PASSIVE RADON PROGENY DOSIMETERS: FEASIBILITY STUDIES

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ABSTRACT

Radon progeny measurements can improve dose estimates based on radon gas measurements alone. The airborne activity-size distribution ratio affects the available dose rate per unit radon. Measurements of surface deposited alpha activity and radon concentration can be used in a semi-empirical model to estimate the equilibrium ratio, the free fraction and airborne dose rate. Since residential atmospheres are dynamic, several measurement approaches, including electret ion chamber and track registration techniques, are being studied to develop passive, integrating detectors. Preliminary tests show good correlation between surface deposited activity or energy, airborne progeny concentrations and dose rate. Tests are underway to assess the performance in other home environments.

INTRODUCTION

Inhaled radon progeny deliver more radiation dose to individuals than radon (^{222}Rn) or thoron (^{220}Rn) gas. The airborne dose available in a room depends mainly on the Potential Alpha Energy Concentration (PAEC) of the radon progeny and their activity size distribution. Most indoor radon radiation exposure estimates are based on gas measurements since radon concentrations are fairly well correlated with radon-related dose rate and progeny measurements are more difficult to make with current technology.
The most common parameter used in quantitative estimate of risk from radon is the equilibrium ratio, designated as F. This is the ratio of the actual airborne progeny PAEC to the maximum that could be generated from the actual radon gas concentration. To calculate the risk factors from radon in US houses, F is often taken as 0.4. Unfortunately F can vary widely depending upon aerosol particle concentration surface deposition and ventilation that affect the progeny activity size distribution. Therefore the actual dose rate depends of the fraction of the progeny that is "unattached" to aerosols, designated as the free fraction or f_p and usually assumed to be about 10% in homes.

Changes of indoor atmospheric conditions can introduce variations of a factor of two in the dose rate per unit radon concentration in upper Midwest houses (Steck and Field 2006).

Unfortunately, radon progeny concentrations have been measured and published in only a few US homes. The "typical" equilibrium ratio and free fraction distributions used in radon risk assessment in the US are based on grab samples and short-term monitoring in fewer than 40 homes in the New York and New Jersey (USEPA 1992, James et al 2004) The radon progeny atmosphere is dynamic in most rooms and grab samples are likely to miss significant temporal variations (Hopke et al. 1995). Active radon progeny measurement devices tend to be hard to maintain and bothersome to home occupants.

Applications that require accurate dose or risk assessments would be better served by combined measurements of radon and radon progeny that integrate both. The goal of this work is to develop and test passive detectors that could estimate airborne radon progeny concentrations and dose rates over periods of several days to several months. Our current approach is to measure the average radon concentration and the average surface-deposited radon progeny during extended sampling period with simple, inexpensive technologies like alpha track detectors and electret ion chambers.

METHODS

Semi-empirical indoor progeny fate and transport model

Our first step in detector development was to identify the activities and other variables that we could measure and use to reliably predict airborne progeny and dose. We extended a semi-empirical room fate and transport model to include surface deposition and implantation (Steck and Field 1999). A Monte Carlo engine(Crystal Ball) was used to generate the transport and fate of radon progeny indoors from distributions of room parameters like surface-to-volume ratio, deposition, attachment, and ventilation rates in a rate-balance model shown schematically in Figure 1. The parameter distributions were selected from literature or regional reports. As much as possible, the distributions reflect, regional estimates for room size, house air-tightness, smoking and cooking patterns.

The airborne dose rates from two published dosimetric models were generated from simulations in 20,000 upper Midwest homes. These dosimetric models were both based on the ICRP65 lung model but differ in the relative weighting factor assigned to different-size progeny clusters (Porstendorfer 2001, James et al. 2004). We use a bimodal version of these models with separate weights for the measurable progeny; unattached (<5 nm) and attached. Between them, the models provide realistic bounds for the true dose rate. Correlations were examined between measurable surface deposited progeny and airborne dose rate to try to find a practical measurement for a passive dosimeter.

Figure 2 illustrates this process. It shows the clear correlation between the attached airborne PAEC and the radon concentration. Less obvious is the correlation between the PAEC and the deposited activity. After extensive examination of different possible combinations of surface deposited progeny we found that two sets of measurables proved reliable estimates of the airborne dose rate: (1)the radon concentration plus the ratio of the deposited ^{218}Po to ^{214}Po activity density and (2) the radon concentration plus the surface deposited PAEC.

In 2007, we also tested the fate and transport model predictions for thoron progeny. Given the lack of experimental and theoretical work done on the indoor thoron activity and dose, we currently use a simple EEC model for the thoron progeny dose (UNSCEAR 2000). This model doesn't use weighting for the different activity-weighting size distributions.

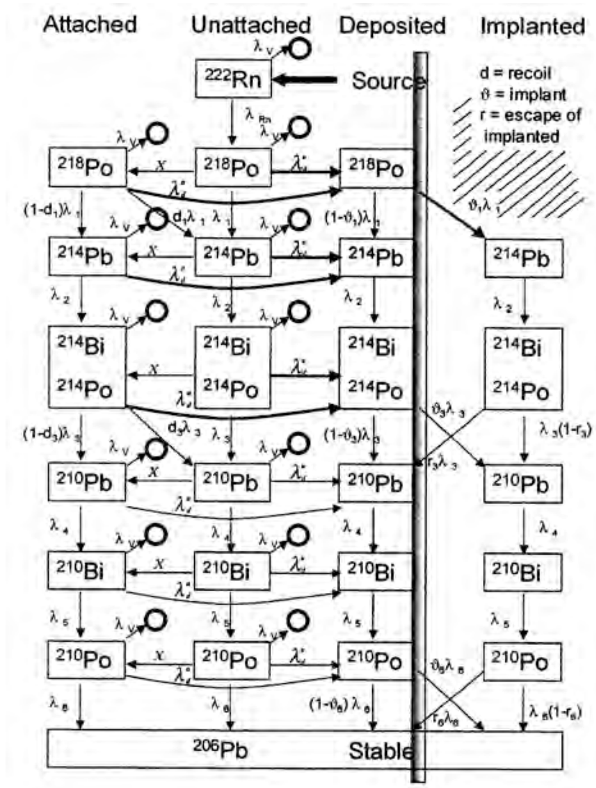

Figure 1: Schematic representation of process and parameters that affect airborne radon progeny activity size concentrations

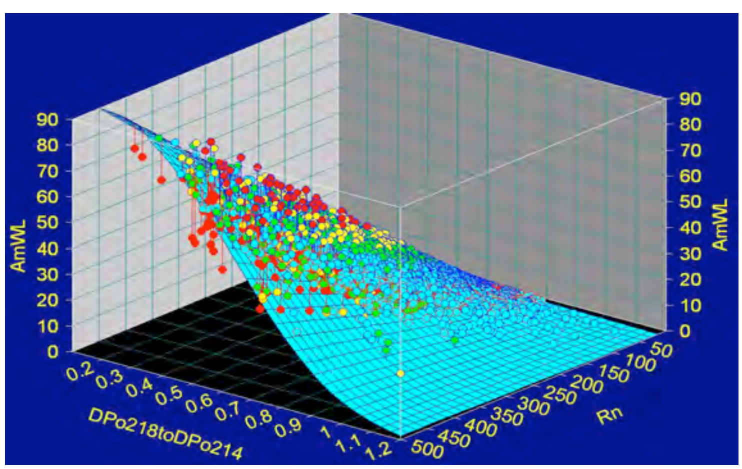

Figure 2. The predicted attached PAEC in 20,000 houses (shown as colored dots) with high aerosol concentrations as a function of radon concentration and the ratio of the deposited ^{218}Po to ^{214}Po.

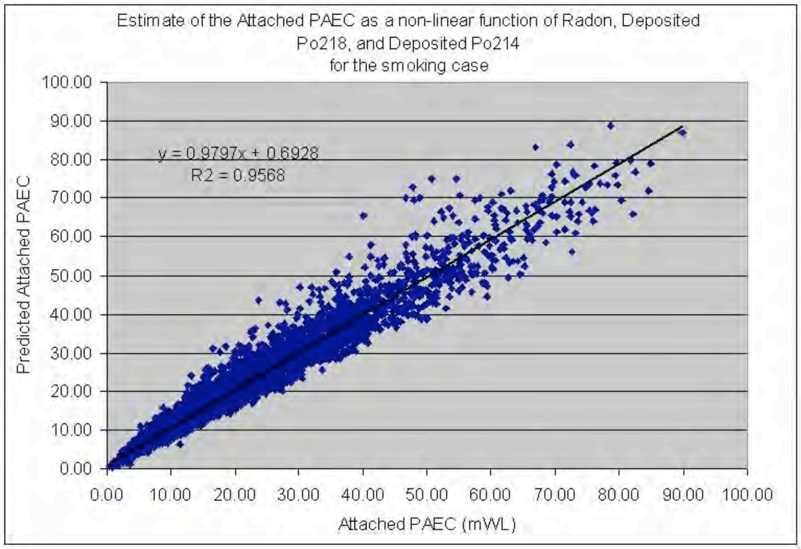

Figure 3 The correlation between the simulation points and the two-variable measurement model predictions at those points. The predictive model is shown as light blue surface in Figure 2.

These model studies led us to develop two types of dosimeters: surface alpha activity track detectors and surface alpha energy electret ion chamber (EIC) detectors.

Surface alpha activity detector

Track registration material responds to alpha particles on or near a surface. The detector's efficiency depends on the energy of the alpha particle. With a suitable set of energy absorbing films between the deposited progeny and the track detector, the activities can be determined for the various alpha-emitting progeny that are on or near the surface. We use three filters to separate the thoron-generated ^{212}Po and radon-generated ^{214}Po and ^{218}Po. These detectors are shown in the top of Figure 4.

Surface alpha energy detector

Electret ion chambers (EIC) respond to the energy lost by the alpha particle in the chamber. Alpha particles from surface deposited progeny can penetrate an EIC with a thin entrance window as shown on the bottom left of Figure 4 (Dua et al 2002). The voltage loss by the electret will depend on the total alpha energy loss in the ion chamber which will depend on the radon and radon progeny in chamber and on the radon progeny and radon on or near the thin entrance window. A companion EIC with a thick plastic lid covering the entrance window is used to correct the thin-window chamber response for the effects of interior radon and progeny.

Figure 4 Track registration detectors (top) and electret ion chambers for measuring surface deposited radon and thoron progeny.

Exposure room and home tests

The theoretical model predictions and the passive detectors were tested in series of exposures in our lab from 2004 to 2007. Exposures took place in a room with dimensions and furnishing similar to a small bedroom. Radon in the room can be varied from 2 to 100 pCi/L with air exchange rates from 0.2 to 2 ach. Most exposures took place with radon varying from 40 to 70 pCi/L and 0.2 ach. A Durridge RAD-7 measured the hourly average radon concentration. Attached and unattached airborne radon progeny concentrations were measured by repeated grab sampling. In 2005-2006, a SARAD EQF3120 ran continuously on a two-hour cycle during exposures. In 2007 the EQF was unavailable so repeated grab samples were taken every few hours using a screen-filter system. This 20007 system's results for the equilibrium ratio (F) and free fraction (f_p) agreed with the values measured simultaneously with an EIC-based integrating progeny sampler (ERIPSU) and past EQF measurements under similar atmospheric conditions.

Surface progeny activities were sampled at the same rate as the airborne progeny. Each sample set consisted of three to six glass pieces that were removed from their holders on the exposure room wall. Large-area semiconductor diode detectors were used for spectroscopic analysis of the glasses which began immediately upon their removal. For radon progeny, a two interval counting protocol that lasted one hour determined the three short-lived progeny activity densities.

Aerosols were generated using common household sources like tobacco smoke, candles, propane, air fresheners, vacuum cleaners, and humidifiers. Aerosols could be removed by increasing deposition with fans or commercial air cleaners using HEPA and/or ion generation. From 2005 to 2006, exposures were conducted with the aerosol sources generating continuously. In the 2007 the sources were run intermittently to better simulate the activity that might occur in a real home. Aerosol concentrations were measured with a condensation nuclei counter during the 2007 exposures.

Thoron exposures for each atmosphere took place over at least four days to allow equilibrium to occur. Thoron concentrations measured with a modified Durridge RAD-7 ranged from about 10 to 15pCi/L while the radon was less than 1 pCi/L. Surface ^{212}Po activity density was measured with an hour-long spectroscopic count. Airborne ^{212}Pb and ^{212}Bi^{-}Po concentrations were measured with a screen-filter scintillation grab sampling system and a 5-hour counting protocol. (Khan et al., 1982)

The surface alpha energy system was tested in the homes of two of the authors (DS and PK). Field tests of the surface activity devices are underway in 40 Iowa homes and 20 Pennsylvania homes.

RESULTS AND DISCUSSION

Our fate and transport model suggests that the integrated average radon concentration and the deposited radon progeny on surfaces can predict dose rates over a wide range of indoor environments. From these measurables, which depend on F and f_p, we can form a proxy F. Theoretically, the proxy F is expected to increase with lowering of F. Lower F is usually the result of lower particle size distribution and number concentration. Under such situation, the diffusion coefficient of particles increases leading to relatively higher deposition on the surface measured. The aim of the present work is to determine the correlation between proxy F, F and f_p. If s an acceptable correlation exists, the dose rate or F can be calculated from proxy F. Since proxy F is measurable over extended periods, days to months, the calculated F or dose rate will be applicable for the entire period.

Exposure room results for deposited activity and alpha energy

Summaries of airborne and deposited radon and thoron progeny activity measurements made in 2007 are shown in Figures 5, 6, 7. Results from earlier experimental runs are similar. Each cluster of data represents the average of repeated measurements over two to four day runs. The horizontal axis reports the environmental conditions, aerosol source and sinks (if any). The progeny activities and dose rates are normalized using the average radon concentration during the run so that different runs can be compared.

Figure 5 shows that deposition is enhanced by air movement and aerosol reduction. A closer look shows that the ratio of the two deposited alpha emitting progeny also changes with different atmospheric conditions. Changes in the two dosimetric models are significant but less dramatic than the activity variation. The nanoparticle aerosol concentrations (Z; units particles cm^{-3}) are shown as grey squares and their logarithms are plotted using the secondary (right) vertical axis.

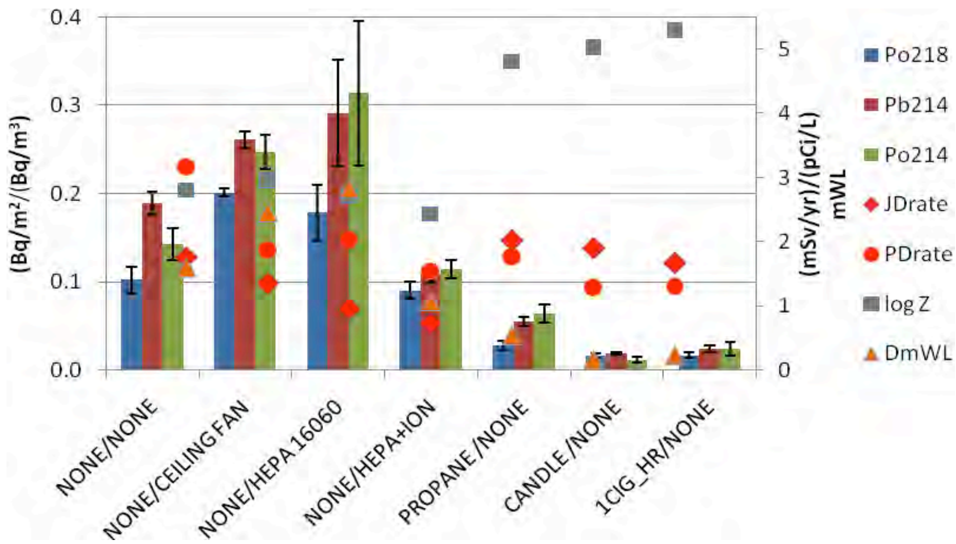

Figure 5. Deposited progeny activities per unit radon concentration are plotted as bars using the primary vertical scale. The airborne normalized dose rates (James and Porstendorfer), the log of the aerosol concentration (Z) and the deposited alpha energy (mWL) are plotted with symbols using the secondary vertical axis. The horizontal axis is labeled with conditions in the exposure room; the first label indicates the aerosol source while the other shows the aerosol removal mechanism.

The airborne progeny concentrations shown in Figure 6 also show dramatic differences as atmospheric conditions are varied. As expected higher aerosol conditions increase airborne progeny activity. But since the progeny are attached to aerosols they have lower dose effectiveness and hence the dose rate variation is smaller that the activity variation.

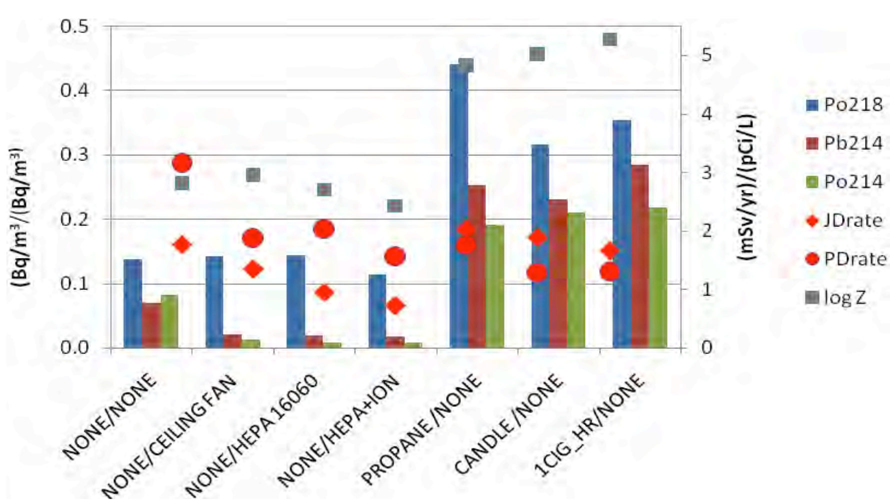

Figure 6. Airborne progeny activities per unit radon concentration are plotted as bars using the primary vertical scale. The airborne normalized dose rates (James and Porstendorfer) and the log of the aerosol concentration (Z) are plotted with symbols using the secondary vertical axis.

Thoron progeny

The long-lived thoron progeny have more opportunity to be removed from the room air by ventilation and deposition than the immediate radon progeny. The normalized airborne and deposited thoron progeny concentrations shown in Figure 7 illustrate this. A comparison of the normalized, deposited progeny in Figure 5 with those in Figure 7 shows that in low aerosol cases, the activities are similar. But in high aerosol concentrations, relatively more ^{212}Po than ^{214}Po is deposited.

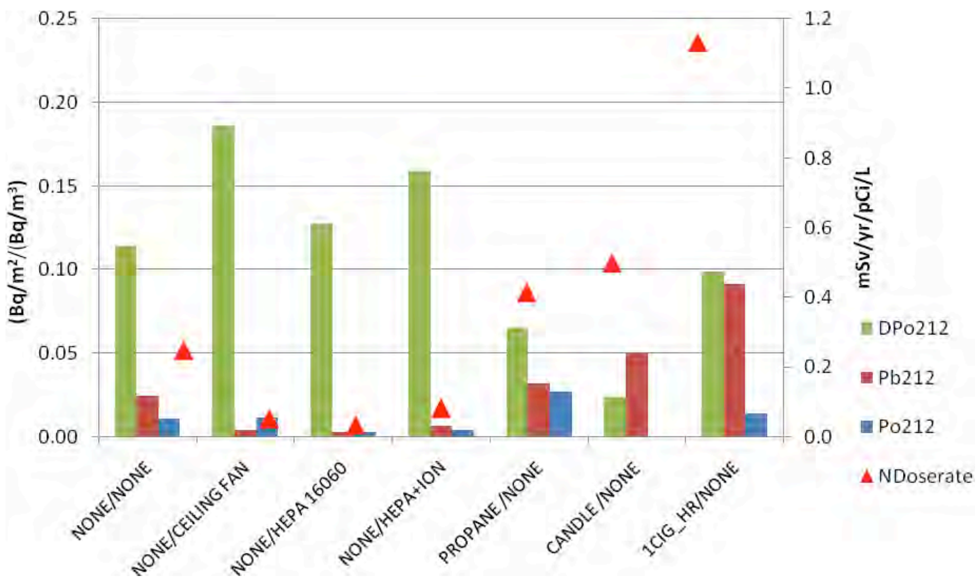

Figure 7 Deposited ^{212}Po and airborne ^{212}Pb and ^{212}Po. Normalized dose rate plotted using secondary vertical axis.

Dose model predictions in exposure room

We tested the reliability of our fate-and-transport model, to predict the measured airborne progeny equilibrium ratio, free fraction and dose rate in our exposure room runs. The reliability of the prediction improved if we treat cases with high aerosol concentrations and low aerosol rooms separately. Figures 8 and 9 show that, in the 2005 experimental runs, reasonably good correlation exists between the predicted and measured dose rates.

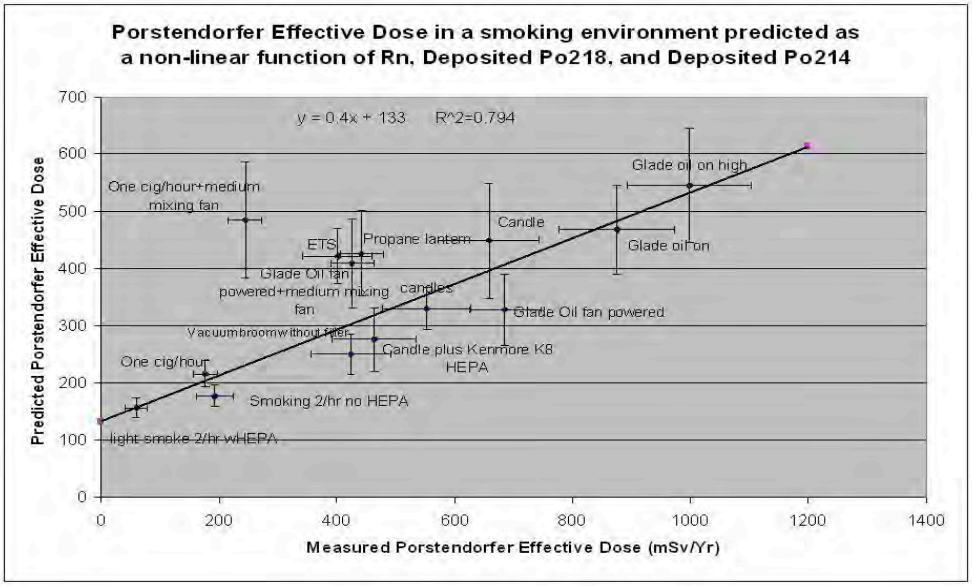

Figure 8 The dose rate predicted from measured radon and deposited progeny concentrations compared to the dose rate based on direct sampling of the airborne progeny concentrations for various high aerosol conditions in the exposure room.

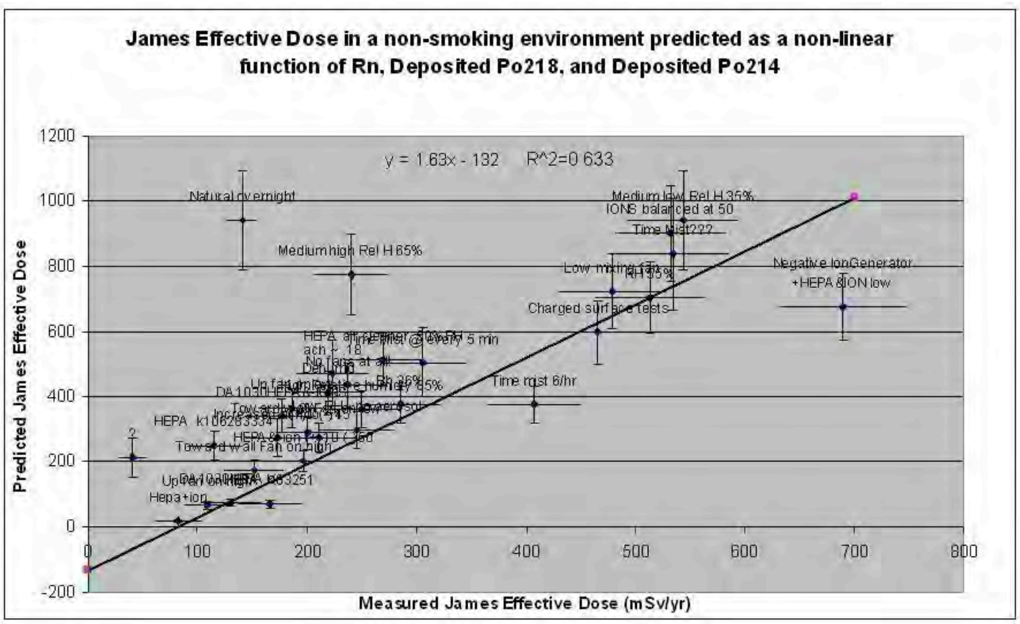

Figure 9. The dose rate predicted from measured radon and deposited progeny concentrations compared to the dose rate based on direct sampling of the airborne progeny concentrations for various low and medium aerosol conditions in the exposure room.

Surface energy dosimeter performance in the exposure room and two houses

The performance of the surface alpha energy detector is shown in Figures 10 and 11.

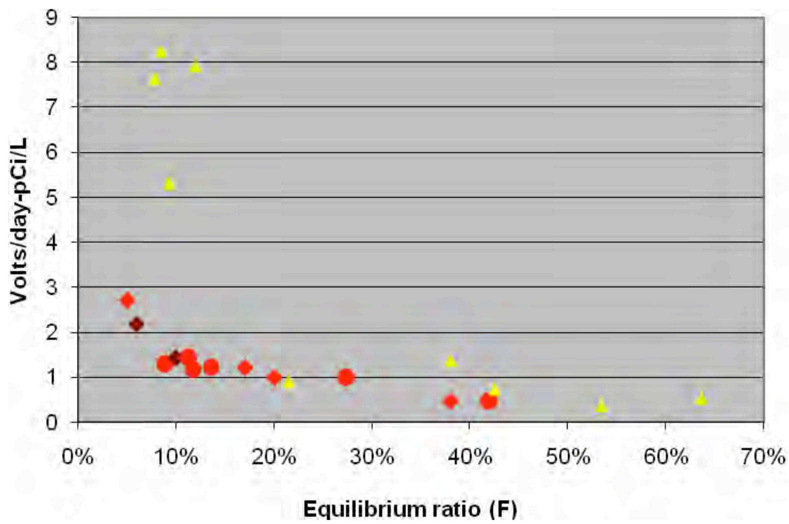

Figure 10. The EIC voltage loss rate in different equilibrium ratio atmospheres. The yellow data points were measurements in one author's house (PK). The bright red symbols were data taken in the exposure room while the darker red diamonds were taken in another author's (DS) house.

The electret voltage loss rate per unit radon concentration for equilibrium ratios above 10% follows a consistent pattern for measurements made in the exposure room and two houses. The disagreement below 10% may result from an increased deposition rate from high free fractions and enhanced air turbulence in PK's house when the HEPA air cleaner was used to achieve the lower F.

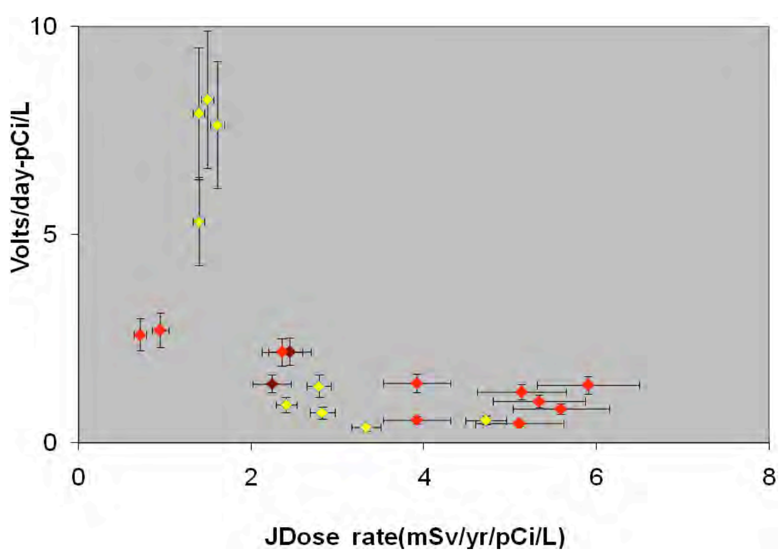

Figure 11. The dose rate and EIC voltage loss rate in different atmospheres. The yellow data points were measurements in one author's house (PK). The bright red symbols were data taken in the exposure room while the darker red diamonds were taken in another author's (DS) house.

The same pattern of agreement is evident in the dose rate.

These limited numbers of measurements appear to correlate our proxy F with measurable F and dose rate. But it is necessary to test the correlation for different types of environments in homes or laboratories where precisely characterized progeny concentration is available. These tests will help quantify the accuracy and precision of the surface activity and energy proxy methods and identify the conditions that limit the methods.

CONCLUSIONS

Our work shows that passive measurements of radon concentrations and surface deposited alpha activity or energy can be used with a semi-empirical model to reliably estimate the equilibrium ratio, free fraction and airborne dose rates in a variety of controlled environments. Field tests in homes are underway to improve and validate the method.

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