# RAPID DETERMINATION OF THE RADON PROFILE IN A STRUCTURE BY MEASURING IONS IN THE AMBIENT ATMOSPHERE

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## ABSTRACT

Normal background radiation produces about five (5) ion pairs per cubic centimeter per second with an average lifetime of about five minutes and an average concentration of about 1,600 ions per cubic centimeter. Elevated radon in the air also increases the ion concentration in the air. A simple and inexpensive electrostatic measuring device<sup>\*\*</sup> using a charged metal sphere and an electrostatic charge detector enables one to detect low ion concentrations in the air. Assuming that the increased ion concentrations are due to radon, radon mitigators may rapidly determine the relative ion concentrations in different parts of a structure and therefore pinpoint the radon entry points at the location of highest ion concentration. A mathematical model of the ion concentration in air as a function of radon concentration will be presented. Actual measured ion concentrations in structures will also be presented.

\*\*Patent disclosure document filed

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# INTRODUCTION

This paper describes an economical, reliable, and sensitive apparatus and method to measure air ions. Since radon produces air ions and, if the air ions are principally produced by radon, then this apparatus enables an individual to indirectly infer the radon concentration. A radon mitigator using the proposed ion collecting apparatus within various locations of a structure and controlling the atmosphere such that most ions are produced by radon, the profile of ions in the structure enables the high ion concentrations to be located and facilitates one to locate the entry of radon into the structure.

The ion collector consists of a metal sphere attached to a cylindrical container by using glue and Teflon. The electrical charge on the sphere is determined, or read, by placing the sphere next to a charged surface which is connected to a sensitive electronic reader.

Sources of air ions may also be produced naturally by background radiation, lightning discharges, high energy ultraviolet radiation, and the friction effects of wind, rain, snow, and hail. Man made sources of air ions consist of air ion generators, high voltage direct current transmission lines, electrostatic precipitators, and friction of the air as it moves over metal surfaces such as in heat ducts. One may easily eliminate the major sources of air ions in a structure with the exception of background radiation and radon by turning the air handling system off.

## AIR ION CONCENTRATIONS PRODUCED BY RADIATION

Background radiation of 9 microroentgens per hour (79 mR/year) produces about 5 ion-pairs per cubic centimeters per second in air. The mean life of the ionpairs is 300 seconds and in ordinary air at sea level there are about  $1.6 \times 10^3$  ionpairs per cubic centimeter produced by background radiation.(1)

Let us now calculate the expected number of ion-pairs per cubic centimeter for four picocuries per liter (4 pCi  $L^{-1}$ ) of radon existing under secular equilibrium conditions. With the disintegration of each radon-222 nucleus, three alpha particles are emitted in the radon series with a total energy of 19.17 MeV. Since it requires about 34 electron volts to produce an ion pair in air, we have

$$19.17 \times 10^{6} \frac{eV}{dis} \times \frac{i.p.}{34eV} = 5.6 \times 10^{5} \frac{i.p.}{dis}$$
 Eq. 1

For a radon concentration of 4 pCi L<sup>1</sup>, we may then determine the average

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phone is presented a special cards it summarian the strength of the

number of ions produced per cubic centimeter per second as follows:

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$$pCi \ L^{-1} \ x \ 3.7x10^{-2} \frac{dis}{\sec} \frac{1}{pCi} \ x \ 5.6x10^{5} \frac{i.p.}{dis} x \frac{1 \ L}{1000 \ cm^{3}}$$
  
= 82.8  $i.p. \ cm^{-3} \sec^{-1}$  Eq. 2

For alpha particle columnar ionization, many of the ion pairs will readily recombine. Even if one-half of the ion-pairs readily recombine, a concentration of 4 pCi L<sup>-1</sup> of radon will have a production rate of air ions(41 i.p. cm<sup>-3</sup> sec<sup>-1</sup>) which is about 8 times that of the normal background level(about 5 i.p. cm<sup>-3</sup> sec<sup>-1</sup>). Kanne and Bearden(2) published an article concerning the collection of ions produced by Columnar Ionization in which they found over 50% of the ions produced by alpha particles were collected even in low electric fields (8 volts/cm).

Let us consider a gas containing  $N_1$  and  $N_2$  positive and negative ions per cm<sup>3</sup> respectively, then we define the recombination coefficient  $\beta$  by the relation

$$\frac{-dN_1}{dt} = \frac{-dN_2}{dt} = \beta N_1 N_2$$
 Eq. 3

and since for our case we assume the positive and negative ions in air are equal, then

$$\frac{-dN}{dt} = \beta N^2 \qquad \text{Eq. 4}$$

-dN/dt is the rate at which ions recombine and values of the recombination coefficient(3) have been measured and  $\beta$  is of the order of  $2 \times 10^{-6}$  cm<sup>3</sup> ion<sup>-1</sup> sec<sup>-1</sup> in air.

At equilibrium, the production rate (p) is equal to the recombination rate; therefore,

$$\frac{dN}{dt} = p - \beta N^2 = 0 \qquad \text{Eq. 5}$$

or

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$$N=\sqrt{\frac{p}{\beta}}$$
 Eq. 6

Assuming that  $\beta$  is a constant and equal to  $2 \times 10^{-6}$  cm<sup>3</sup> ion<sup>-1</sup> sec<sup>-1</sup>, we obtain the number of ions per cubic centimeters at equilibrium for different production rates and the values appear in Table 1 and are plotted in Figure 1. Note that under these assumptions that a production rate increase over the original production rate of four times yields double the original equilibrium concentration of ions. It is then obvious, as the radon concentration increases, the equilibrium ion concentrations also

increases even through  $\beta$  may change and increase significantly as the ion concentration increases.

Radon (pCi L <sup>-1</sup> )	p * (ions cm <sup>-3</sup> sec <sup>-1</sup> )	N (ions cm <sup>-3</sup> )
0	5	1600
4.3	50	5000
47.8	500	15811
482.6	5000	50000

	TA	BLE 1			
Number of Equilibrium	Ions '	Versus	Production	Rate of	Ions(p)

\*Assume one-half of the ions produced by radon readily recombine and have only included one-half of the produced ions under p.



Figure 1. The logarithm of the equilibrium concentration of ions versus the logarithm of the production rate, assuming the recombination coefficient  $\beta$  is constant and equal to  $2 \ge 10^{-6}$  cm<sup>3</sup> ion<sup>-1</sup> sec<sup>-1</sup>.







# DESIGN AND DEVELOPMENT OF A SPHERICAL ION COLLECTOR

The Spherical Ion Collector (SIC) was designed to contain sufficient charge so that its magnitude was easily measured, yet the quantity of charge was such that it was sensitive to a change in charge for a small number of ions collected. It is also necessary to maintain the electric field near its surface so that the electric field is below the magnitude which produces avalanche ionization. In order to enclose the spherical ionization collector (SIC), the metal ball was glued onto a 1/16" thick piece of PTFE Teflon<sup>1</sup> and the Teflon was then glued to the bottom of a 30 mL cylindrical container. The container could be closed by placing a conducting top on the cylindrical can. The SIC ion collector is illustrated in Figure 2.



Figure 2. Illustration of the Spherical Ion Collector

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<sup>&</sup>lt;sup>1</sup> Teflon<sup>R</sup>PTFE fluorocarbon, manufactured by E.I. duPont De Nemours and Co.Inc., Wilmington, DE 19898

To charge the sphere, a high voltage power supply potential of +400 volts was applied to the sphere and the ground was connected to the outer cylindrical can. The sphere may also be charged by using the method of induction. That is, a negatively charged object is brought near the sphere, the sphere is touched for an instant to allow part of the negative charge to leave the sphere, then the charged object is removed and the sphere is now positively charged. This produced a charge on the sphere of about  $1.2 \times 10^{-10}$  coulombs. The electric field at the surface of the sphere is given by using Gauss's law to yield

$$E = 9 \times 10^9 \frac{N m^2}{C^2} \times \frac{1.2 \times 10^{-10} C}{(0.009 m)^2} = 13333 \frac{volts}{meter}$$
 Eq. 7

A method known as the shutter method(4), also called the capacitive probe method, was used to measure the surface potential, and therefore the charge on the sphere. A homemade inexpensive meter as well as an electret reader produced and sold by RAD ELECT INC.(5) were used to measure the charge on the spherical ion collector. The cylindrical can is positioned where electrets are usually placed on the reader to determine their charge (or voltage). In this instrument, a metallic shutter shields the electrostatic field of the charged sphere while the meter is "zeroed." When the shutter is opened, a charge is induced on a conducting plate connected to a high impedance Op-Amp circuit and the read out is displayed on an LCD meter. The reproducibility of this reading was within one volt over the range of voltages used for the SICs.

After closing the SIC container with negligible radon in the container, a discharge rate of 7.5 volts per day was observed because of background radiation producing ion pairs in the 27 cm<sup>3</sup> (30 cm<sup>3</sup>-sphere volume) of air. The background radiation was about 10 microroentgens per hour and the total negative charge ( $\Delta q$ ) produced in the 27 cm<sup>3</sup> air volume was about

$$\Delta q = 10 \times 10^{-6} \frac{R}{h} \times 24h \times \frac{2.08 \times 10^{3} (i.p. cm^{-3})}{1 \times 10^{-6} R} \times \frac{1 e^{-}}{i.p.} \times \frac{1.6 \times 10^{-19} C}{e^{-}} \times 27 cm^{3}$$
$$= 2.2 \times 10^{-12} C \qquad \text{Eq. 8}$$

Since the change in voltage per day produced by background radiation was 7.5 volts, we obtain the charge per volt constant(k) for the SIC to be

$$K = \frac{2.2 \times 10^{-12} C}{7.5 \text{ volts}} = 2.9 \times 10^{-13} \frac{C}{V}$$
 Eq. 9

For curious individuals who use short term electrets to make radon measurements, the short term electrets have a constant of about  $9.5 \times 10^{-12}$ C/V. Therefore, the SIC is about 30 times as sensitive as the short term electret.

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# EVALUATION OF THE SPHERICAL ION COLLECTOR

# CHARGE STABILITY

The stability of charge on the spherical ion collector depended upon the background radiation, radon in the cylinder containing the sphere, and leakage due to moisture. Background radiation alone caused the voltage to decrease about 7.5 volts per day. By using reasonable care not to expose the ion collector and Teflon to overly humid environments, no difficulty with leakage currents were encountered. The closed detectors were placed in an open pan where rain actually wetted the tops of the relatively closed containers and after one day the voltage on these detectors had decreased about 10 volts, which is slightly more than the voltage decrease caused by background radiation.

## CALIBRATION OF THE SPHERICAL ION COLLECTOR

The air handling system was turned off in a house for thirty minutes before making any radon measurements. Four charged SIC detectors were placed with open tops on a table for six minutes. The detectors discharged from 400 volts to about 304 volts over a period of six minutes. During the exposure of the SICs, an open container was allowed to come to equilibrium with the air in the center of the table and E-PERMS placed in this container for 24 hours indicated a radon concentration of 17.5 pCi L<sup>-1</sup>. One day later, three SICs detectors were exposed to a radon concentration of nine pCi L<sup>-1</sup> and the average voltage on these detectors during a 6 minute exposure decreased from 400 volts to 332 volts or a decrease of 68 volts. This indicates a total negative charge collection of

68 volts 
$$\times 2.9 \times 10^{-13} \frac{C}{V} = 1.97 \times 10^{-11} C$$
 Eq. 10

## MEASUREMENTS IN A STRUCTURE

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The SICs have been used to measure the ions in various structures. The ion concentration and the radon concentration were studied in a structure. The air handling system was turned off thirty minutes prior to initiating measurements and three fans located upstairs were turned on for thirty minutes to increase the radon concentration in the structure. After turning off the fans, the SICs were charged to 400 volts, read on a reader, and then placed in the appropriate room. The top was removed to allow each SIC to be exposed for ten minutes. A Lucas Cell and the associated apparatus were used to determine the radon concentration. Table 2 contains the change in voltage of the SICs and the radon concentration at each

location.

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Location	Mean Change in Voltage (volts)	Radon Concentration (pCi L <sup>-1)</sup>
1. Basement (Radon Room)	131	22.5
2. Radon Room (At Peak Level)	161	28.5
3. Adjacent to Radon Room	85	10.0
4. Opposite Side of Basement from Radon Room	73	8.4
5. Upstairs	-73	3.8
6. Outside Air	7	less than 1.0

TABLE 2 Changes in voltages of the SICs and the Radon Concentrations

Typical fractional standard deviations for three SICs used at each location were about ten percent. Note that in Table 2, as the radon concentration increased the mean of the change in voltage also increased. These measurements obviously provide a good correlation between the magnitude of the radon concentration and the change in voltage of the open SICs. Note that for the fifth measurement, the radon concentration was only 3.8 pCi L<sup>-1</sup> but the change in voltage was 73 volts. A fire was burning in the fireplace near this location. The ions produced by the fire probably provided the elevated ion concentration.

For other measurements, significant exceptions were found. Using the open SICs, the detector is vulnerable to environmental effects such as charged objects or ions produced by other means. One structure had high radon concentrations, about 20 pCi  $L^{-1}$ , but a very low ion concentration. Many objects, such as plastics, are electrically charged and when located near the SICs, they compete with the SICs for the ions. This, of course, can cause false readings.

## CONCLUSIONS

Spherical ion collectors used with charge readers to detect air ion concentrations are economical, reliable and sensitive. By directly exposing the SICs

to the environment, one must be careful to eliminate other sources or sinks of ions in order to obtain accurate results. By placing the SICs in one or two liter containers, sufficient sensitivity should still exist for the SICs to provide rapid radon measurements and also eliminate many of the competing environmental factors. These measurements are in progress.

Two significant advantages for using the spherical ion collectors over electrets for air ion collection are the relative ease with which one can recharge the spheres and the sensitivity of the spherical ion collector. One may utilize a charged object to readily recharge the sphere.

> The work described in the paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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## INTERCOMPARISON OF ACTIVITY SIZE DISTRIBUTION MEASUREMENTS WITH MANUAL AND AUTOMATED DIFFUSION BATTERIES - FIELD TEST

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#### ABSTRACT

To compare the performance of the Graded Screen Array (GSA) technique for measurement of radon decay product activity size distributions in a real house environment, a series of experiments were performed in a single family house in Princeton NJ. The present study was designed as a field test following to the laboratory intercomparison measurements carried out previously in a radon-aerosol chamber. Two different systems were used:

1) Disk-type Diffusion Battery from the Environmental Measurements Laboratory.

2) Automated, Semi-Continuous System (ASC-GSA) from Clarkson University.

Several sets of parallel measurements of radon gas concentration, particle concentration and activity size distributions were performed with and without additional aerosol sources. The conditions of the experiments were as follow: radon gas concentration varied from about 2500 Bq m<sup>-3</sup> to 3500 Bq m<sup>-3</sup>, and the particle concentration from 8,000 cm<sup>-3</sup> to 140,000 cm<sup>-3</sup>. The results of the measurements generally have shown a very good agreement between two instruments. However, the Clarkson University ASC-GSA yielded activity values from 1% up to 20% higher than the diffusion battery from EML. Despite this minor discrepancy, the tests performed proved the viability of the GSA systems for activity weighted size distribution measurements.

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#### INTRODUCTION

Exposure to radon (222Rn) and radon decay products inside homes is now recognized as the main source of radiation doses to the general public and in some situations may present a significant health risk. Two approaches to the estimation of the health risk coefficient from indoor radon have been used. The epidemiological approach derives the risk factor from studies of the incidence of desease in an exposed population (1,2). Alternatively, the dosimetric approach is based on the calculations of radiation doses from physical and biological models and from that dose develops the risk factor (3,4,5,6,7). The second approach requires detail knowledge of the physico-chemical properties of radon and its progeny. Two parameters in early lung dosimetry models used to estimate radiation doses from inhaled radon decay products (3,4,5,6), were the activity median diameter of the "attached" radioactive aerosol and the "unattached" fraction of 218Po. Traditionally defined, the "unattached" fraction constitutes free molecular daughter atoms or ions possibly clustered with other molecules such as  $H_2O$ , typically within the size range of 0.5 to 3-5 nm, as distinct from daughter atoms "attached" to particles in the pre-existing ambient aerosol.

Recently, James modeled the relationship between monodisperse size of radon decay products and dose per unit exposure (7), instead of dose conversion factor or the "attached" or "unattached" fractions only. The resulting dose conversion factors per unit exposure from monodisperse activity are presented in Figure 1.





This development stressed the importance of measurements of the radioactive aerosol size distribution spectrum, especially in the range 0.5 nm up to 1000 nm.

The measurements of the "unattached" fraction of <sup>218</sup>Po and of the potential alpha energy concentration (PAEC) or in more general terms the size distribution of the particles carrying <sup>218</sup>Po, <sup>214</sup>Pb or <sup>214</sup>Bi has been the subject of extensive research. A critical review of such measurements was provided by Hopke (8). The majority of the techniques utilized to obtain the size distributions are based on the diffusional properties of radon progeny and use of the diffusion battery-type systems or the multiple wire-screens systems.

To assure the quality of the size distribution measurements, intercomaprison exercises have been conducted involving different laboratories (9). The reported tests were carried out in laboratory settings. In August 1990 two systems, one from the Environmental Measurements Laboratory (EML) and the second from Clarkson University (CU) were intercompared during series of tests in an unoccupied house in Princeton, NJ. It was, to the best knowledge of the authors, the first intercomparison done in the real house environment.

## ACTIVITY-WEIGHTED SIZE DISTRIBUTION

A major consideration in evaluating the radiological health risk from indoor radon is the size spectrum of particles carrying the radon decay products, described by the term "activity-weighted size distribution".

The size distribution of particles may vary slightly for each decay product due to coagulation or size-dependent loss of the aerosol between the time of attachment and the time of decay. Therefore, for direct measurement of the activity-weighted size distribution, the sampling system should be able to segregate particles according to size, so that the activity of each size fraction can be determined separately. Several factors make this a difficult task, especially in the domestic environment: the activities, usually low, become lower yet when segregating the particles into several groups; the activities are shortlived, dictating short sampling times. Owing to these difficulties, the direct measurement of the activity-weighted size distribution is a relatively recent development.

Two methods for size segregations of particles according to size are generally used: diffusion batteries (DB), and graded screen arrays (GSA). Two methods are also commonly utilized for estimation activity concentrations of radon decay products : gross-alpha counting and alphaspectroscopy.

The diffusion battery generally consists of number of channels, typically of cylindrical shape. The deposition of aerosols in the channels of the diffusion battery is determined by its dimensions, the sampling flow rate, and the diffusion coefficient of the diffusing species. The theorey for diffusional deposition in circular tubes is presented in several papers (10-12). Diffusion batteries have been widely employed for aerosol size distribution measurements. The theoretical and operational aspects of deposition in different types of diffusion bateries have been investigated by many workers (13, 14).

The use of the wire screens to segregate particles in radon research was developed by James et al. (15), Thomas and Hinchliffe (16) and George (17). The degree of penetration, P = (1 - fractional deposition), through wire screen is dependent upon the particle size, wire screen parameters and sampling face velocity. For  $D_p <100$  nm, the dominant wire screen collection mechanism is Brownian diffusion. For larger particle size ( $D_p > 500$  nm), collection by interception and inertial impaction becomes predominant. The theory describing particle penetration through a wire screen was developed by Cheng and Yeh (18) and Cheng et al. (19).

The Graded Screen Array (GSA) technique is a development of more recent years (20). GSA systems consist of varying mesh number, single/multiple wire screen stages operated either in series or in parallel, with a choice of a wide range of wire screen parameters and sampling flow rates. The parameter that is used to describe a GSA stage is the particle diameter that leads to 50% penetration through the stage,  $D_p(50\%)$ . A GSA system may consist of two distinctly different configurations of individual wire screens that may be defined as "serial" or "parallel" (21). The "serial" system consists of a number of individual wire screens. In contrast, the "parallel" configuration GSA system consists of a number of individual GSA stages operated in parallel, in which each stage containing a specific set of wire screens.

To measure the activity concentration, gross-alpha counting and alpha spectrometry techniques are used. Gross alpha counting is based on counting of all of the alpha particles coming from the source (usually a filter) in several different time intervals. A minimum of three count periods are required (22) to solve the set of equations yielding concentration of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi. Raabe and Wrenn (23) proposed a technique in which more count intervals than unknowns are used. A least-squares fit to the count rate versus time provides activities of radon decay products.

Alpha-spectroscopy method was first reported by Martz et al. (24). The counts from the radon decay products distinguished on the basis of their energy with a multichannel analyzer are accumulated in two time intervals. Tremblay et al. (25) optimized the count-interval timing for the case in which the counting and sampling intervals overlap. The counting while sampling technique greatly improved the measurement precision especially with regards to <sup>218</sup>Po with its half life of only 3.1 min.

These methods of sizing of the active aerosols and alpha counting were used in the intercompared systems.

#### HOUSE CHARACTERISTICS

The intercomparison measurements were performed in a basement of a one-story residence with a living room, dining room, kitchen, study, two bedrooms and two bathrooms on ground floor. The outer basement dimensions were as follow: 11.3 m x 10.5 m x 2.5 m and the volume about  $250 \text{ m}^{-3}$ . The schematic plan of the basement with location of the measuring devices is presented in Figure 2. The house was instrumented by the Center for Energy and Environmental Studies, Princeton University for continuous measurements of the radon concentration, temperature, humidity and differential pressure. The house is equipped with sub-slab ventilation system which was turned off during the measurements. The radon concentration in the basement during tests varied between 2600 Bq  $m^{-3}$  (70 pCi L<sup>-1</sup>) and 3700 Bq m<sup>-3</sup> (100 pCi L<sup>-1</sup>). The particle concentration without additional sources was about  $8000 \text{ cm}^{-3}$  as measured with a Rich 100 condensation nuclei counter, and up to 140,000 cm<sup>-3</sup> with additional sources (cigarette smoke, a 1320 watt electric space heater and handyman's propane torch).





### APPARATUS AND PROCEDURE

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The measurements of the size distribution of particles carrying radon decay products were performed with two systems: EML Disk-type Diffusion Battery (DDB) and Clarkson University Graded Screen Array (ASC-GSA). Activity-weighted size distribution measurements were complemented with continues radon gas concentration measurements (EBERLINE, Radon Gas Monitor RGM-3), continues PAEC measurements (EBERLINE, WL-meter) and the aerosol concentrations (Rich 100 CNC).

## EML Disk-Type Diffusion Battery

The EML Disk-type Diffusion Battery (DDB) has a 19 year history of use, thereby providing a link between past and current measurements. The apparatus consists of one open face filter and four filters preceded by diffusion elements (26). Each element consists of a number of perforated stainless steel discs (27) arranged in a series in a tube. Each disc contains 14,500 collimated holes (tubes), 0.0228 cm in diameter. The characteristics of the four batteries are listed in Table 1.

Battery	Number of Sections	Actual Total Lenght of Sections [cm]	Effective Lenght of Battery [cm]	Dp <sub>50</sub> [nm]
1	1	0.34	5.0 x10 <sup>3</sup>	11
2	2	0.95	1.37x10 <sup>4</sup>	21
3	5	8.08	1.17x10 <sup>5</sup>	83
4	10	25.75	3.73x10 <sup>5</sup>	212

TABLE 1. CHARACTERIS	TICS O	F DIFFUSION	BATTERIES
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A sampling time of 10 minutes, and a flow of 3.0 Lpm through each of the four sections of DDB, and through an open face filter is sufficient to achieve good counting statistics. These values were used during tests. Millipore Type AA membrane filters were used (Glass fiber filters may have been more appropriate; see discussion below). After sampling the filters were manually transferred to five identical drawer-type counters (EML Type TH-29 B (28)). The counts were accumulated in a pair of CTM-05 five-channel scalers (Metrabyte Corp., Taunton, MA) plugged into backplane slots of a portable personal computer. A Pascal program was used to supervise counting and to record data in one minute increments. Normally, 40 minutes of count data were collected in each experiment. The one-minute count data were analyzed using the weighted least squares procedure by Raabe and Wrenn (23) yielding the activity concentration of the  $^{218}$ Po,  $^{214}$ Pb and  $^{214}$ Bi on each filter.

Once the EML data has been assembled, it became obvious that many of the <sup>218</sup>Po results obtained from counting the filters were not reliable. This problem was easy to determine because the amount of activity found on the five DDB filters were sometimes badly out of sequence. (They should be in a monotonic decreasing sequence). The probable reason for these erratic results was plateout of <sup>218</sup>Po onto the Millipore filters during transfer from the filter holders to the counters. Glass fiber filters, or other media not subject to substantial electrostatic charging, should be used when radon concentrations are as high as they were in these experiments.

To ensure that the data reported here are reliable, the following data acceptance criteria were adopted for the EML data:

1) the  $^{218}$ Po data were used only as an indicator of data quality; 2)  $^{214}$ Pb,  $^{214}$ Bi and PAEC were accepted only for those tests for

which the <sup>218</sup>Po were in reasonable sequence.

To "unfold" the data, yielding activity-weighted size distributions the Expectation-Maximization (EM) algorithm (29) was used. EML used 16 midpoint values for "unfolded" activity-weighted size distributions: 1.00 nm, 1.58 nm, 2.51 nm, 3.98 nm, 6.31 nm, 10.00 nm, 15.85 nm, 25.12 nm, 39.81 nm, 63.10 nm, 100.00 nm, 158.49 nm, 251.19 nm, 398.11 nm, 630.96 nm and 1000.00 nm. The EML DDB system is meant for research, so experienced operators are required to collect and interpret the data.

#### Clarkson University Graded Screen Array

The ASC-GSA system used by the Clarkson University group involves the use of 6 compact sampler-detector units operated in parallel (30, 31). Each sampler-detector unit couple wire screen penetration, filter collection and activity detection in a way as to minimize depositional losses while being sufficiently rugged for field operations. The system samples air of the rate of about 15 Lpm, simultaneously in all of the units through the sampler slit between the alpha surface barrier detector (ORTEC Model DIAD II, 450 mm<sup>2</sup>) and filter (25 mm Millipore 0.8 m, Type AA) section in each unit. One of the sampler-detector units is operated with an uncovered sampler slit, thus providing information on the total ambient radon decay products concentrations. The sampler slits on the remaining units are covered with single or multiple wire screens of differing wire mesh number. The operating parameters of the system are presented in Table 2.

Unit	Sampler Slit Width [cm]	Sampler Diameter [cm]	Screen Mesh	Dp <sub>50</sub> [nm]
1	0.5	5.3	-	-
2	0.5	5.3	145	1.0
3	0.5	5.3	145x3	3.5
4	0.5	5.3	400x12	13.5
5	1.0	12.5	635x7	40.0
6	1.0	12.5	635x20	98.0

TABLE 2. THE PARAMETERS OF THE SIX SAMPLERS OF THE ASC-GSA SYSTEM

The signals from alpha detectors are connected through amplifiers into an 8-segment multiplexer and routed to a personal computer-based multichannel analyzer. The computer controls acquisition of the alpha spectra, operation of the sampling pump, sample time sequencing and data analysis as well. The sequence of sampling counting and analysis permits automated, semi-continuous operation of the system with a frequency of between 1.5 to 3 hours. The alpha counts from <sup>218</sup>Po and <sup>214</sup>Po detected by each alpha detector in the two counting intervals are used to calculate of the radon decay product concentrations penetrating into each unit (25). The observed concentrations of <sup>218</sup>Po, <sup>214</sup>Pb and <sup>214</sup>Bi are used to reconstruct the corresponding activity-weighted size distributions using the Expectation-Maximization (29) algorithm. The Clarkson University program is design to use six optimized size range bins: 0.5-1.58 nm, 1.58-5.00 nm, 5.00-15.81 nm, 15.81-50.00 nm, 50.00-158.11 nm, 158.11-500.00 nm (32).

The ASC-GSA system was design for field measurements and involves very little attention during operation (31).

#### RESULTS

There were four sets of measurements taken simultaneously with EML and CU systems. Table 3 gives the test conditions for the four test runs.

Run no.	Type of aerosol	Particle concentration [particle.cm <sup>-3</sup> ]	Radon concentration [Bq m <sup>-3</sup> ]
1	background	8000	3110
2	background	8000	3108
3	cig. smoke	24000	3290
4	heater+torch	35000	3220

TABLE 3. TEST CONDITIONS FOR THE SIZE DISTRIBUTION INTERCOMPARISON

To compare the results, both sets of data were brought to a common basis. The EML analyses yield the  $dA/d(logD_p)$  in terms of activities, expressed in Bq m<sup>-3</sup>, whereas the CU analyses yield the results in form of the activity fractions. The CU results were therefore converted to activities  $(dA/dlogD_p)$  and these values were used for direct comparison. Because both devices are equipped with an open face filter for total airborne activity measurements, it was possible to compare the performance of both systems for direct radon decay product concentrations and PAEC measurements. The obtained values are presented in Table 4 a), b), c).

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a)			
	<sup>214</sup> Pb Concentr	ation [Bg m <sup>-3</sup> ]	
Run no.	EML	ĊU	CU/EML Ratio
1	$1309 \pm 20$	$1478 \pm 8$	1.13
2	$1243 \pm 18$	$1418 \pm 8$	1.12
3	$1738 \pm 18$	2063 ±18	1.17
4 ·	$1680 \pm 30$	2002 ±29	1.19
b)			
	<sup>214</sup> Bi Concentr	ation [Bq m <sup>-3</sup> ]	
Run no.	EML	CU	CU/EML Ratio
1	$1088 \pm 34$	$1248 \pm 14$	1.15
2	$1012 \pm 26$	$1023 \pm 5$	1.01
3	$1380 \pm 31$	$1298 \pm 6$	0.94
4	1274 ± 50	1343 ± 4	1.05
c)			
	PAEC [	nJ m <sup>-3</sup> ]	
Run no.	EML	CU	CU/EML Ratio
1	7351 ± 48	8004 ± 41	1.09
2	6883 ± 36	$7402 \pm 40$	1.08
3	9214 ± 44	9983 ± 71	1.08
4	9038 ± 76	9842 ± 68	1.09

## TABLE 4. COMPARISON OF ACTIVITIES OF<sup>214</sup>Pb (a), <sup>214</sup>Bi (b) and PAEC (c) MEASURED DURING INTERCOMPARISON BETWEEN THE ENVIRONMENTAL MEASUREMENTS LABORATORY (EML) AND THE CLARKSON UNIVERSITY (CU).

The right column in Table 4 a), b), c) shows the ratio of values obtained by the Clarkson University group and the EML group. In nearly all cases (except one) the CU values were higher then EML, but the maximum difference was not larger then 20%, and in case of PAEC less then 10%.

The main purpose of the tests was however, to measure the activity-weighted size distributions. The results of four runs presented in Figures 3, 4, 5, and 6 are the size distributions of PAEC, <sup>214</sup>Pb and <sup>214</sup>Bi measured by EML and CU. Figures 3 and 4 present the results of measurements performed in low acrosol concentration conditions (no additional sources) and Figures 5 and 6 present the high aerosol concentration samples. For the low (background) aerosol case (Runs 1 and 2) the EML spectra generally showed a mode near 100 nm, whereas the CU spectra generally showed the mode in the largest size class. This difference is fairly minor, and reflects the differing

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Figure 3. Comparison of the Clarkson University and the Environmental Measurements Laboratory results of the activity weighted size distribution measurements in a real house environment: Run no. 1 (09-22-90, 11:00)



Figure 4. Comparison of the Clarkson University and the Environmental Measurements Laboratory results of the activity weighted size distribution measurements in a real house environment: Run no. 2 (09-22-90, 12:00)

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Figure 5. Comparison of the Clarkson University and the Environmental Measurements Laboratory results of the activity weighted size distribution measurements in a real house environment: Run no. 3 (09-22-90, 16:00). Aerosol: smoke from two cigarettes, aged 45 minutes.



Figure 6. Comparison of the Clarkson University and the Environmental Measurements Laboratory results of the activity weighted size distribution measurements in a real house environment: Run no. 4 (09-23-90, 10:00) Aerosol: fumes from a 1320 watt electric space heater plus a handyman's propane torch, aged 45 minutes

capabilities of the two devices at the large end of the size spectrum. A similar behavior was observed for the cigarette smoke aerosol (Run 3). For the torch+heater aerosol (Run 4), the two methods agreed very well as to location of the peak near 50 nm. However, there is a substantial difference between the two methods regarding the amount of activity in the 2 to 10 nm range. Most EML curves show virtually zero activity in this range, while the CU results show a small but finite amount. In addition, the EML results generally show a sharper upturn at the smallparticle end of the size range, than do the CU results. The EML results correspond to the classical idea of a well defined, uniformly sized, "unattached" fraction. The difference discussed above could result in part from a difference of assumptions concerning the smallest possible particle size. The bottom class in the EML calculation is 0.79 to 1.25 nm, compared to 0.5 to 1.58 nm for the CU calculation. This subject is a matter of further study.

As a practical, single number index of differences between EML and CU results, the dose to bronchial secretory-cells nuclei per unit exposure for adult male was calculated, based on recent James model (7). The results are presented in Table 5.

CALCOLATED	TOR TREO	DIZE DID	INIDO	110110	ODIAINED	DI	THE	LIIL	AND	INE	00.	
		Secretory	Cell	Dose	[Gy/J m <sup>-3</sup>	h]						-
Run no		EMI.			CU			CU	/EMI	Rat	io	

21.65

24.93

13.60

28.41

1.62

1.78

2.17

1.80

TABLE 5. DOSE TO SECRETORY-CELL NUCLEI FOR ADULT MALE ( $B = 3 \text{ m}^{-3} \text{ hr}^{-1}$ ) CALCULATED FOR PAEC SIZE DISTRIBUTIONS OBTAINED BY THE EML AND THE CU.

The dose estimates by using data from both systems did not agree very well. This difference was probably due to the much sharper peak at about 1 nm in the EML system and no activity in the 2 to 10 nm range. Since the peaks in the dose per unit exposure curves shown in Figure 1 occur around 1.5 nm, small difference in the amount of activity in this size range can make significant difference in the estimated dose.

However, the overall performance of both systems was good and the observed differences were within the expected variability of two different techniques that have their own internal errors in addition to the statistical counting errors.

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13.39

14.02

6.27

15.75

;

1

2

3

4

#### CONCLUSION

The performed test in a real house environment proved that the reliable measurements of the activity-weighted size distributions of radon decay products could be made. The results obtained with two systems, which utilize different particle segregation and alpha counting methods, have shown very good agreement. The agreement between the manual diffusion battery and the automated graded screen array system indicates that the GSA does provide useful activity size information with simplicity in its use. Automated systems of this type could be a good source of reliable information of sizes of active aerosols. That are important for proper dose estimates from inhalation of radon decay products.

#### ACKNOWLEDGEMENT:

The work at Clarkson University was funded in part by the New Jersey Department of Environmental Protection under contract No. P32108 and the U.S. Department of Energy under Grant No. DE-FG02-90ER61029.

The work described in this paper was not funded by the U.S. Environmental Protection Agency and therefore the contents do not necessarily reflect the views of the agency and no official endorsement should be inferred.

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