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SPATIAL AND TEMPORAL VARIATIONS OF INDOOR AIRBORNE RADON DECAY PRODUCT DOSE RATE AND SURFACE-DEPOSITED RADON DECAY PRODUCTS IN HOMES

Daniel J. Steck[1], **Kainan Sun**[2], **R. William Field**[3]

[1]Eagle River, MI;

[2]Department of Occupational and Environmental Health, College of Public Health, University of Iowa, Iowa City, IA;

[3]Department of Occupational and Environmental Health, Department of Epidemiology, College of Public Health, University of Iowa, Iowa City, IA.

Abstract

The temporal variations of the airborne radon decay product dose rate and deposited radon decay product activities, as well as the within-house and house-to-house variations of radon concentrations, were evaluated through repeated field measurements. Long-term average radon and surface-deposited radon decay product concentrations were measured in 76 rooms of 38 houses. Temporal variation of radon, as well as airborne and surface-deposited radon decay products, were measured in 11 of the 38 houses during two different seasons. Environmental factors that have the potential to influence airborne dose rate and deposited radon decay products were also studied. Airborne dose rates were calculated from the unattached and attached potential alpha energy concentrations using two dosimetric models. For one model, the observed dose variations were 103%, 74%, 58%, and 60% for the total, house-to-house, within-house, and within-room temporal variations, respectively. For the other model, the dose variations were 100%, 66%, 61%, and 46%, respectively. Surface-deposited ^{214}Po showed variations of 79%, 57%, 42%, and 48%, respectively. These substantial radon decay product concentration variations suggest that multiple locations and time-integrated measurements are needed to make an accurate assessment of the chronic radon-related doses in homes. Smoking was the environmental factor that had the largest temporal and spatial effect on airborne radon decay product dose rates.

Keywords

detector; alpha-track; naturally occurring radionuclides; radiation dose; radon

For correspondence contact R. William Field, Department of Occupational and Environmental Health, Department of Epidemiology, College of Public Health, S327 CPHB, 145 North Riverside Drive, University of Iowa, Iowa City, IA 52242, or bill-field@uiowa.edu.

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INTRODUCTION

Epidemiologic studies that investigate the relationship between prolonged radon exposure and lung cancer, as well as other adverse health outcomes, would greatly benefit from measurements of the long-term average radon decay product (RDP) concentrations in dwellings since the radon decay products, rather than the radon gas, deliver the radiologically significant dose to the respiratory epithelium of the lung (Field et al. 1996). The use of contemporary radon gas concentrations as a surrogate for radon-related dose has the potential to introduce significant uncertainty in dose assessment (Steck and Field 1999, 2006). Effective dose estimates based on radon gas measurements generally have larger uncertainties than those based on radon decay product measurements (Singh et al. 2007). Few simultaneous measurements of radon and its decay products in occupied homes have been reported. In addition, human activities like breathing patterns (e.g., mouth vs. nose breathing), room occupancy patterns, and aerosol generation can also significantly impact the accuracy of the cumulative radon dose estimates for individuals.

Radon and its decay products show substantial spatial and temporal variations in many domestic environments, which poses additional obstacles for reconstructing an individual's long-term radon-related exposures (Abu-Jarad and Fremlin 1984; Cohen 1991; Martz et al. 1991; Hotzl and Winkler 1994; Miles 2001; Winkler et al. 2001; Kobayashi 2002; Baysson et al. 2003; Martinez et al. 2004; Sahota et al. 2004; Baciu 2005; Krewski et al. 2005; Hadad et al. 2007; Seftelis et al. 2007, 2008; Singh et al. 2007; Steck and Field 2006; Steck 1992, 2005, 2009). Several research studies, performed in the US Upper Midwest, highlighted the need to account for some of these variations to obtain accurate long-term average dose estimates for individuals (Fisher et al. 1998; Zhang et al. 2007; Steck 1992, 2005, 2009).

Previously, we reported the results of the field investigation (Sun et al. 2009) and modeling (Sun et al. 2010) of the behavior of radon-related decay product behavior in occupied homes. The primary aim of the current paper is to report the within-house and house-to-house spatial variations of the radon concentrations, the airborne radon decay product dose rate, and the deposited radon decay product activities as well as the seasonal temporal variation of those variables. An additional aim of the study was to examine the influence of environmental factors on those variations.

MATERIALS AND METHODS

Study design

Field tests were conducted between the summer of 2005 and spring 2007, with equipment placed and retrieved by trained radon measurement personnel. We designed the radon gas and active radon decay product measurements to be as nonintrusive as possible to allow measurements that covered the same time span as most common radon gas exposure assessments in actively occupied homes. Thirty-eight volunteers provided access to their homes for measurement periods that lasted up to 90 d. The houses were located in southeast Iowa and were occupied by either active smokers or lifetime nonsmokers. The houses were selected from a larger sample based on radon screening tests that indicated that they had elevated radon gas concentrations in the living area. The majority of the homes had been

occupied by the current resident for at least 5 y. In each of the 38 houses, two frequently occupied rooms in either the basement, first, or second floor having a clean, unobstructed glass object in them were deemed eligible for study of contemporary radon decay product deposition on exposed glass surfaces. Additional details of site selection and participation can be found in Sun et al. (2009).

Radon and radon decay product measurements

Table 1 provides a summary of the radon gas and decay product measurements. Radon gas concentrations were measured in each room over short (3 to 7 d) and long (90 d) time periods. Commercially available electret ion chambers (EICs) were used for short-term measurements while long-term radon concentrations were measured with alpha track detectors (ATDs) manufactured, calibrated, and analyzed in an author's (DS) laboratory. Radon gas concentrations in the lab's radon exposure room during calibration ranged from 400 to 1,000 Bq m^{-3} and were monitored with a Durridge RAD7 (Durridge Company, Inc., Billerica, Massachusetts, US) whose calibration was periodically checked in the manufacturer's calibration chamber. The ATDs successfully passed tests during national and international comparison exercises and were listed in national proficiency programs.

Airborne and surface-deposited RDP measurements used the same track registration material as the ATD. A retrospective radon detector (RRD) was placed on the selected glass object for 3 mo. This passive detector provided the long-term average surface-deposited radon decay product concentrations (Steck and Field 1999; Field et al. 1999; Steck et al. 2002, 2005). Contemporary airborne radon decay products were measured in each identified eligible room in the house for periods up to a week with an active radon decay product detector (ARPD) (Sun et al. 2009). National Institute of Standards and Technology (NIST)-traceable alpha sources were used to calibrate the material's efficiency response to RDPs. The specific response of the material to the alpha-emitting radon decay products when used in the RRD and the ARPD was determined through repeated exposures in a laboratory walk-in room under a variety of environmental and aerosol conditions, with the radon concentration ranging from 1,500 to 3,000 Bq m^{-3}. During these exposures, a German Federal Office for Radiation Protection (Bfs)-calibrated SARAD EQF 3120 (SARAD GmbH, Dresden, Germany) was used to measure the radon gas, unattached (<5-nm) and attached (>5-nm) activity-size fractions. Large-area semiconductor diode detectors measured the deposited RPD activities. Repeated laboratory exposures gave an estimate of the precision of the detectors under different exposure conditions (Steck et al. 2005, 2007). All track registration detectors were analyzed independently by two of the authors (DS, KS). The performance quality of the track registration detectors was monitored by analyzing unexposed detectors and detectors exposed to known activities, and by using side-by-side (i.e., collocated) exposures.

Domestic activities that might affect environmental parameters, such as indoor aerosols and ventilation conditions, were gathered through face-to-face interviews. The information gathered was categorical, such as the presence or absence of smoking in a room, and whether windows were open or closed. Additional detailed information about the

measurement devices and procedures used in the field study of radon decay product behavior can be found elsewhere (Sun et al. 2009, 2010).

Seasonal variation

The temporal variation of radon gas and radon decay products was evaluated in a sample of 11 houses, selected from the original 38, for a second measurement round that included both heating and nonheating seasons. Most of the 11 homes selected for this phase of the study were located in Kalona, Iowa, in southeast Iowa. Three of the 11 houses were occupied by at least one current smoker. For each room in each test season, one short-term radon measurement was performed at the end of the long-term measurement period, which provided another measurement from a different period. The original measurements for these 11 houses took place primarily in the winter 2005 or spring 2006 heating season. All the radon and radon decay product measurements, as well as the concurrent documentation of the indoor environmental conditions, were repeated in the summer and fall of 2006 in the selected 11 houses following the same protocols used in the original investigation (Sun et al. 2009).

Dosimetric models

In this paper, two models are used to calculate the effective dose rate from the RDP measurements. Doses calculated from RDP measurements depend on numerous assumptions about the indoor environment, occupant behavior, and fate of inhaled RDPs. We used two well-established dosimetric models available in 2006. Both models were based on the International Commission on Radiological Protection (ICRP) lung model (ICRP 1993). While newer models are now available, the models used in this study illustrate the effects that different assumptions have on the activity-size weighting factors even when the models start from the same root model. These model predictions are not intended to produce highly accurate estimates of the actual available effective dose rates, but rather they show the variation possible in the interpretation of RDP measurements under different aerosol and environmental conditions.

For both the model referred to as Jdose (James et al. 2004) and the model referred to as Pdose (Porstendörfer 2001), we assumed that an equal prevalence of mouth and nose breathing for one residential year was equal to 42 WM and that the absorbed-dose-to-effective-dose conversion was 1 Gy = 2.4 Sv. Since the ARPD recorded activity from two size fractions, unattached (up to 5 nm) and attached (5 to 1,400 nm), we averaged the activity-size-to-dose conversion factors from each model to assign the dose from that fraction. The averaged weighting factors were 85 and 186 mSv y^{-1} WLM^{-1} for the unattached Jdose and Pdose fractions, respectively, and 10 and 5 mSv y^{-1} WLM^{-1} for the attached fraction. Details regarding these dose models can be found in our previous papers (Sun et al. 2009, 2010).

Data analysis

The linear mixed model was used to assess temporal as well as within-house and house-to-house variances in the radon concentrations, airborne potential alpha energy concentrations (PAECs), radon decay products, and surface-deposited radon decay products. The maximum

likelihood method was used to estimate the variance components. The house and rooms were treated as random factors, and the level of the room within the house and other environmental variables were treated as fixed factors in the analyses. The percent coefficient of variation (COV) was calculated as:

$$\mathrm{COV} = 100^{*}\mu / < V >, \quad (1)$$

where μ is the square root of the estimated variance component from the linear mixed model and $< V >$ is the average value of the variable.

Simple linear regression was used to evaluate the correlation between repeated measurements of radon concentrations, airborne PAECs, radon decay product dose rates, and surface-deposited radon decay products from different periods. Univariate analysis, using a linear mixed model, was used to identify important environmental factors affecting either spatial or temporal variations in airborne dose rates and surface-deposited radon decay products. Statistical analyses were conducted using SAS statistical software (SAS Institute Inc., Cary, North Carolina, US).

RESULTS AND DISCUSSION

Among all 98 sets of complete measurements, 26 sets of the surface-deposited radon decay product measurements had to be excluded from the analysis because their deposited ^{218}Po concentrations were below the detection limit of the RRD. Extremely low surface deposition can occur in environments where most of the decay products are attached to aerosols that have lower deposition rates than unattached decay products or where air flow rates on surfaces are low (Sun et al. 2009, 2010).

The observed variations in the measurements include a component from instrumental variation. The instrumental variations for the ATD, ARPD, and RRD detectors were estimated from side-by-side (i.e., collocated) measurements done repeatedly in our walk-in radon exposure room under a variety of radon, aerosol, and ventilation conditions as described above. The estimates for the performance of the different detectors for the conditions encountered in the homes are specified below and given in Table 2 and Table 3 footnotes.

Spatial variations

House to house.—Seventy-six original sets of measurements were used to assess the total spatial variations. Sixty-one sets of deposited ^{218}Po detectors whose track densities were likely ($p > 0.8$) to be greater than zero were used for the variations of surface-deposited radon decay products. The spatial variations for short-term and long-term radon gas, airborne PAECs, radon decay product dose rate, and surface-deposited radon decay products by smoking status are presented in Table 2. The total spatial variation in long-term radon concentrations was slightly smaller than that in short-term radon concentrations (COV 64% vs. 79%). Instrumental uncertainties in the gas measurements were approximately 15% and 10%, respectively. When the instrumental uncertainties are subtracted in quadrature from the total spatial COVs, the results are COVs of 62% and 78%. Since the homes were selected

based on high radon screening tests, the true spatial variation between Iowa houses may be higher. The airborne attached and unattached PAECs and airborne dose rates (Pdose and Jdose) all have higher total variations (COV ~100%) than the radon gas concentrations. The instrumental uncertainty in the radon decay product concentrations measured by the RRDs and ARPDs is typically on the order of 20%, which suggests that the majority of the COV comes from spatial variation. Airborne radon decay product concentrations are more variable than radon gas concentrations in homes where smoking is present. The total variation for deposited ^{214}Po was lower than the variation found for deposited ^{218}Po (COV 79% vs. 117%) and lower than the airborne PAECs as well but similar to the total variation for radon gas concentrations.

Within house.—Generally large variations exist within a house for most variables (Table 2). Unattached PAEC, Pdose, and Jdose have larger total variations than the subgroup variations in both smoking and nonsmoking categories, which may indicate that the higher aerosols in smoking rooms create additional variation within a house. It should be noted that the within-house variation for attached PAECs and dose rates were lower in homes where individuals smoked as compared to rooms where smoking did not occur, which could imply a more stable depositional environment within smoking houses where the majority of radon decay products may attach to aerosols.

Temporal variations

To assess seasonal temporal variations, the 22 rooms with repeated measurements (44 measurements) from a heating and a nonheating season were included in the analysis.

Radon concentrations in the same season

For each room in each test season, one short-term radon measurement was performed at the end of the long-term measurement period. The relationship between short-term radon measurements and long-term radon measurements in the same season is presented in Fig. 1. The modest correlation (R^2 = 0.53) is similar to results in other studies.

Seasonal variations

The seasonal temporal variations for short-term and long-term radon gas, airborne PAECs, radon progeny dose rates, and surface-deposited radon decay products by smoking status are presented in Table 3. Thirty-three deposited ^{218}Po measurements, whose track densities were likely ($p > 0.8$) to be greater than zero, were included in the temporal variation analysis. Somewhat counterintuitively, the long-term radon concentrations yielded larger temporal variation than the short-term radon concentrations (COV 55% vs. 37%, R^2 = 0.35 vs. 0.49) (Fig. 2). Further examination of the data found two outlier rooms, both belonging to the same house. The original long-term radon concentrations measured in winter for these two rooms were 230 and 360 Bq m^{-3}; the measurements in summer provided the results 37 and 19 Bq m^{-3}, yielding seasonal COVs of 103% and 128%, respectively. The whole house was closed tightly all day during the winter, but during the summer the windows were kept open. This special case illustrates that household operation can cause a large seasonal variation even in long-term domestic radon gas measurements.

The correlations between repeated seasonal PAEC measurements were not as strong as the radon correlations as shown in Table 3 and Fig. 3. Attached PAECs had slightly better correlation than unattached PAECs (COV 61% vs. 67%, R^2 = 0.3 vs. 0.2), which could explain the better correlation between repeated Jdose as compared to Pdose (COV 46% vs. 60%, R^2 = 0.47 vs. 0.27) as shown in Table 3 and Fig. 4. Similar to the spatial variation findings, surface-deposited ^{214}Po also displayed less temporal variation as compared to deposited ^{218}Po and airborne PAECs as shown in Table 3 and Fig. 5. A correlation plot was not performed for deposited ^{218}Po due to the large fraction of missing values. For all variables listed in Table 3, the temporal variations noted for the rooms where individuals smoked were less than those in rooms where smoking did not occur.

Environmental factors affecting the between-room and within-room variations

As shown in Table 4, the between-room variance in Pdose and Jdose was reduced by adding smoking to the model. The residual variance, which includes both temporal variation and remained variation, showed the same reduction when smoking was included in the model. The between-room variation is likely attributable to the inherent differences in aerosol conditions between smoking rooms and nonsmoking rooms. The within-room variation had a significant contribution from a single case where smoking was present during the initial testing but not during the second measurement because the individual who smoked had died. Fan and fireplace usage could also explain some of the temporal (or within-room) variation in both Pdose and Jdose, but it could not reduce the between-room variance in the model because fan and fireplace usage has obvious seasonal patterns. However, room level and room type could only reduce between-room variance since they are fixed.

The surface-deposited ^{214}Po variation behaved the same way as Pdose and Jdose variations for smoking, fireplace usage, and room level (Table 5). Opening windows and candle usage could reduce both between-room variance and temporal variance in the model because they are relevant to indoor room aerosol changes and have seasonal patterns. For example, householders tend to open windows more frequently in summer and burn candles more often during fall and winter holidays.

The effects of the environmental factors selected by the current analysis should be interpreted cautiously since the statistical power is limited by the small sample size (22 rooms, 2 measurements per room).

CONCLUSION

Few studies have been published of the measured temporal and spatial variations of radon decay products in large samples of US houses compared to the extensive literature on indoor radon gas variations. Our results provide insights on the complex relationship between radon, radon decay products, surface-deposited radon decay products, and radon-related dose in occupied homes. The substantial radon decay product concentration variations from house to house as well as within a house and from season to season indicates that multiple locations and time integrated measurements are needed to make an accurate assessment of chronic radon-related doses. A limitation of the study is the small number of sampled homes and rooms; therefore, it is not scientifically defensible to broadly generalize the results of

this study especially to other geographic regions or housing stocks. Nonetheless, the findings may provide important insights for developing research methods for future radon-related epidemiologic investigations.

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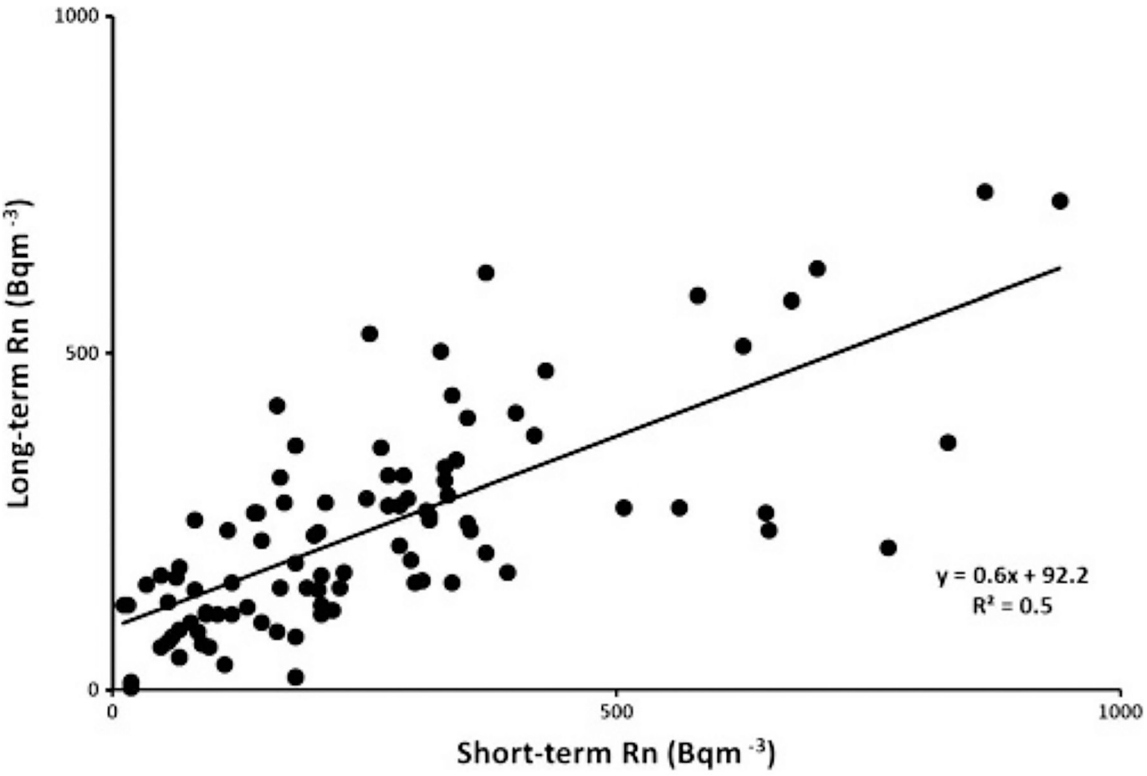

Fig. 1.

Correlation between short-term (7 d) and long-term (>90 d) radon concentrations (Bq m^{-3}) measured in winter and summer seasons.

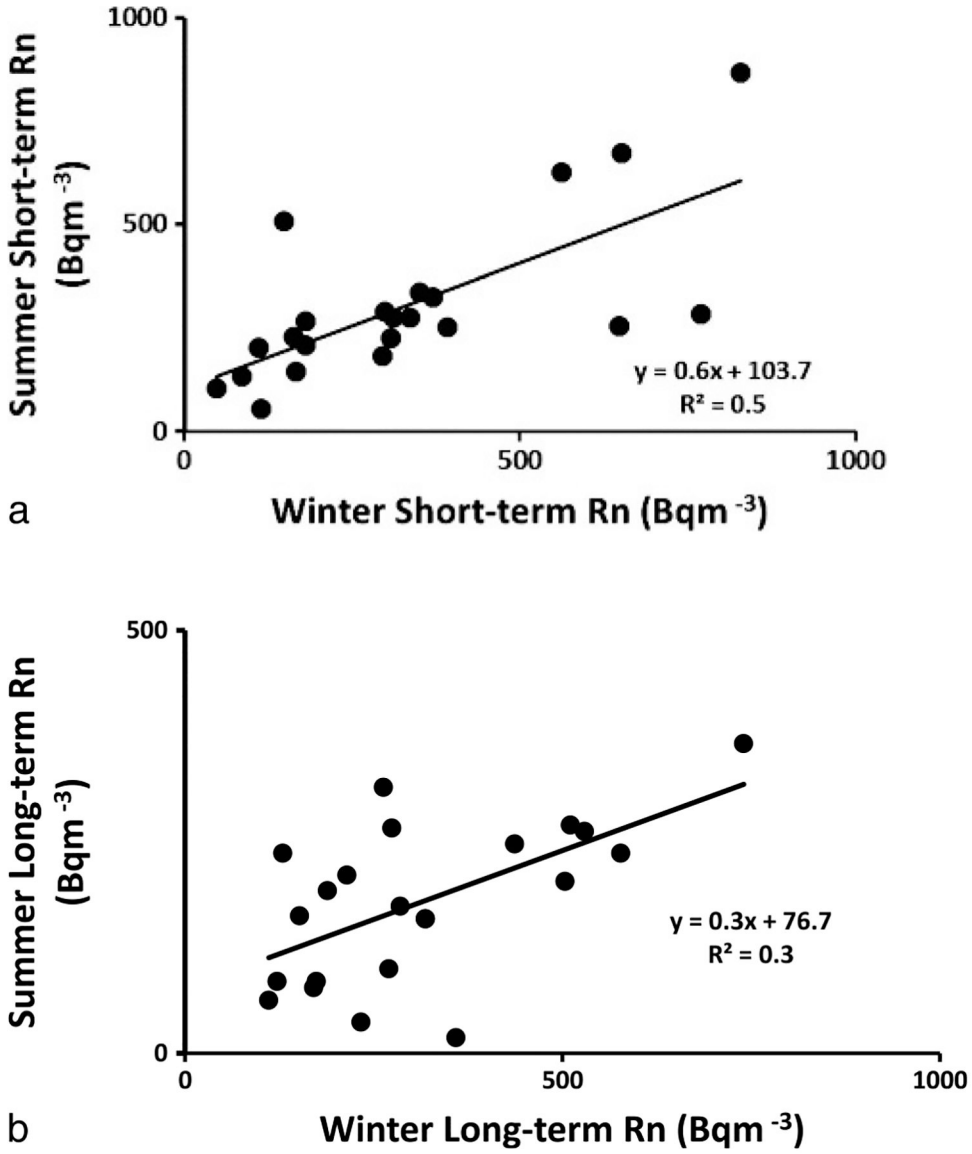

Fig. 2.

Correlation between winter and summer radon concentrations (Bq m^{-3}) for (a) short-term measurement and (b) long-term measurements in 22 rooms of 11 houses.

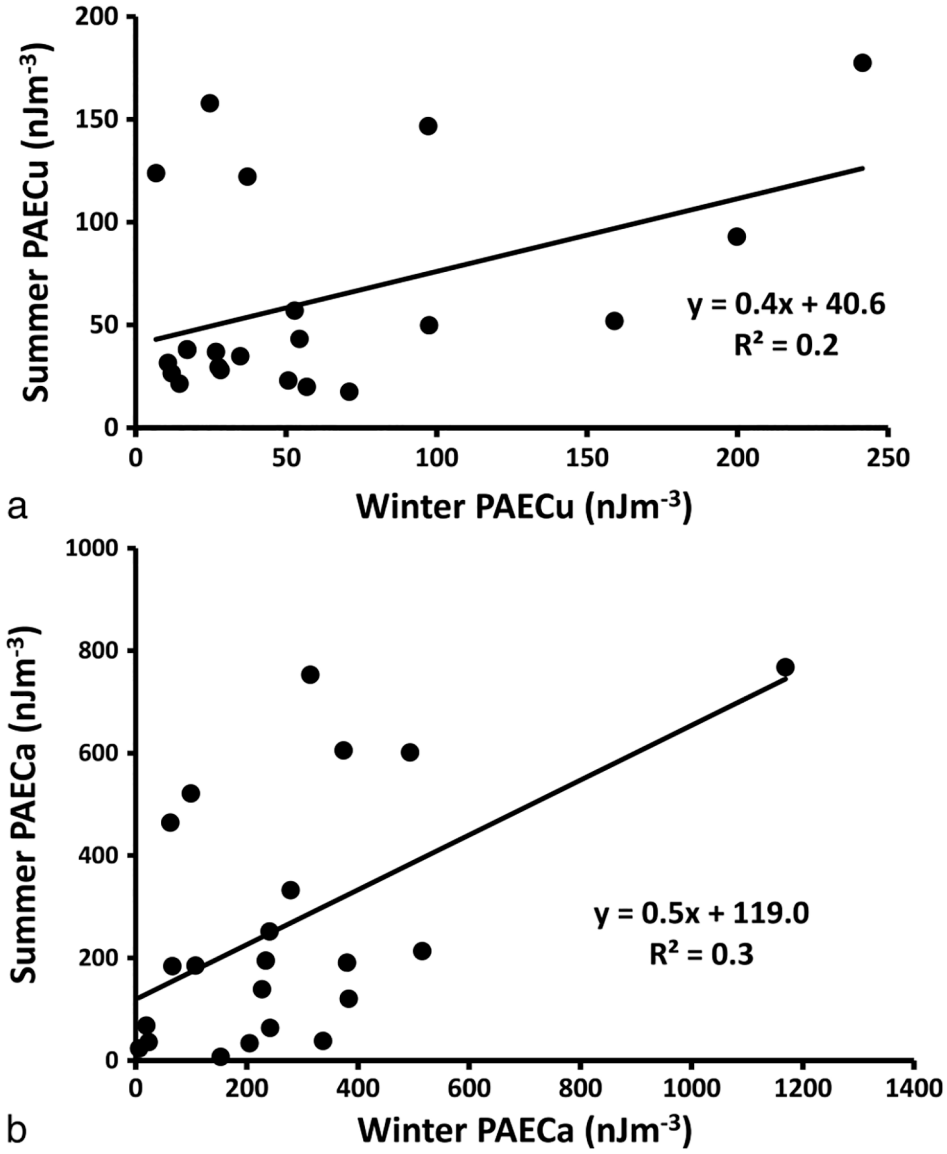

Fig. 3.

Seasonal variations of airborne radon decay product potential energy concentration (nJ m^{-3}) by size for (a) aerosol unattached and (b) aerosol attached.

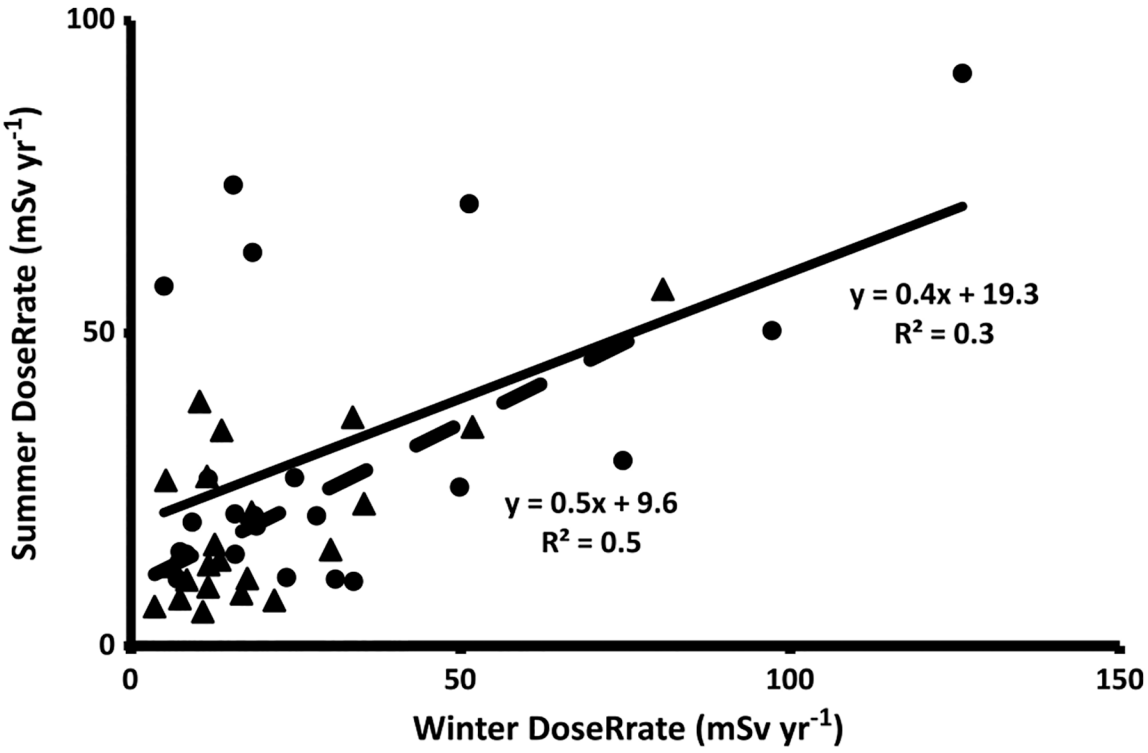

Fig. 4.

Seasonal variations of dose rates (mSv y^{-1}) estimated from airborne radon decay products using two dose models (Pdose shown as circles; Jdose shown as triangles).

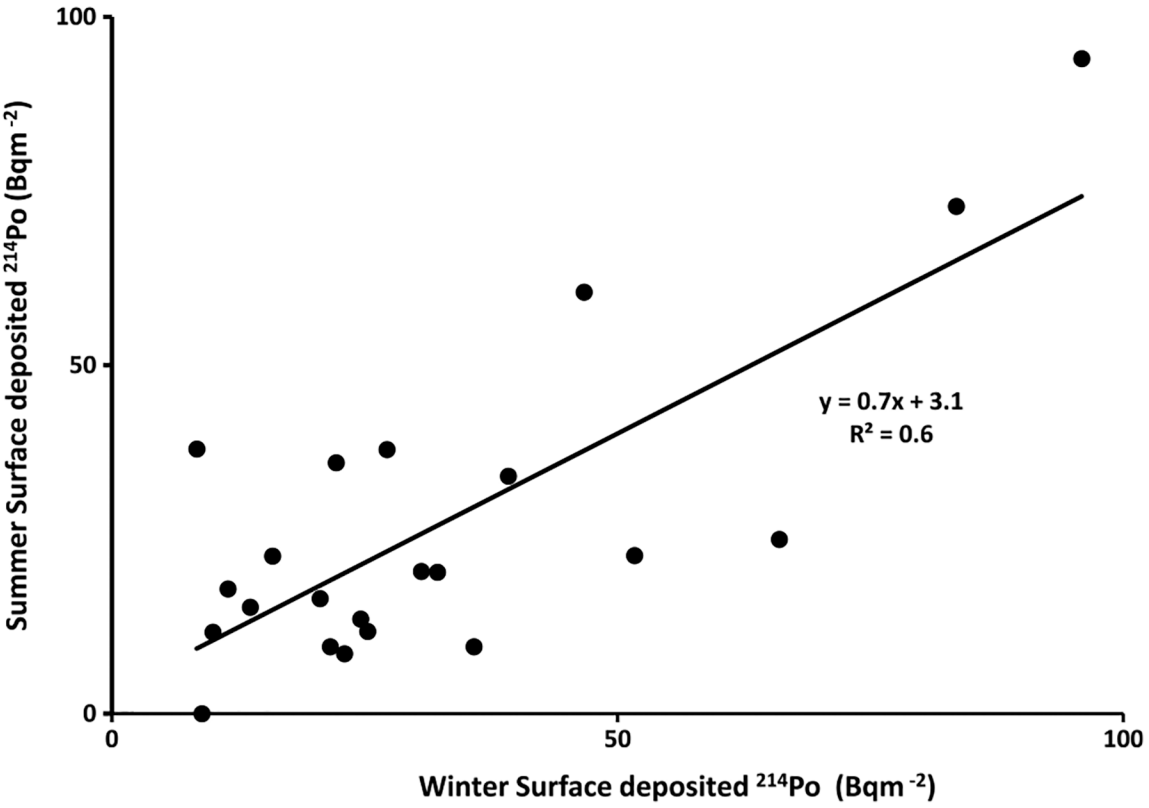

Fig. 5.
Seasonal variation of surface-deposited ^{214}Po (Bq m^{-2}).

Table 1.

Radon-related variables measured in each home with two occupied rooms that had a clean, unobstructed glass surface.

Variable name[a]	Measured	Integration/duration	Season	Detector type
Long-term radon	Radon gas concentration	90 d	Winter-spring/summer-fall	Alpha track detector (ATD)
Deposited ^{218}Po, deposited ^{214}Po	Surface-deposited concentrations	90 d	Winter-spring/summer-fall	Retrospective radon detector (RRD)
Short-term radon	Radon gas concentration	3 to 7 d	End of each 90-d period	Electronic ion chamber (EIC)
Unattached PAEC	^{218}Po, ^{214}Po collected on screen (<5 nm)	3 to 7 d	End of each 90-d period	Airborne radon progeny detector (ARPD)
Attached PAEC	^{218}Po, ^{214}Po collected on filter (>5 nm)	3 to 7 d	End of each 90-d period	Airborne radon progeny detector (ARPD)

[a]The variables Pdose and Jdose were calculated from the unattached and attached PAEC (Sun et al. 2009).

Table 2.

The mean values and spatial coefficient of variation (COV) for short-term and long-term radon gas, airborne PAECs, dose rate, and surface-deposited radon decay products by smoking environment.

Variables	Smoking environment (n)	Mean value	COV,[a] total %	House to house, %	Within house,[b] %
Short-term radon (Bq m^{-3})	Nonsmoking (44)	302	68	48	43
	Smoking (32)	150	72	65	12
	Total (76)	**238**	**79**	**62**	**42**
Long-term radon (Bq m^{-3})	Nonsmoking (44)	313	57	40	34
	Smoking (32)	179	60	53	12
	Total (76)	**257**	**64**	**51**	**32**
Unattached PAEC (nJ m^{-3})	Nonsmoking (44)	102	86	59	53
	Smoking (32)	34	84	55	50
	Total (76)	**74**	**105**	**77**	**58**
Attached PAEC (nJ m^{-3})	Nonsmoking (44)	376	138	69	99
	Smoking (32)	368	100	84	54
	Total (76)	**374**	**124**	**72**	**86**
Pdose (mSv y^{-1})	Nonsmoking (44)	38.4	87	57	54
	Smoking (32)	14.4	83	63	43
	Total (76)	**28.3**	**103**	**74**	**58**
Jdose (mSv y^{-1})	Nonsmoking (44)	23.0	93	54	61
	Smoking (32)	11.9	85	73	39
	Total (76)	**18.3**	**100**	**66**	**61**
Deposited ^{218}Po (Bq m^{-2})	Nonsmoking (40)	43.1	116	36	105
	Smoking (21)	24.9	76	60	68
	Total(61)	**36.8**	**117**	**43**	**106**
Deposited ^{214}Po (Bq m^{-2})	Nonsmoking (40)	31.8	74	53	38
	Smoking (21)	17.3	64	37	50
	Total(61)	**26.8**	**79**	**57**	**42**

[a]The instrumental uncertainties in radon gas concentrations are typically about 10 to 15%. The uncertainties in airborne radon and deposited radon decay product concentrations are in the range of 20 to 25%.

[b]The results for within-house COV have been adjusted for the fixed effect of a room's level within a house.

Table 3.

The mean values and temporal coefficient of variation (COV) for short-term and long-term radon gas, airborne PAECs, dose rate, and deposited radon decay products by smoking environment.

Variables	Smoking environment (n)	Mean values	Variation between winter and summer measurements, %
Short-term radon (Bq m^{-3})	Nonsmoking (35)	348	37
	Smoking (9)	207	15
	Total (44)	**319**	**37**
Long-term radon (Bq m^{-3})	Nonsmoking (35)	249	58
	Smoking (9)	232	31
	Total (44)	**246**	**55**
Unattached PAEC (nJ m^{-3})	Nonsmoking (35)	96	60
	Smoking (9)	31	44
	Total (44)	**85**	**67**
Attached PAEC (nJ m^{-3})	Nonsmoking (35)	340	65
	Smoking (9)	450	51
	Total (44)	**362**	**61**
Pdose (mSv y^{-1})	Nonsmoking (35)	36.2	54
	Smoking (9)	14.3	46
	Total (44)	**31.7**	**60**
Jdose (mSv y^{-1})	Nonsmoking (35)	21.5	43
	Smoking (9)	13.0	44
	Total (44)	**19.8**	**46**
Deposited ^{218}Po (Bq m^{-2})	Nonsmoking (25)	51.2	112
	Smoking (8)	28.8	88
	Total (33)	**45.8**	**115**
Deposited ^{214}Po (Bq m^{-2})	Nonsmoking (25)	30.1	53
	Smoking (8)	18.5	10
	Total (33)	**27.3**	**48**

Table 4.

(a) Univariate analyses of environmental factors as fixed factors affecting variation in Pdose.
(b) Univariate analysis of environmental factors as fixed factors affecting variation in Jdose.

Variable	*p* value for *F* test of fixed effect[a]	Between-room variance	Residual variance
—	—	377	367
Humidifier	0.002	163	383
Room type	0.01	87	367
Smoke	0.03	348	321
Level	0.05	282	367
Fan	0.12	374	338
Fireplace	0.15	389	336

Variable	*p* value for *F* test of fixed effect[a]	Between-room variance	Residual variance
—	—	159	82
Humidifier	0.001	87	76
Room type	0.03	63	82
Fireplace	0.03	172	65
Level	0.08	133	82
Smoke	0.09	156	74
Fan	0.17	161	75

[a]Fixed effects with p value < 0.2 were included in the table.

Table 5.

Univariate analysis of environmental factors as fixed factors affecting variation in surface-deposited ^{214}Po.

Variable	*p* value for *F* test of fixed effect[a]	Between-room variance	Residual variance
—	—	387	132
Open window	0.03	366	113
Smoke	0.05	378	115
Level	0.05	313	132
Fireplace	0.10	400	115
Candle	0.12	365	125

[a]Fixed effects with p value < 0.2 were included in the table.