per minute. This flow rate controls the radon concentration in the chamber, since all other air transfers to and from the chamber are operated in a closed loop (i.e. sampled air is returned to the chamber). The radon concentration in the chambers can be modified by adjusting the radon source flow rate or by adding radon from a radium solution source (source activity 460 kBq).

(c) Aerosol concentration. A low temperature flow through furnace based on an EML design (Tu, 1982) was used to produce a monodispersed carnauba wax aerosol of 100 nm diameter over a concentration range of 1000 to 100000 aerosol per cm^3. The generator output maintained the aerosol concentration within a ± 10% range over periods of days.

(d) The equilibrium factor in the chambers can be varied independently of the aerosol concentration by adjusting the air residence time. The fan for the air-conditioning unit can be adjusted to vary the amount of plate-out on the condenser coils. Further control is achieved by recirculating the chamber air through a filter using an air pump. This pumping system comprises two Thomas compressors with a combined maximum throughput of 0.4 m^3 per minute, adjustable down to 0.30 m^3 per minute.

A2.3 MONITORING OF CHAMBER CONDITIONS

Most of the measurement and data acquisition associated with the chambers is handled by a CAMAC based PDP11-02 mini-computer. This system includes a 4096 channel, 50MHz pulse height analyzer, a 16 channel, 12 bit resolution monitoring analog to digital converter (ADC), a 4 channel, 100MHZ scaler, together with dual disk drives for data storage. This system continuously monitors the chamber conditions, using a variety of sensors, and logs the data to disk for later analysis. The sensors include;

(a) Analog sensors for temperature, humidity, flow rate of radon source and aerosol concentration (measured using an Environment One Rich 200 condensation nuclei counter).

(b) Digital inputs from the continuous radon and radon daughter monitors connected to chamber #1. The radon monitor is a 1 litre volume, flow-through scintillation cell. The radon daughter monitors consist of two sampling heads, each containing a filter and solid state alpha detector. Monitor #1 collects both attached and unattached radon daughters. Monitor #2 has a 200 mesh stainless steel screen on its input from the chamber and collects only radon daughters attached to aerosols. These two monitors continuously sample the chamber atmosphere and allow the determination of potential alpha energy concentration (PAEC) and of the unattached fraction of PAEC.

A2.4 SAMPLING FACILITIES

Access to the chambers is through an air-tight hatch on one wall of each chamber. These hatches are rectangular, 0.90 m high and 0.60 m wide, and are used to move large instruments into the chambers. Chamber #1 also has a small airlock (approximately 0.40 m square) allowing smaller equipment (i.e. scintillation cells or filter holders) to be transferred into the chamber. A pair of heavy duty rubber gloves, mounted on the wall above this airlock box, allows equipment to be moved from the glove box onto the equipment racks within the chamber.

There are five sampling ports on chamber #2 and one on chamber #1. Four of these ports are used for side-by-side measurements of radon daughters. These ports accept filter holders up to 55 mm in diameter, and there is provision for the sampled air to be returned to the chamber.

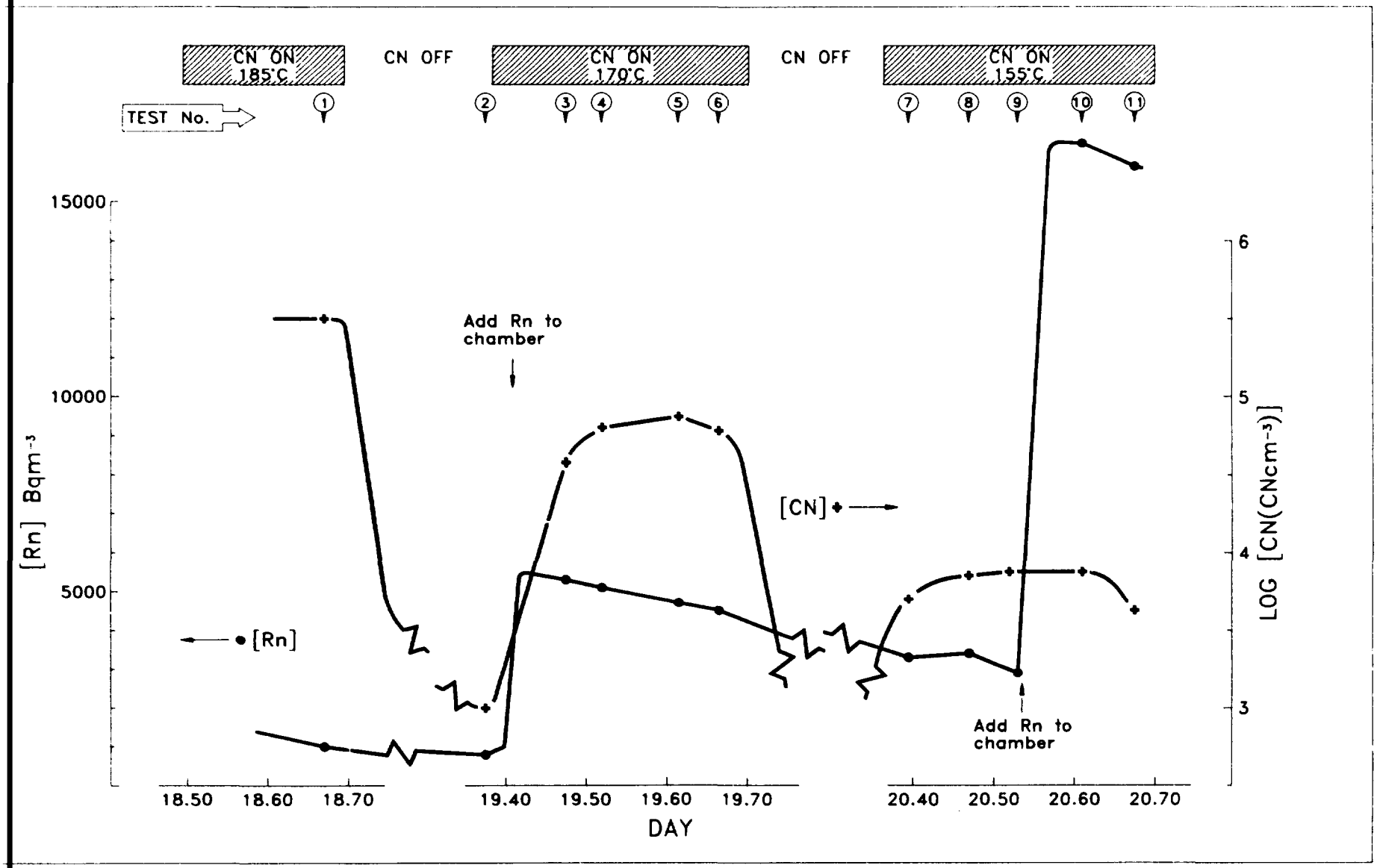


Figure 1. Radon and Aerosol Concentrations in ARL Radon Test Chamber during Radon Daughter and Particle Size Intercomparison. The label "CN" indicates periods of operation of the aerosol generator.

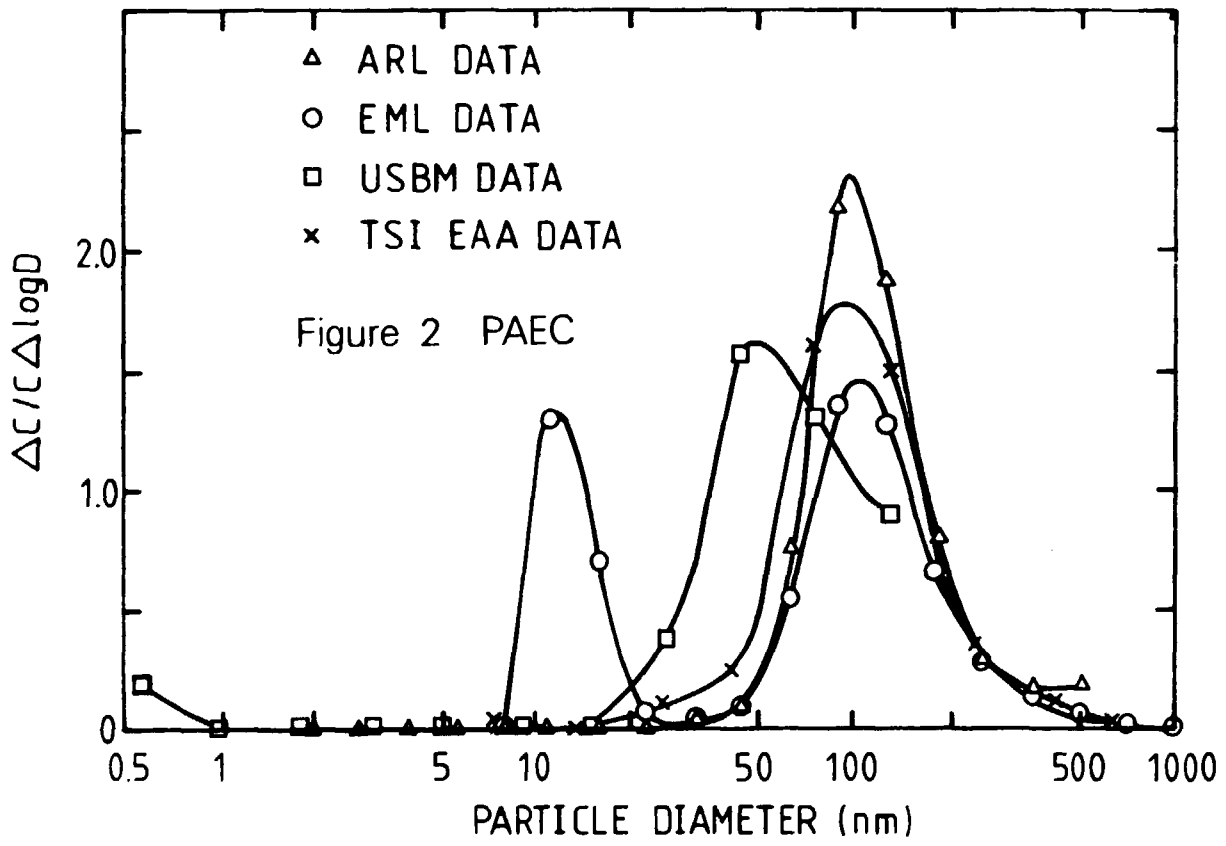
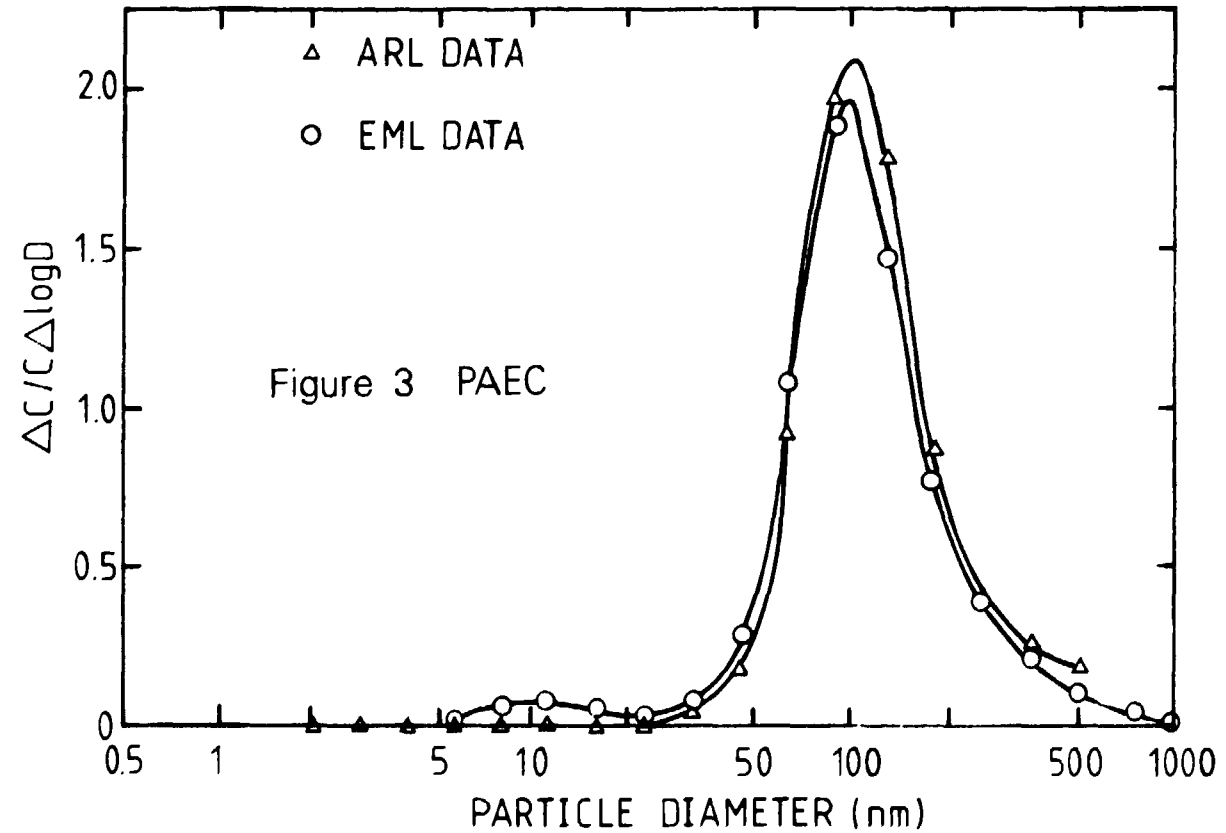


Figure 2. Derived PAEC Particle Size Distributions for ARL Test #4.



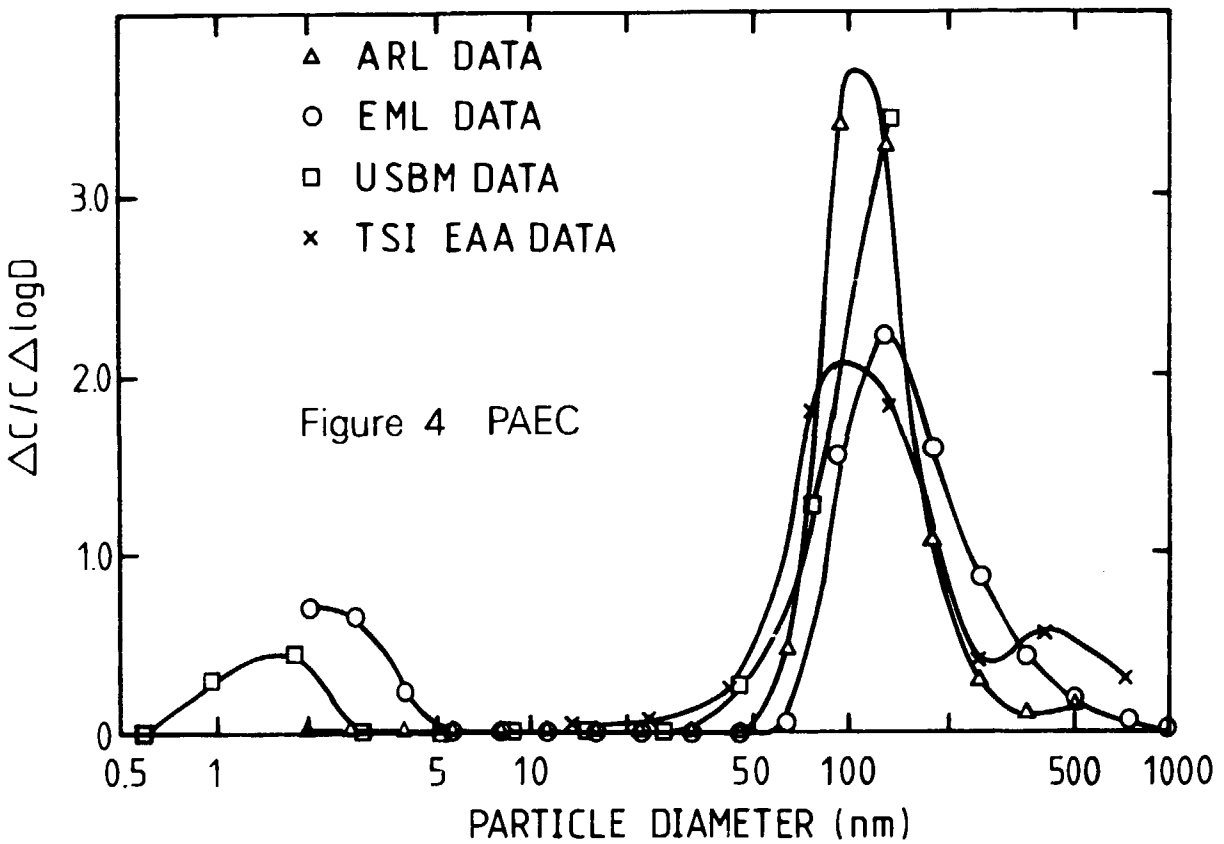
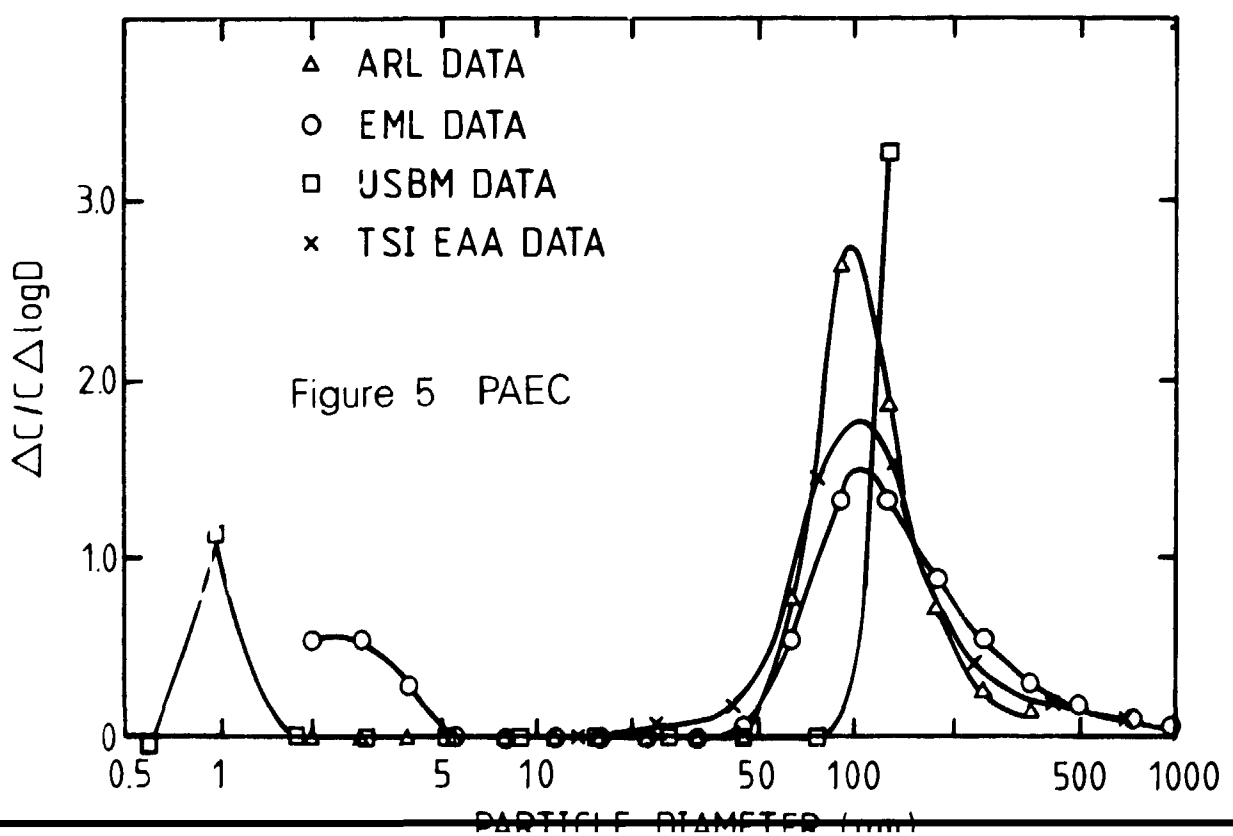


Figure 4. Derived PAEC Particle Size Distributions for ARL Test #8.



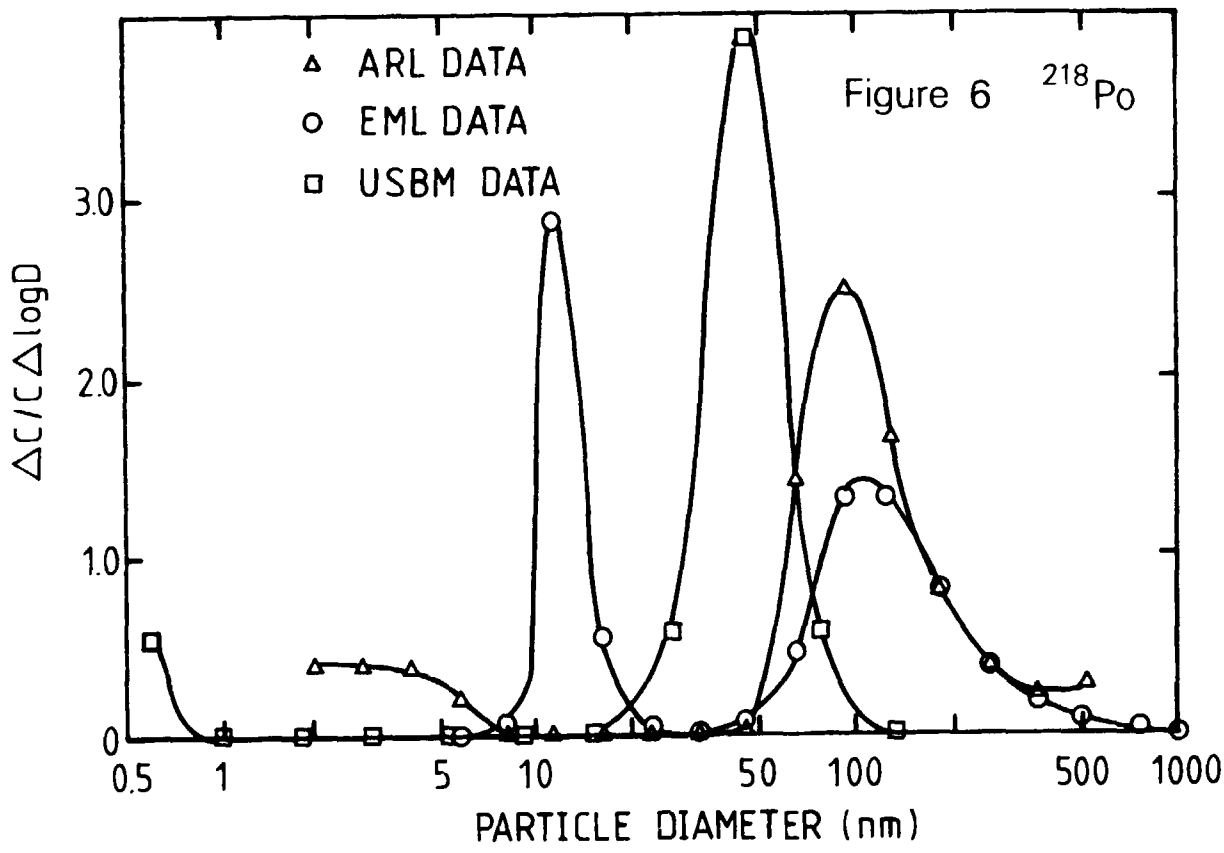
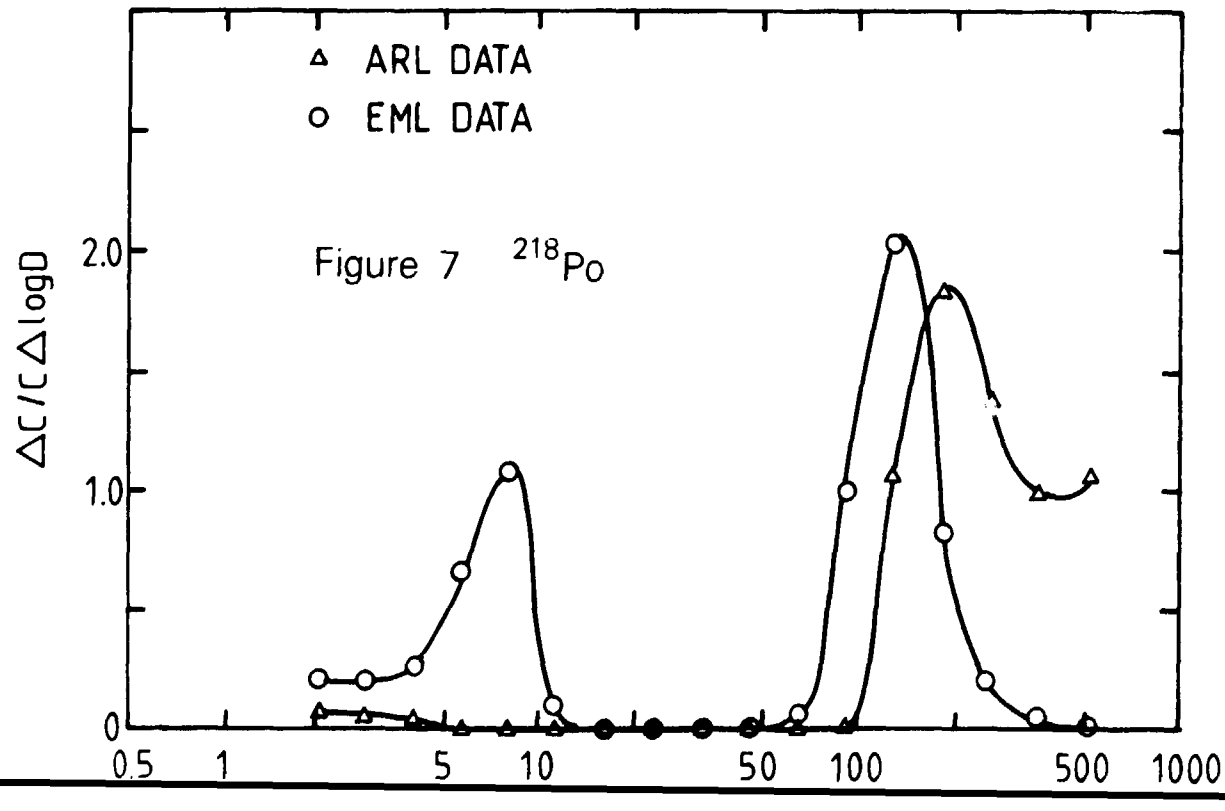


Figure 6. Derived ^{218}Po Particle Size Distributions for ARL Test #4.



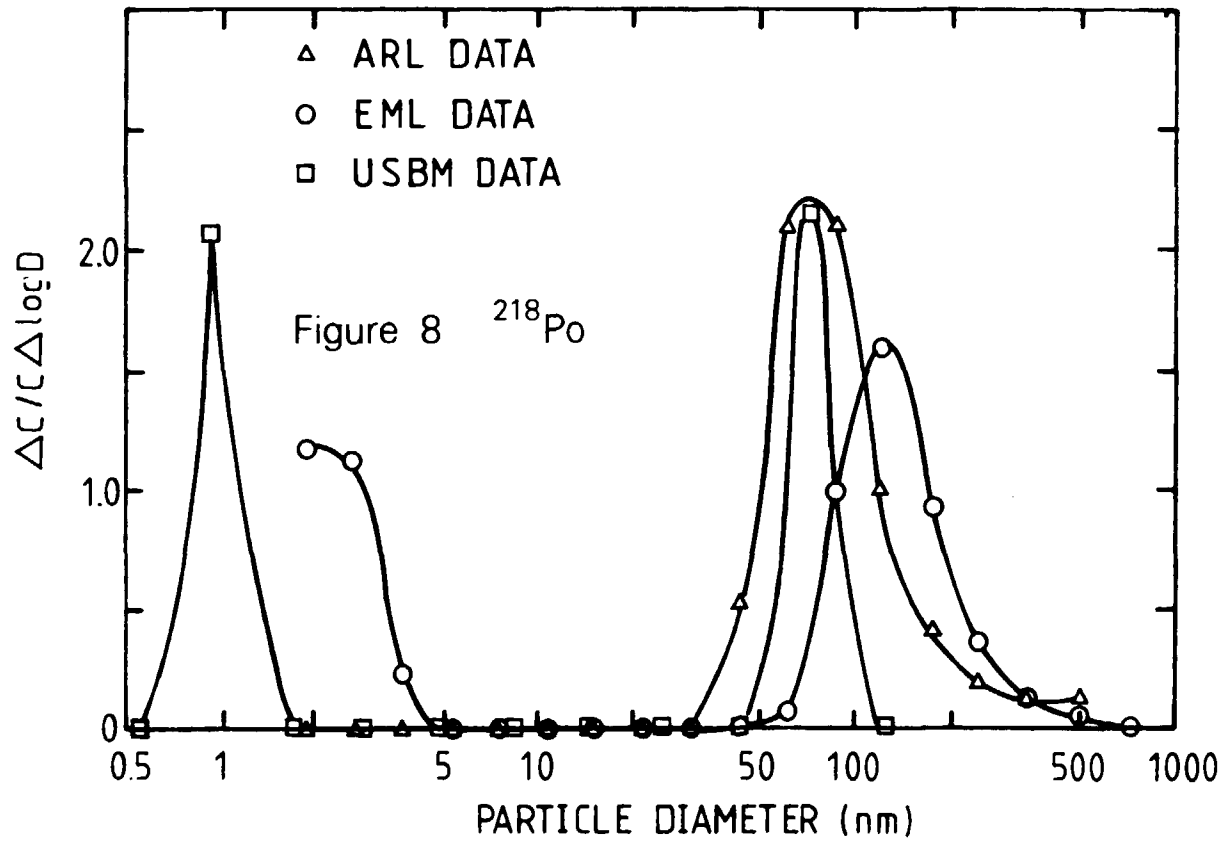
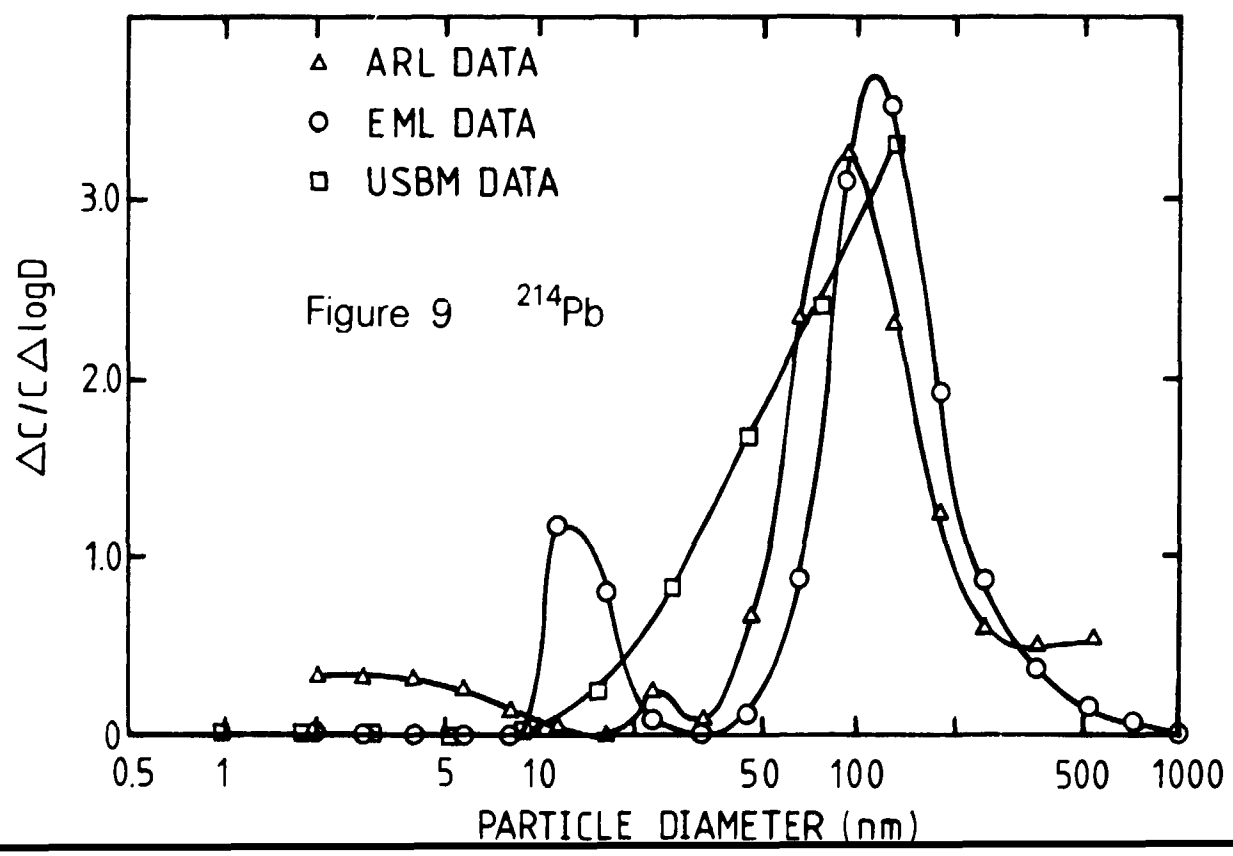


Figure 8. Derived ^{218}Po Particle Size Distributions for ARL Test #10.



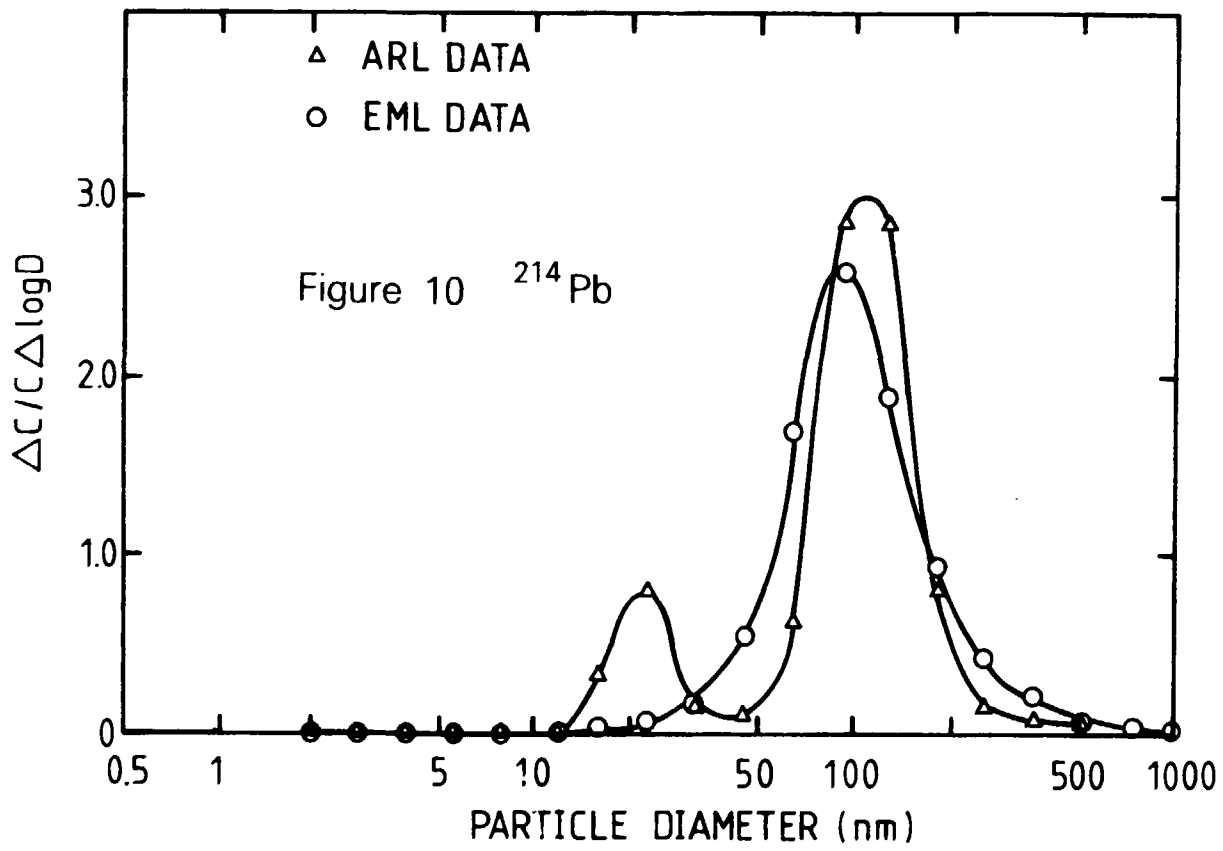


Figure 10. Derived ^{214}Pb Particle Size Distributions for ARL Test #6.