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Recommended Citation
Field RW, Steck DJ, Parkhurst MA, Mahaffey MA, Alavanja MCR. 1999. Intercomparison of retrospective radon detectors. Environmental Health Perspectives 107(11): 905-910.
Intercomparison of Retrospective Radon Detectors

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We performed both a laboratory and a field intercomparison of two novel glass-based retrospective radon detectors previously used in major radon case–control studies performed in Missouri and Iowa. The new detectors estimate retrospective residential radon exposure from the accumulation of a long-lived radon decay product, 210Pb, in glass. The detectors use track registration material in direct contact with glass surfaces to measure the α-emission of a 210Pb-decay product, 210Po. The detector’s track density generation rate (tracks per square centimeter per hour) is proportional to the surface α-activity. In the absence of other strong sources of α-emission in the glass, the implanted surface α-activity should be proportional to the accumulated 210Po, and hence to the cumulative radon gas exposure. The goals of the intercomparison were to a) perform colocated measurements using two different glass-based retrospective radon detectors in a controlled laboratory environment to compare their relative response to implanted polonium in the absence of environmental variation, b) perform colocated measurements using two different retrospective radon progeny detectors in a variety of residential settings to compare their detection of glass-implanted polonium activities, and c) examine the correlation between track density rates and contemporary radon gas concentrations. The laboratory results suggested that the materials and methods used by the studies produced similar track densities in detectors exposed to the same implanted 210Po activity. The field phase of the intercomparison found excellent agreement between the track density rates for the two types of retrospective detectors. The correlation between the track density rates and direct contemporary radon concentration measurements was relatively high, considering that no adjustments were performed to account for either the residential depositional environment or glass surface type. Preliminary comparisons of the models used to translate track rate densities to average long-term radon concentrations differ between the two studies. Further calibration of the retrospective detectors’ models for interpretation of track rate density may allow the pooling of studies that use glass-based retrospective radon detectors to determine historic residential radon exposures. Key words: case-control studies, dose–response relationship (radiation), epidemiologic methods, epidemiologic studies, lung neoplasms, radon, radon progeny, smoking. Environ Health Perspect 107:905–910 (1999). [Online 15 October 1999] http://ehpnet1.niehs.nih.gov/docs/1999/107p905-910/abstract.html

The National Research Council of the National Academy of Sciences (NAS) estimates that approximately 18,600 lung cancer deaths (range 3,000–32,000) in the U.S. population each year may be caused by residential exposure to radon-222 (radon) decay products (1). NAS researchers caution that these risk estimates derived from radon-exposed underground miners and applied to the general nonoccupationally exposed population must be cautiously interpreted because of inherent differences in lifestyle factors between these populations, as well as differences between the mine and the home environments.

The most direct way to derive risk estimates for residential radon decay product (progeny) exposure is to compare residential radon progeny exposure among people who have lung cancer with the exposure received by individuals who have not developed lung cancer. Numerous case–control epidemiologic investigations have attempted to examine the relationship between residential radon gas exposure and lung cancer (2–11). The historic reconstruction of radon exposure presents a formidable challenge in these studies. The major obstacles impeding accurate radon exposure estimates for the epidemiologic studies (12) include the studies’ inability to account for missing radon measurements for homes that were previously occupied by the subjects and were inaccessible for radon testing (2–10), temporal and spatial variation of residential radon concentrations (2–10), and the use of current residential radon gas concentrations as a surrogate for past residential radon progeny concentrations (2–10).

Previous residential radon case–control epidemiologic studies have imputed from 17 to 40% of their radon measurements for dwellings occupied by the study participants for the 20-year period preceding study enrollment (2–10). The missing measurement data create significant gaps in the participants’ exposure history, which compel the investigators either to analyze a reduced data set or to impute radon concentrations for missing homes (13). These gaps in radon measurements seriously decrease a study’s statistical power to detect an association (14), especially if the gaps occur 5–15 years before study enrollment (12).

Studies that fail to consider temporal radon gas and progeny variation will also have higher exposure misclassification. Residential radon gas and progeny concentrations vary hourly, diurnally, monthly, seasonally, and annually. These variations are influenced by numerous factors including radon infiltration rates, heating and air conditioning system design and usage, pressure differentials, soil characteristics, house construction methods and materials, water usage, weather conditions (e.g., rainfall, wind speed), and occupant behavior (15–17).

The epidemiologic studies published to date have examined the relationship between radon gas exposure and lung cancer (2–11). However, it is radon progeny rather than radon gas itself that delivers the actual radiation dose to the lung tissues (1). The effective dose conversion coefficient for radon progeny strongly correlates with the size of the aerosol cluster associated with the radon progeny. Radioactive clusters in most domestic atmospheres usually contain multiple size fractions. The smaller particles (3–10 nm) provide greater exposure to the airways than radon progeny that are attached to larger aerosols (diameters of ~ 100 nm), primarily because of their high rate of deposition in the bronchial region. The particle size distribution varies with changes in radon concentration and changes in the domestic atmosphere that include aerosol density, air movement, and the air exchange rate. Thus, both natural factors (e.g., weather patterns) and homeowner activities (e.g., cooking) can dramatically alter the delivered dose over short periods. The use of radon gas rather than progeny concentrations alone can routinely...
introduce an uncertainty of 50% in the exposure estimates (18). Improved residential radon exposure estimates require measurements that depend on actual airborne radon decay product concentrations.

To overcome these exposure assessment obstacles, detectors that analyze the α-activity implanted in glass surfaces have been developed for reconstructing past residential progeny concentrations (19–23). The persistent α-activity in glass was observed early in this century by Crookes (24), but its use as a retrospective radon–radon progeny monitor is recent (25,26). The new detectors use the accumulation of a long-lived radon decay product, 210Po, in glass. Radon's radioactive decay chain produces a daughter product, 210Pb, with a long half-life (approximately 22 years). A fraction of the 210Pb implants in glass surfaces in a room, which provides a long-lasting marker for past radon concentrations. 210Pb produces a shorter lived daughter product, 210Po. The 210Po decay can be captured by measuring the etched tracks created in a suitable piece of plastic by the emitted α-particles.

The possibility of using household glass as an indicator of historical radon concentrations is potentially of great importance to epidemiologists studying radon, because retrospective radon exposure assessment over many years otherwise usually requires going into individuals' former homes and making long-term radon measurements. However, many of those homes may no longer exist, others may have current owners not interested in cooperating with the study, and still other homes may have been modified in ways that affect the residential radon concentrations. On the other hand, a piece of glass (e.g., that in front of a treasured family picture) may have been owned for a long time, relocated with the subject, and been displayed in the current and former homes. Thus, such glass can serve as a long-term exposure integrator.

210Po α-emissions can be measured by a variety of detecting techniques. Track registration detectors are suited to this task for domestic surveys. The α-particles from the glass produce microscopic damage tracks in the plastic that can be easily developed and measured. The track generation rate is then a measure of α-activity of the surface. Previous work has demonstrated that an excellent correlation exists between cumulative radon exposure and the activity of implanted 210Po for glass surfaces exposed under laboratory conditions (19). Additional studies of this relationship in a sample of homes have also shown moderate to good correlation between contemporary year-long radon gas concentrations and historically derived radon gas concentrations from detectors that measure the implanted progeny in glass (19–21,27). The ratio between the cumulative radon exposure and the implanted activity can vary with the aerosol and atmospheric conditions in each room. This behavior presents a challenge in accurately reconstructing either the airborne radon concentration or the radon-related dose based on implanted activity alone. However, this behavior also presents an opportunity to reconstruct the airborne concentrations when the implanted activity is combined with contemporary radon gas and deposited radon progeny measurements.

This paper reports the results of an intercomparison study between two devices for assessing historical exposure to radon progeny using household glass. The goals of this study were to perform collocated measurements using two retrospective glass-based radon detectors in a controlled laboratory environment to compare their relative response to implanted polonium in the absence of environmental variation, perform collocated measurements using two retrospective radon progeny detectors in a variety of residential settings to compare their detection of α-decays due to implanted polonium, and examine the correlation between α-track density rates and contemporary radon gas concentrations.

**Methods**

The two detectors compared in this paper have been used in major epidemiologic residential radon studies (28–30) to estimate exposure from long-term radon progeny delivered to individuals in their homes. The Missouri Radon Lung Cancer Study [MRLCS; (28)] and the Iowa Radon Lung Cancer Study [IRLCS; (29)] were case-control epidemiologic studies that evaluated the lung cancer risk posed by residential radon exposure. The studies used both traditional retrospective radon gas detectors and retrospective radon gas and progeny detectors to estimate historic radon concentration. The MRLCS inclusion criteria allowed subjects to have lived in more than one home over the 20 years before enrollment. The IRLCS limited enrollment of subjects to those individuals who lived in the current home a minimum of 20 years.

**Retrospective surface monitor (RSM).** The RSMs for the MRLCS were developed by Pacific Northwest National Laboratory in Richland, Washington. The RSM measures implanted 210Po activity in glass surfaces. The RSMs were produced from dosimetry grade CR-39 plastic sheets manufactured by American Technical Plastics, Inc. (Stratford, CT). Each 5 cm × 5 cm monitor had a protective polyethylene film, which was removed before placement. The detecting side of the RSM was placed against a glass surface and held in place by taping the perimeter of the monitor with polypropylene tape.

During the MRLCS study, the RSMs were affixed to the glass surface for 4–5 weeks. On their return to the laboratory, the RSMs were chemically etched in a 75°C solution of 6.25 N NaOH for 5.5 hr. After the RSMs were etched, a matrix of 50–100 fields (0.002 cm²/field) depending on track density, was manually evaluated under an optical microscope at 200× magnification. The number of α-tracks counted was converted to tracks per square centimeter and divided by the exposure duration (in hours) of the RSM on the glass surface to produce the track density rate (tracks per square centimeter per hour). Additional details concerning the RSM, which was previously identified as the CR-39 surface monitor, are available elsewhere (21,22,28).

**Retrospective reconstruction detector (RRD).** The RRDs used in the IRLCS were developed by the Physics Department at St. John's University in Collegeville, Minnesota. Both the RRD and the RSM measure the 210Po activity implanted in glass surfaces, but the more complex, multicomponent RRD also measures the activities of the contemporary airborne radon gas concentration and surface-deposited 210Po and 214Po. The detector (3 cm × 8 cm × 1 cm) (Figure 1), manufactured from dosimetry grade CR-39 plastic sheets obtained from Landauer, Inc. (Glenwood, IL), used three track registration chips per detector. Chip G, which measured surface-implanted 210Po, faced the glass surface (similar to the RSM), whereas the contemporary surface-deposited radon progeny...
measuring chip faced away from the glass and toward the room (chip D). The third chip (chip C), which measured the contemporary radon gas concentration, was located inside the filtered enclosure of the RRD.

Each of the three chips (1 cm x 2 cm) of the RRD had a protective covering, which was removed before placement. The RRD was secured to glass surfaces with a self-adhesive on the detector housing. The RRDs were placed on glass surfaces for a 1-year exposure period. After the exposure period, the RRDs were disassembled in the laboratory and developed at 75°C in a 6.25-NaOH solution for 6 hr. The number of \( \alpha \)-tracks counted was converted to tracks per square centimeter and divided by the exposure duration (in hours) of the RRD on the glass surface to produce the track density rate (tracks per square centimeter per hour). The detector (Figure 1) contained four track-bearing areas (CT, GT, DT, DD) and three contamination-monitoring areas (CB, GB, GP). Each track-bearing area on each chip was read under a microscope at 100 x until at least 150 tracks in three or more distinct regions were counted. The radon gas chip (Figure 1, chip C) served as the quality assurance monitor for the module during quality assurance exposures (spikes). The calibration of the radon gas chip was established in a private radon chamber and confirmed by exposures at the U.S. Department of Energy Environmental Measurements Laboratory (New York, NY). The \( \alpha \)-detection efficiencies of the other regions of the other chips were calibrated by exposure to calibrated surface \( \alpha \)-sources. Additional details concerning the RRD, which was previously identified as the historic reconstruction detector, are available elsewhere (19,20,25).

Design of laboratory intercomparison. The RRDs and RSMs were exposed to implanted \(^{210}\)Po sources under controlled conditions to compare their relative responses. These exposures allowed comparison of the relative performance of the devices in the absence of high airborne radon concentrations, intrinsic glass radioactivity, or varying environmental conditions. We used the probability that an observable track is produced by an \( \alpha \)-particle emitted from surface-implanted \(^{210}\)Po as the measure of the detector's relative response. This probability, which we refer to as the efficiency, can be calculated from the ratio of the observed track generation rate to the implanted \(^{210}\)Po activity density. The calibrated \(^{210}\)Po sources, glass surfaces exposed to high radon concentrations for \( \geq 1 \) year, had activity concentrations of approximately 2–5 kBq/m². Their activity was calibrated through a large-area \(^{230}\)Th source, whose calibration is traceable to the National Institute of Standards and Technology (Gaithersburg, MD). The exposure times were short (20–700 min) to produce track densities similar to those encountered under field conditions for each device type. RRD track densities ranged from 500 to 10,000 tracks/cm², whereas RSM densities ranged from 50 to 500 tracks/cm² to simulate their shorter field exposure period.

Design of field intercomparison. The intercomparison study of glass-based retrospective radon detectors started in 1995 with the placement of detectors in a sample of homes that had previously participated in either the MRLCS or the IRLCS. Informed consent to participate in the study was obtained from each subject. Twenty-two homes in Iowa and 23 homes in Missouri were chosen to take part in the intercomparison. The 45 study homes were selected based both on the willingness of the homeowner to take part in the study and the availability of suitable glass surfaces. The selection of study homes in Missouri was also weighted to include sample homes with higher estimated radon concentrations based on previous measurement results.

One technician from the MRLCS and one from the IRLCS were trained on the proper protocols for the placement of the detector used in each study. The technicians followed guidelines for the selection of an appropriate glass surface and the placement of the detectors according to the following criteria:

- the glass surface must be ordinary, smooth glass without visible coatings or colorings (not lead crystal)
- the glass must be vertical and facing the interior of the home
- the glass surface must have a known age (preferably between 20 and 70 years old) and the age must be accurate to within 10%, if possible
- the glass item must have been purchased new by the subject
- the glass surface must be large enough to accommodate the placement of both detectors
- the glass surface must be unobstructed, with no large objects, such as curtains, within 10" of the glass surface
- the glass surface must be isolated from strong air currents such as vents, fans, and open windows.

Recommended items for detector placement were photo frames, picture glass, mirrors, and cupboard or interior door glass. At each study site, the technician completed a data sheet on characteristics of the glass (type, age, history, washing history, storage history, films, and an air-movement test) and the room (type, size, ventilation, smoking presence, heating and air-conditioning ducts).

The detectors were placed on alcohol-washed areas as close to the center of the glass as the homeowner would allow (Figure 2). The detector pairs (one RSM and one RRD) were placed on the same glass surface in the master bedroom and living area (usually the kitchen) in each home. The RSM and RRD were used for their normal placement periods of 1 month and 1 year, respectively. Duplicate RSMs and RRDs were placed at 10% of the placement sites to examine the precision of the measurements.

The measurement results from the retrospective detectors were expressed as an \( \alpha \)-track density rate (tracks per square centimeter per hour). One track/cm² x hr corresponded to 0.3 pCi/m² (11 Bq/m²) at a detection efficiency of 25%. Interpreted retrospective radon concentrations are not presented because the RRD's adjustment factors for various depositional environments and surfaces (19,21) are still undergoing calibration. Statistical analyses included the Pearson product moment correlation coefficient (\( \rho \)), to assess the correlation between the track density rates for the RSM and RRD, and either a paired t-test (for normally distributed data) or a Wilcoxon signed rank test (for data that could not be transformed to a normal distribution) to assess the existence of significant systematic bias between the track density rates for the RSM and RRD. The coefficient of variation (CV) was calculated to assess the variation (precision) between the RSM and RRD track density rates. The Kolmogorov-Smirnov test was used to assess the normality of the data. Geometric means and geometric
standard deviations were used as summary descriptors for the field intercomparison results because of the log-normal nature of the track density data.

**Results**

**Laboratory intercomparison.** The $^{210}$Po detection efficiency for the RSMs was 26% with a CV of 44% (Table 1). The RRD exhibited an efficiency of 22% and a CV of 14%. A Wilcoxon signed rank test did not detect a significant difference ($p = 0.30$) between the detection efficiency of the devices for the common exposure class (500 tracks/cm²).

**Field intercomparison.** The technicians successfully retrieved the 46 RRDs and 46 RSMs from the 23 Missouri homes and 44 RSMs and 43 RRDs from the 22 Iowa homes. Placement duration adhered to established protocols for each detector, with the exception of one RRD in Iowa, which was inadvertently discarded by a family member. The detectors in Missouri were placed on mirrors ($n = 20$), picture glass ($n = 18$), or cabinet glass ($n = 8$). The detectors in Iowa were placed on mirrors ($n = 14$), picture glass ($n = 13$), cabinet glass ($n = 11$), or the interior surface of window glass ($n = 5$). The mean age and standard deviation (SD) of the glass surfaces in Missouri and Iowa were 37 ± 10 years and 26 ± 6.7 years, respectively. Ninety-eight percent and 64% of the glass surfaces in Missouri and Iowa, respectively, were exposed to cigarette smoke for at least 2 months during their residency in the home. All of the glass objects selected for Iowa and 52% of the glass objects in Missouri resided only in the current home. The five sets of duplicate RSMs exhibited a mean and SD of the CV of 22% ± 13% at an average track rate of 0.33/(cm² × hr). The five sets of duplicate RRDs, conducted on a separate sample set, exhibited a mean and SD of the CV of 5 ± 3% at an average track rate of 0.44 tracks/(cm² × hr).

**Agreement between retrospective measurement results.** Table 2 presents summary measurement results for the two retrospective detectors. The collocated retrospective detectors in Missouri produced geometric mean track rate densities of 0.39 tracks/(cm² × hr) for the RSM and 0.42 tracks/(cm² × hr) for the RRD. The track rate densities for both detectors were log-normally (ln) distributed. A paired $t$-test on the ln-transformed data did not find any systematic bias ($p = 0.10$) between the observed track rates for the collocated detectors in Missouri. The Pearson product moment correlations between the RSM and RRD both for the raw data and ln-transformed RSM data were 0.89 and 0.84, respectively.

Geometric mean track rate densities of 0.48/(cm² × hr) for the RSM and 0.55/(cm² × hr) for the RRD were observed for the Iowa detector placements (Table 2). A paired $t$-test on the ln-transformed data did not detect any systematic bias ($p = 0.26$) between the observed track rates for the collocated detectors in Iowa. The Pearson product moment correlation between the two sets of detectors for the raw and ln-transformed data was 0.93 and 0.87, respectively. The overall correlation for the 89 sets of collocated retrospective detectors for the Missouri and Iowa placements combined was 0.92 for the raw and 0.86 for the ln-transformed track density rates (Figure 3).

A paired $t$-test on the ln-transformed data noted a systematic bias ($p = 0.009$) between the observed track rates for the 29 sets (RSM and RRD) of smoke-exposed glass surface collocated measurements in Iowa. The Pearson product moment correlation between the 29 sets of detectors for the ln-transformed data was 0.78. However, a paired $t$-test on the ln-transformed data did not detect any systematic bias ($p = 0.16$) between the observed track rates for the 14 sets of non-smoke-exposed glass surfaces in Iowa. The Pearson product moment correlation between the sets of detectors measuring the non-smoke-exposed glass surfaces for the ln-transformed data was 0.95. Subset comparisons by smoke exposure were not performed for the Missouri data because of the small number (one) of non-smoke-exposed glass items.

**Agreement between track density rates and contemporary measurement results.** In Missouri, the Pearson product moment correlation between the ln-transformed contemporary (1-year) radon gas concentrations, measured using the filtered chip (Figure 1, chip C) of the RRD, and the ln-transformed track rate densities from the RSM and RRD measurements for the subset of 24 glass surfaces that were only in the current home, was 0.62 and 0.63, respectively (Figure 4A and B, respectively). The Pearson product moment correlation between the ln-transformed contemporary (1-year) radon gas concentrations, measured using the filtered chip of the RRD, and the ln-transformed track density rates from the RSM and RRD measurements for Iowa placements was 0.76 and 0.78, respectively (Figure 5A and B, respectively).

The Pearson product moment correlation between the ln-transformed contemporary (1-year) radon gas concentrations, measured using the filtered chip of the RRD, and the ln-transformed track density rates from the RSM and RRD measurements in Iowa for...
the 29 smoke-exposed glass surfaces was 0.64 and 0.58, respectively. For the 14 non-smoke-exposed glass surfaces in Iowa, the Pearson product moment correlation between the In-transformed contemporary (1-year) radon gas concentrations, measured using the filtered chip of the RRD, and the In-transformed track density rates from the RSM and RRD measurements was 0.89 and 0.95, respectively.

**Discussion**

The absolute detecting efficiency served as the performance metric for the laboratory calibration intercomparison exposure. The results suggest that the detecting materials and track-reading protocols in Missouri and Iowa produced similar efficiencies. The Missouri and Iowa field phase of the intercomparison found good agreement between the track density rates for the two types of retrospective detectors.

The correlations both between the track density rates produced by the two detectors and between the track density rates and contemporary radon concentrations were slightly higher for the Iowa study sites. Several factors may have reduced the correlation for the Missouri placements including the older age of glass surfaces and the higher percentage of smoke-exposed glass at the Missouri subset placement sites. The retrospective radon concentrations most likely varied more in homes where the measurement covered a longer retrospective period. The subset analyses performed on the non-smoke-exposed glass surfaces found higher correlation, as compared to the smoke-exposed glass surfaces, both between the track density rates produced by the two collocated detectors and between the track density rates and contemporary radon concentrations.

This finding suggests that previous smoke exposure to the glass surfaces may increase the variation in track densities between collocated measurements. To improve retrospective radon concentration estimates using retrospective detectors, the source of this variation requires further investigation. Nonetheless, the agreement between the track density rates and contemporary radon concentrations was excellent, especially for the non-smoke-exposed glass surfaces, considering that no adjustments were performed to account for either the depositional environments or surface type.

However, preliminary comparisons of the models used to translate track rate densities to average long-term radon concentrations differed between the two studies. The RSM track density generation rate has been used to derive historic radon gas concentrations using a calibration factor based on linear regression of the contemporary residential radon gas concentrations and the track density generation rates in MRLCS homes (22). The age of the glass was used to correct the observed track density rates for radioactive decay (20). The RRD track densities from the IRLCS homes were similarly analyzed to produce a calibration factor (20,23). The two calibrations had similar multiplicative factors, but differed enough in detail that further work is needed to reconcile the two approaches. Some of the differences may arise from the variation of room atmospheres in the two studies. For example, more smokers were present in MRLCS houses than in IRLCS houses. The effect of atmospheric differences is being investigated using the results of the RRD’s deposition chip and IRLCS questionnaire data on aerosol generation such as from smoking and other factors. If categorical variables common to both MRLCS and IRLCS can be identified that significantly reduce the effects of room atmosphere variation on the calibration, those variables will be used to adjust both RSM and RRD track densities for comparisons.

In summary, laboratory and field intercomparisons found that two glass-based retrospective radon detectors, previously used in major epidemiologic studies, produced similar track density rates. Further calibration of these glass-based retrospective detectors for various depositional environments and surfaces will further refine this measurement technique and may allow pooling of glass-based retrospective radon measurements from two large radon epidemiologic studies performed in the United States (28, 29).

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