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THE IOWA RADON LUNG CANCER STUDY: CONTEMPORARY AND HISTORICAL AIRBORNE RADON (^{222}Rn) AND RADON PROGENY CONCENTRATIONS

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The use of contemporary radon (^{222}Rn) gas concentrations to estimate retrospective radon-related doses can introduce substantial uncertainties in epidemiological analyses. These uncertainties tend to bias the results of radon-lung cancer epidemiologic studies towards the null. Temporal variability of radon progeny over past decades and the variability in the dose effectiveness of airborne radon progeny caused by indoor atmospheric differences are among the main sources of uncertainties in our region. Studies of glass exposed in radon chambers and in homes show that radon progeny deposited on, and implanted in, glass hold promise for reconstructing past radon and radon progeny concentrations in a variety of atmospheres.

We developed an inexpensive track registration detector for the Iowa Radon Lung Cancer Study that simultaneously measures contemporary airborne radon concentrations, surface deposited alpha activity density, and implanted ^{210}Po activity density. The contemporary surface deposited activities are used to estimate the contemporary airborne radon progeny concentrations. They are also used to interpret the implanted activity ^{210}Po as a retrospective measure of the cumulative radon and radon progeny exposure.

Over two thousand retrospective reconstruction detectors were placed in more than 1000 homes for a one-year exposure period. A preliminary analysis of approximately 1500 of these detectors showed that: the detectors performed well in the field with >95% retrieval after 1 year; the detectors met accuracy and precision goals (10%); that there is good correlation (r^2~0.5) between the total radon exposure estimated from contemporary annual-average radon gas measurements and the reconstructed historical average radon concentrations determined retrospectively from implanted ^{210}Po activity; and that the use of the contemporary implanted activity improves the correlation between the retrospectively reconstructed and contemporary radon concentration by about 50% . Work is underway to test the reconstruction in homes with measured radon histories and to compare our methodology with other groups.

Key words: radon, radon progeny, retrospective, dose, glass, surface activity, implanted activity, lung cancer, epidemiology.

INTRODUCTION

The possible health consequences of exposure to the radiation dose from radon (^{222}Rn) and its progeny has been the focus of public and scientific interest during the past 15 years. Epidemiologic studies of uranium miners suggest that chronic exposure to the radon levels equivalent to those encountered in some homes may pose a increased risk for lung cancer (BEIR VI, 1998). Epidemiologic studies in homes have generated mixed conclusions. Many studies have failed to detect a significant association. Reconstructing the long term dose associated with radon exposure in homes is a serious challenge in residential studies and a failure to do so accurately may be the reason for the mixed conclusions. Accurate estimates are needed for these low exposure studies that have limited sample sizes in order to overcome the statistical fluctuations in risk models associated with lung cancer confounders like smoking (Lubin *et al.,* 1995). Among the factors that cause variation in dose estimates are: the temporal and spatial variation of radon and radon progeny within homes; the individual's mobility within and outside the home; and the use of surrogate measures, like contemporary radon gas concentrations to estimate long-term radon-related dose.

The Iowa Radon and Lung Cancer Study (IRLCS) was designed to reduce some of theses uncertainties through participant eligibility criteria, year-long extensive radon gas measurements, mobility tracking, and historical average radon and radon progeny exposure assessment (Field *et al.,* 1996). This retrospective case-control study involved 1027 female Iowa residents, 413 cases and 614 controls, who had resided in their present homes for at least 20 years.

This paper describes our attempts to improve the estimates for long-term radon exposures and radon progeny dose delivered to those individuals in their homes. We measured the ^{210}Po activity implanted in glass surfaces, the annual average contemporary airborne radon gas concentration, and the surface-deposited ^{218}Po and ^{214}Po activity densities to retrospectively reconstruct the contemporary and historical average airborne radon and radon progeny concentrations.

While persistent alpha activity in glass was observed early in this century (Crookes, 1903), its use as a retrospective radon-radon progeny monitor is recent (Lively and Ney, 1987; Samuelsson, 1988). The basis for this method is the accumulation of a long-lived radon decay product, ^{210}Pb, in a stable matrix like glass. ^{210}Pb has an alpha-emitting decay product, ^{210}Po, that can be readily measured with a variety of techniques. Good correlation exists between total radon exposure and implanted ^{210}Po activity for glass surfaces exposed under laboratory conditions (Lively and Steck, 1993). Because few household glass surfaces have known radon exposure histories, this correlation is not well established in domestic environments. Several factors, present in the home but not yet studied in the lab, can affect the connection between airborne radon, radon progeny activity size distribution, and implanted activity. Nevertheless, previous surveys of the cumulative radon exposure and implanted polonium activity relationship in a few homes has shown moderate to good correlation (Lively and Steck, 1993; Mahaffey *et al.,* 1993; Samuelsson *et al.*, 1992; Steck *et al.*, 1993).

METHODS

We developed a detector module that simultaneously measures contemporary radon gas concentrations, surface-deposited ^{218}Po and ^{214}Po activity, and surface-implanted ^{210}Po (Steck, *et al*; 1990, Steck *et al,* 1993). We have previously referred to this detector as the HRD (Historical Reconstruction Detector), but in keeping with the more commonly accepted terminology for the retrospective reconstruction method have change the name to RRD (Retrospective Reconstruction Detector).

The glass-surface detector is designed to reconstruct the historical-average airborne radon and radon progeny concentration in living spaces through measurements and interpretations of alpha emitting radon progeny deposited on and implanted in surfaces. The surface deposited and implanted activities are interpreted in terms of the current airborne and historical-average airborne activities through a semi-empirical model (Cornelis *et al.*, 1992; Knutson, 1988; Porstendorfer, 1994). This steady-state model relies on four semi-empirical parameters; the ventilation rate, surface–to-volume ratio, aerosol attachment rate, and deposition rate. The model predicts that the ratio of total exposure to age-corrected, implanted ^{210}Po is ~0.5 ky m^{-1} in a typical room but that the ratio may vary by a factor of two in different rooms where the deposition and aerosol environments differ. Figure 1 shows the results of a Monte Carlo simulation of that ratio using measured and literature parameter values that reflect the expected conditions in the IRLCS homes. The geometric mean of this distribution is 0.6 ky m^{-1}. In a clean, small room like a bedroom, the ratio is twice as large, and it is twice as small in a big room with an open window and smoking. We tested the model by measuring 25 glass surfaces in a home with a known radon history (Steck and Field. 99). The surface activities were measured with both electronic and track-registration detectors (RRDs). The model-derived ratio for each room environment ranged from 0.3 to 0.4 kym^{-1}. The coefficient of variation (COV) between the 8 year average radon concentration and the interpreted implanted ^{210}Po result was 24%.

Detector design

Track registration material is ideally suited for use in epidemiologic studies to measure alpha activity in both air and glass surfaces since it is both rugged and inexpensive. Our modules use three chips of LANTRAK®[1] polymer attached to or enclosed in a 3 cm x 8 cm x 1 cm plastic container. Each chip has an area of ~200 mm^2. Approximately 20 mm^2 of the area is protected from exposure to monitor for contamination. All chips are protected from UV damage with metalized mylar (0.4 to 0.8 mg cm^{-2}). A chip inside the container serves as an airborne radon monitor. A chip held in contact with the glass measures implanted alpha activity and has an additional region protected by a thin filter (8.4 mg cm^{-2}) to monitor the glass for high concentrations of natural alpha emitters by detecting ^{212}Po+^{214}Po. A third chip faces the room to measure surface deposited alpha emitters in three regions. Two regions have filters (8.2 and 8.9 mg cm^{-2} to reduce incident alpha energy so that only ^{212}Po and ^{212}Po+^{214}Po alphas register. The deposited ^{218}Po activity is found from the third region by subtracting the minor contributions from ^{212}Po+^{214}Po and airborne ^{222}Rn.

Detector placement

Technicians from the IRLCS placed RRDs using the following guidelines. Normally two detectors were placed in each house; one in the subject's bedroom and one in another frequently-occupied room on another level. Detectors were attached to interior-facing, vertical glass surfaces that were free of visible coatings or colorings. We preferred glass that had been in the house more than 20 years but did accept surfaces as young as 5 years old. We accepted glasses that may have been in another house with the participant. We also preferred large surfaces that were free of obstructions or strong air currents, but frequently we had to compromise. Detectors were placed on washed areas as near the center of the glass as the homeowner would allow. The technician completed a data sheet on characteristics of the glass (type, age, history, washing history, films, and an air-movement test) and the room (type, size, ventilation, smoking presence, HVAC ducts).

Detector analysis

Detectors were retrieved after approximately one year, disassembled and developed at 75^0 C in a 6.25N NaOH solution for 6 hours. The detector contains 4 track-bearing areas and 3 contamination-monitoring areas. Each major track-bearing area on each chip was read under a microscope at 100X

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[1] A proprietary polymer from. Landauer, Inc., 2 Science Road Glenwood, IL 60425-1586, IL

until at least 150 tracks in 3 or more distinct regions were counted. In the three contamination monitoring areas, 20 mm^2 were counted. Two people read the detectors and each of them showed an individual variability of ~10%. The two readers agreed with each other to within 10%. The radon gas chip served as the quality assurance monitor for the module during QA exposures (spikes). The calibration of this chip was established in our radon chamber and confirmed through exposures at the U.S. Department of Energy's Environmental Measurements Laboratory. During the survey, 36 detectors exposed at the U.S. EPA's Montgomery chamber at rates equivalent to annual exposures at 74, 150, and 220 Bq m^{-3} agreed to within 10% of the target value, thus satisfying our QA goals. The alpha detection efficiencies of the other regions of the other chips were calibrated by exposure to thin sources traceable to NIST. Laboratory and field blanks showed no appreciable exposure after a minor storage contamination problem was solved shortly after the start of the IRLCS. A preliminary analysis of 52 field duplicate pairs (placed on the same glass but not necessarily side-by-side) show coefficient of variations (SD/mean) of 7, 7, 15, 8% for the radon gas, implanted ^{210}Po, deposited ^{218}Po and deposited total alpha activity regions respectively.

RESULTS

We placed 2445 HRDs in 1072 homes in Iowa. We retrieved 97% of those detectors, and based on a sub-sample of 1440 HRDs, approximately 90% of the retrieved detectors had a complete set of chips and the information necessary to interpret the chip results. All retrieved chips had sufficient tracks in all regions to meet the minimum analysis requirements. The HRDs were usually attached to mirrors (51%), windows (23%), or picture glass (11%) that were located predominantly in bedrooms (53%), living rooms (23%), or dining rooms (9%). The glass ranged from 5 to 150 years old, with a median of 31 years. Because heavy films can reduce implantation of heavy ions, we recorded washing habits and visible film thickness. Surfaces were primarily washed monthly (51%), yearly (22%), or infrequently (15%). The installers noted that the films on most glass were either non-existent (44%), light (41%), or moderate (11%).

Figure 2 shows the measured distribution of the surface to volume ratio, one of the parameters needed for the transport model. It is best fit with bimodal normal distributions with medians at 3.3 and 4.5 m^{-1}. These values are somewhat higher than the recommended typical value of 3 m^{-1} (Knutson 1988) that we used previously, but more in line with more recent recommendations (Porstendorfer 1994).

DISCUSSION

We have investigated the results from 1190 HRDs whose data sheets and track analysis have been reviewed and found to contain no errors or anomalies. Although we do not anticipate major changes in the data when the full set is analyzed, the following discussion and conclusions should be considered as preliminary.

We tested the historical reconstruction method by comparing the estimated total radon exposure of the surface with the implanted ^{210}Po activity. Although we don't have a measurement of the true radon exposure for these surfaces, we can estimate the total exposure from the product of the contemporarily measured radon gas concentration and the estimated age of the glass. If we assume that the year-to-year radon variation (Steck, 1995) is ~30% and the uncertainty in the age is ~20%, then we would expect a variation of ~ 35% in the data from the exposure uncertainties introduced by using the contemporary radon value as the historical average. Figure 3 shows the radon exposure estimated from contemporary gas measurements and the glass' age and the age-corrected implanted ^{210}Po surface activity. The correlation coefficient (r^2) is 0.5 and the average percentage difference between the measured contemporary radon and reconstructed historical average radon is only 45%, not much more than one would expect from the exposure variation alone. The slope of the linear regression line (0.44 ky m^{-1}) agrees with the standard model prediction for typical rooms and with previous observations (Steck *et al.,* 1993) but is smaller than our Monte Carlo simulation average. This difference will be investigated upon completion of the QA review of the complete data set. The fit of the implanted activity to the cumulative radon exposure is improved (r^2 ~0.7) when the measured deposited activities were added to the fit as linear regression variables. ^{218}Po and ^{214}Po are sensitive to the atmospheric conditions and can be used to correct the reconstructed airborne concentrations for variations in the airborne progeny's deposition effectiveness in different rooms.

Since the theoretical model for deposition effectiveness is not linear in these activities, we have investigated other ways of improving the reconstructed radon through ^{210}Po interpretation. One attractive approach uses the deposited activities to determine the model parameters. If these parameters can be found, then more accurate predictions can be made for the exposure-activity slope and other useful measures of risk, like the available dose. The Monte Carlo simulation sensitivity analyses suggests that the variation in dose in Iowa houses depends mainly on the radon gas variation (92%) and the surface deposition rate (7%). On the other hand, the variation in the implanted ^{210}Po activity depends somewhat less on the radon gas variation (64%), while the deposition rate and the attachment rate play larger roles (12% and 10% respectively).

Therefore, accurate retrospective reconstructions of the dose from implanted ^{210}Po require that the attachment and deposition rate be determined. We are investigating the use of the ratio of surface deposited ^{218}Po to ^{214}Po in to select the attachment rate and deposition velocity. We use categorical data from field notes and questionnaires to select the type of room. Our current categories are room size (small, larger), smoking (yes, no), ventilation (normal, drafty), and air movement (normal or turbulent). Next, we establish the functional relationship between the activity ratio and the model parameter through a fit of the Monte Carlo simulation results for that room type. Figure 5 shows the results for a small, nonsmoking room, with normal ventilation and air movement.

We are currently testing this methodology in a group of homes in Minnesota where the radon concentration has been measured for more than a decade. We also plan to test parts of the methodology in radon chambers before making a final interpretation of the results from surface measurements in the IRLCS homes.

CONCLUSIONS

Track registration detectors can provide valuable information in epidemiologic studies because the implanted ^{210}Po activity in glass correlates well with the historical average Rn concentration. The historical average radon exposure is key to the lifetime dose assessment that is critical for epidemiologic studies. We can improve the assessment by a measure of the deposition and attachment rates in the living spaces. Additional work is underway to test and refine this technique under realistic domestic conditions.

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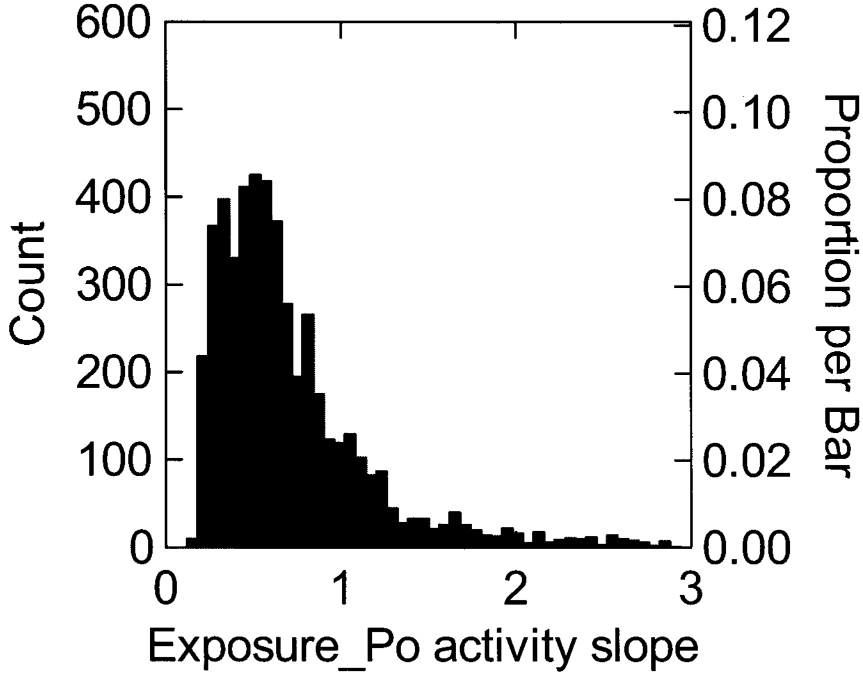


Figure 1: The surface to volume ratio distribution in IRLCS houses. The ratio was calculated from measured room dimensions and the fraction of the floor covered by furniture.

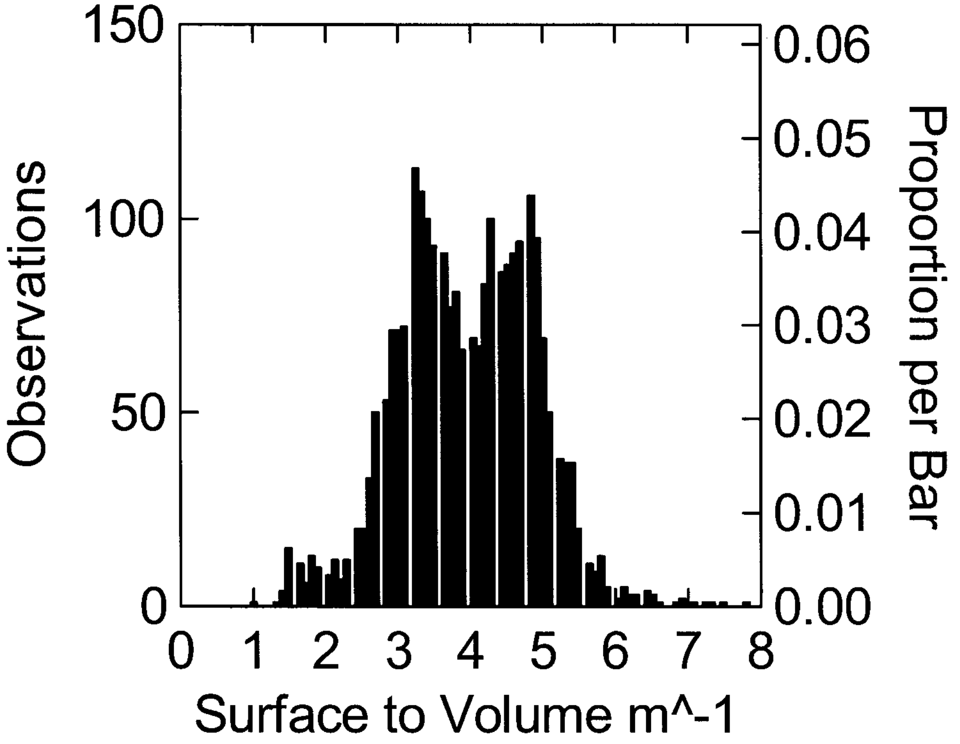


Figure 2: The distribution of the ratio of cumulative Rn exposure to the implanted ^{210}Po activity for a Monte Carlo simulation of the semi-empirical model using parameters that are believed to reflect indoor conditions in Iowa energy.

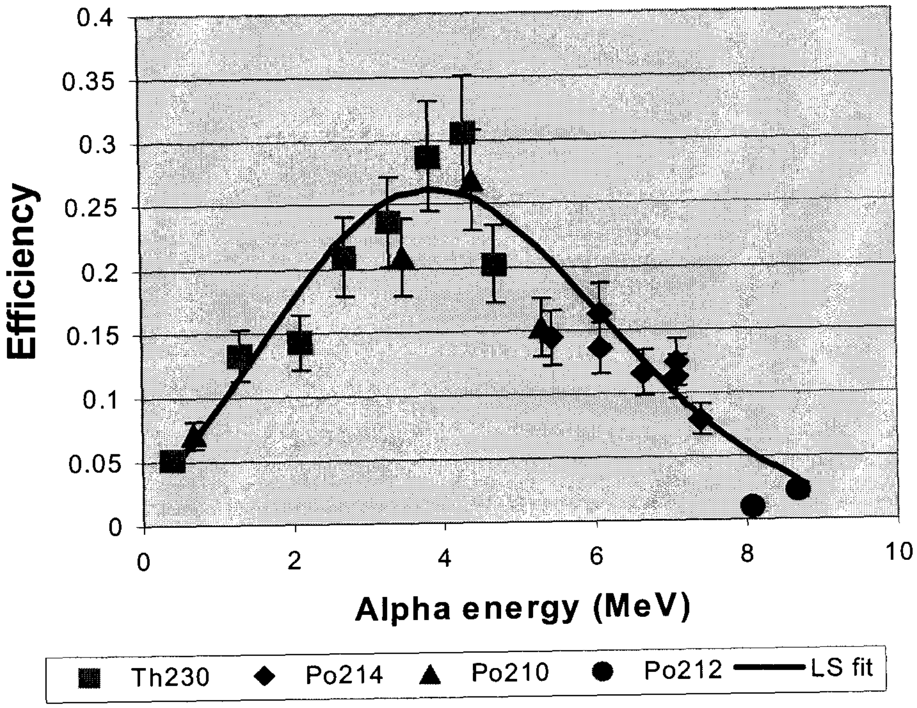


Figure 3: The detection efficiency of the track registration material as a function of incident alpha particle energy.

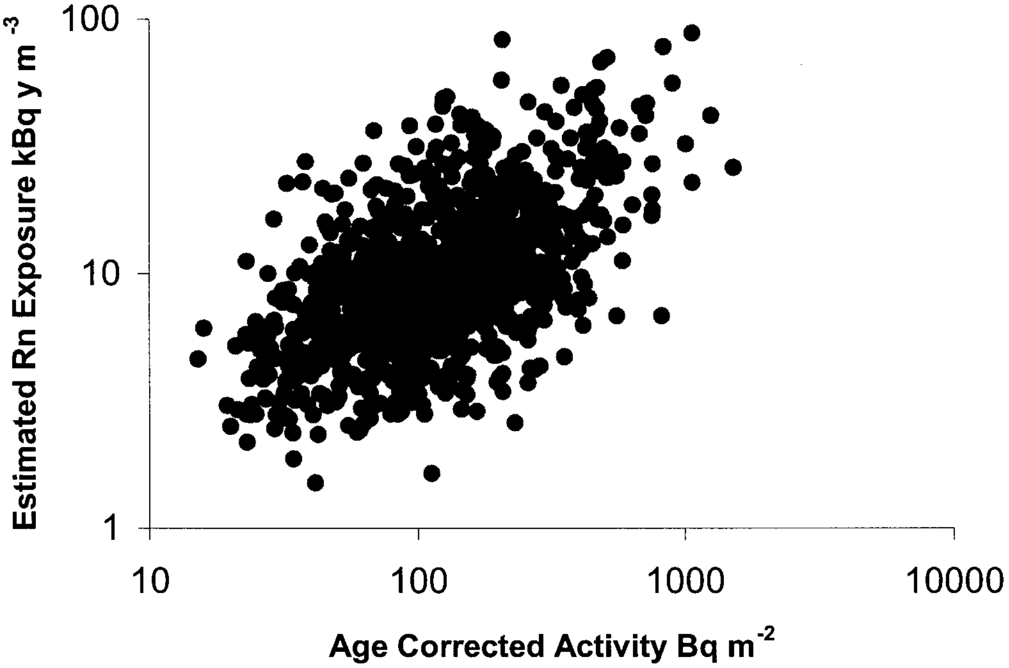
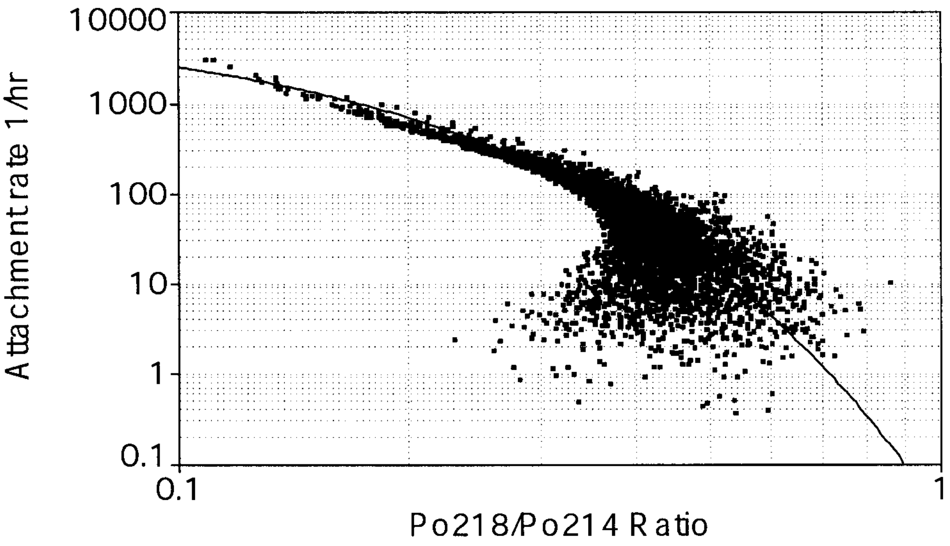
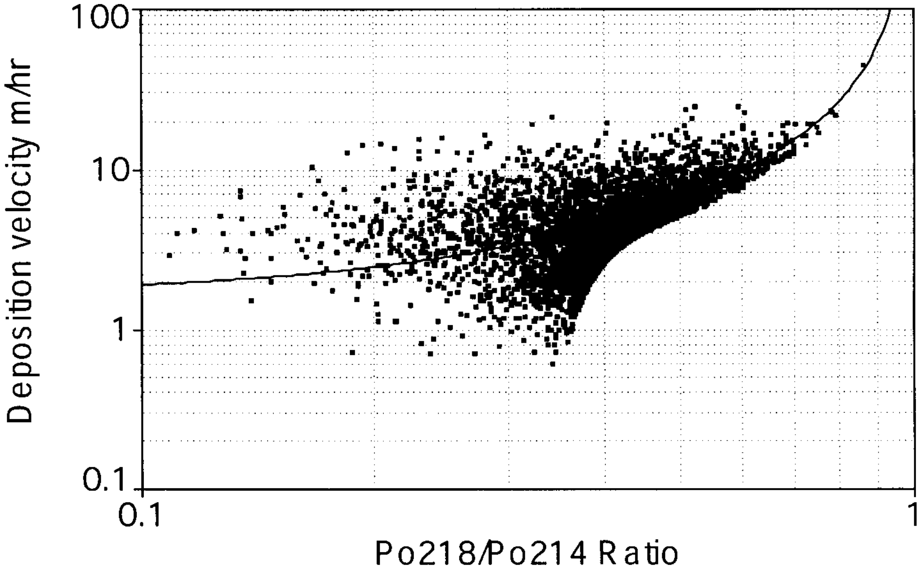


Figure 4: The total radon exposures estimated from contemporary radon gas measurements in more than 600 homes are plotted against the implanted surface activity of more than 1200 surfaces.



(a)



(b)

Figure 5: The relationship between the deposited $^{218}Po/^{214}Po$ surface activity ratio and (a) the attachment rate ; (b) the deposition velocity .