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TIME INTEGRATED MEASUREMENTS OF THE ACTIVITY-WEIGHTED SIZE DISTRIBUTION OF RADON PROGENY

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The dose to the lung is dependent on the size distribution as well as the concentration of radon progeny. The principal indicator of particle size is the diffusion coefficient. The diffusion coefficient of radon progeny is the most important parameter in determining whether the particle will deposit its alpha energy in the tracheobronchial region of the respiratory tract. Due to the importance of the size distribution of progeny when characterizing health effects, a detector was developed that exploits the difference in diffusion coefficients to provide integrated measurements of the activity-weighted size distribution.

Several radon chamber tests were performed in which the diffusion coefficient and unattached fraction of radon progeny were varied by the use of trace gasses and aerosol particles. The technical feasibility of this new detector design was demonstrated in all cases.

Key Words: Radon Progeny, Activity Weighted Size Distribution, Time Integrated, Diffusion Coefficient

Introduction

RAD-X Ltd. is developing a radon progeny detector capable of measuring the time-integrated activity-weighted size distribution of radon progeny (Tetley et al. 1989). The work described in this paper was to determine the feasibility of this detector design. The work done demonstrated that the detector performs extremely well. This paper also includes our plans for future work.

Detector Design and Analysis

The configuration for the detector is shown in Fig. 1. The detector is composed of two stages and operates as follows:

-The air (with radon progeny) is drawn through both stages by a low volume (1 LPM) air pump.

-In the first stage, a parallel plate diffusion battery, the ultrafine progeny plates out (the smaller the particle the closer to the entrance the particle plates out).

-One of the plates in the diffusion battery is made of CR-39. The decay of the progeny (^{218}Po and ^{214}Po) which plates out in the diffusion battery is recorded in the CR-39.

-Analysis of the CR-39 yields the activity weighted size distribution.

-After leaving the diffusion battery the progeny is drawn through the second stage. Here a piece of filter paper traps the remaining (attached) progeny.

-A piece of CR-39 opposite the filter paper records the decay of the attached progeny.

-Analysis of the CR-39 yields the amount of attached progeny.

To analyze the CR-39 it is etched in 6.5 Molar NaOH for 5 hours at 70° C. This etching enlarges the damage tracks left by the alpha particles. The damage tracks are then visible under a 400 power microscope.

The size distribution of the ultrafine particles is determined by analyzing the track density versus distance from the opening. Qualitatively the larger the diffusion coefficient the closer to the entrance the progeny plates out. The penetration probability for a parallel plate diffusion battery can be approximated by (Soderholm 1979):

$$P = .0.91035 \exp(-1.88517\mu)$$

where $\mu = (2)WLD/FH$

W = the width of the plates (cm)
L = the length of the battery (cm)
D = the diffusion coefficient (cm^2/sec)
F = the volume flow rate (cm^3/sec)
2H = the gap between plates (cm).

From this equation the track density versus distance from the opening is, for a monodisperse aerosol, an exponentially decaying curve. The e-folding factor is proportional to the diffusion coefficient. This is most easily analyzed by plotting the natural log of the track density versus the distance from the opening. In the monodisperse case the curve is a straight line, whose slope is related to the diffusion coefficient. The total amount of progeny with that diffusion coefficient is calculated from the y-intercept and the slope.

The case of a polydisperse ultrafine progeny distribution is more difficult to analyze. For a distribution with two distinct components each with a different diffusion coefficient, the plot is made of two line segments with the slope of each related to the diffusion coefficient of one of the components.

The parallel plate design has the potential to provide extremely detailed information regarding the activity-weighted size distribution. In a graded screen array the number of size groups is limited by the number of screens (typically 3). In a parallel plate design the number of size groups is limited only by the amount of analysis performed. This offers the researcher a large amount of flexibility. Also the number and mean size of the size groups need not be chosen prior to the run. Thus the size distribution information can be said to be in an "analog" form.

Prototype Testing

To test the above design a series of tests were run. These tests were designed to determine the ability of the detector to measure both the amount and the size distribution of ultrafine progeny. The tests can be broken down into to classes. In the first, a trace gas (SO_2) was used to alter the ultrafine size distribution. In the second, the concentration of background aerosols was varied. This was done to vary the amount of ultrafine progeny relative to attached progeny present.

The first set of tests were run at Clarkson University with the assistance of Dr. Philip Hopke. The radon chamber used has excellent control over conditions. The chamber is kept clean of all normal background aerosols (< 25 particles/cm^3). The radon level was maintained at 400 pCi/l. The progeny levels varied depending on the trace gasses. Three runs were made. In these runs the SO_2 level was varied. The levels were 0 ppm (Clean Dry Air), 5 ppm and 15 ppm. In the SO_2 runs the relative humidity was about 30%.

In the Clean Dry Air runs the progeny was a monodisperse aerosol with a diffusion coefficient of .054 cm_2/sec. The detector was able to accurately measure (± 5%) the diffusion coefficient. The amount of progeny detected was found to be a function of the gap size. This is due to plate out prior to entrance into the detector. For a given gap size the progeny detected was found to be consistent within ± 5%. For a gap size of .04 cm, the fraction of progeny which is detected is about 50%.

The introduction of SO_2 leads to the formation of a growth mode with particle sizes ranging from .8 to > 20 nm. This is a result of radiolysis of the water and subsequent interaction with the SO_2 to form H_2SO_4. The detector clearly showed this growth mode. A typical plot of the ln of track densities versus distance from the opening is shown in Fig. 2. This figure shows the presence of a bimodal ultrafine particle distribution seen as a change in the slope of the line. The first mode has a diffusion coefficient of about .05 cm^2/sec and accounts for about 35% of the progeny. The second mode has a particle size of about 3.75 nm and accounts for about 17% of the progeny. In addition to the ultrafine modes there is a fraction which has a much larger particle size (>10 nm). This larger mode progeny passes through the diffusion battery and is captured on the filter paper. This mode accounts for the remainder of the progeny (about 48%). The consistency between detectors in a given run was very good (± 10%).

The second set of tests was run at the Environmental Measurements Laboratory with the assistance of Dr. Andy George. In these tests the aerosol concentration was varied. Two runs were made, one with aerosols concentrations < 10,000 particles/cm^3 and one with concentrations > 50,000 particles/cm^3. This varied the total ultrafine fraction (the higher the aerosol concentration the lower the ultrafine fraction). In these tests no trace gasses were used, so the ultrafine progeny was monodisperse. The detector demonstrated the capability of distinguishing between high and low ultrafine fractions. Unfortunately no independent measurements were available to confirm the performance. However, the results were consistent with calculations relating ultrafine fractions to aerosol concentrations.

Additional tests were run to determine the feasibility of separating the two alphas (^{218}Po and ^{214}Po). This can be done by separating the alphas based on the size of the tracks. To do this the alphas are slowed down to 1.0 and 2.7 MeV respectively. This maximizes the size difference. Our testing showed this to be a practical method, although for large scale use an automatic track counting system is required.

Future Work Planned

The present work has demonstrated the feasibility of this detector. Future work will is directed in the following areas:

-Extent the range of ultrafine progeny detected. The present detector is capable of detecting progeny as large as 10 nm. Progeny as large as 20 nm can still have a significant health effect.

-Perform calibration measurements. Monodisperse aerosols in the 5-20 nm range will be used to verify detector penetration curves.

-Improve the size distribution reconstruction analysis routine used. Computerized procedures using the Expectation-Maximization Algorithm are envisioned.

Conclusion

Upon completion, the detector will be ideal for researchers conducting surveys of private homes to reduce the uncertainty of risk relating to radon. In a home with 4 pCi/l of radon the detector will have a run time of 1-3 months. In higher radon concentrations the run time can be reduced accordingly. This flexibility in run times should be very conducive to experiments set up to examine the physical and chemical interactions of radon progeny in air. Experiments could be arranged to determine the effects that cigarette smoke, cooking, gas pilot lights, wood stoves, air cleaners and different ventilation rates have on the activity size distribution for the same concentration of radon gas. Previous research with our detector has demonstrated that the activity-weighted size distribution information that is obtained on ultrafine progeny is "analog" and only limited by the amount of analysis performed on the CR-39.

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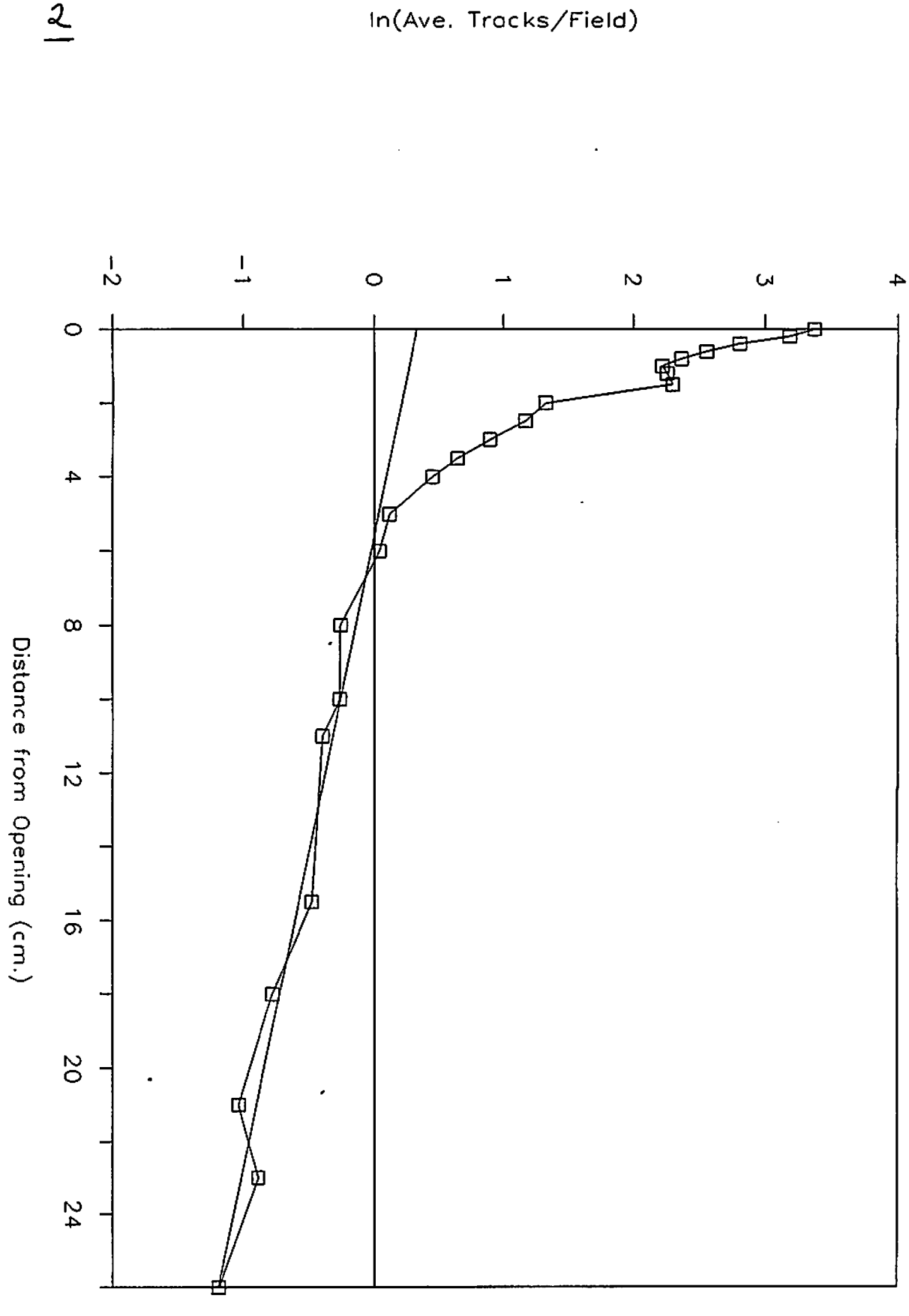
List of Figures

1


ln(Ave. Tracks/Field)
-2
-1
0
1
2
3
4
0
4
8
12
16
20
24
Distance from Opening (cm.)