



Radon transfer from groundwater used in showers to indoor air

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ABSTRACT

Estimation of Rn transfer from water to indoor air based on multi-day measurements may underestimate alpha exposure that occurs at short time scales in confined spaces, such as from showering, in houses with high Rn activities in the water supply. In order to examine one such incremental increase in exposure, variations in Rn in water and indoor air in 18 houses with private wells in western North Carolina (USA) were investigated. Radon in well water ranged from 158 to 811 Bq L⁻¹ (median 239 Bq L⁻¹). After 20-min showers in bathrooms with closed doors, peak Rn in air increases (above background) ranged from 71 to 4420 Bq m⁻³ (median 1170 Bq m⁻³). Calculated transfer coefficients at the scale of a 40-min closed bathroom (20-min shower plus 20 min post-shower) are described by a lognormal distribution whose geometric mean exceeds the widely-used $\sim 10^{-4}$ whole-house transfer coefficient by about one order of magnitude. As short-lived decay products grow from shower-derived Rn, short-term alpha energy exposure occurs in bathrooms in addition to the exposure caused by Rn mixed throughout the volume of the house. Due to the increasing ratio of Rn decay products to Rn, alpha energy exposure is greatest several minutes after the shower is turned off. For a 7.2-min shower with 10 min of additional exposure before opening the door, a geometric mean 5.6% increase in exposure over the $\sim 10^{-4}$ whole-house transfer coefficient derived from longer measurement periods was estimated. In addition to Rn activity in water, short-term shower exposure to Rn progeny depends on exposure time, ventilation, attachment and deposition, among other variable factors that characterize individual houses and residents.

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1. Introduction

Naturally occurring radon-222 (Rn) in indoor air is a significant cause of lung cancer (Krewski et al., 2006), and houses are an important exposure location due to the large proportion of time spent at home. Radon is believed to be the second-largest cause of lung cancer, after smoking, and may cause an estimated 21,100 lung cancer deaths/a in the United States (Environmental Protection Agency (EPA), 2003). Although direct transfer from soil and rock is the largest aggregate exposure pathway, Rn can degas from water into buildings and constitutes an important secondary source of Rn exposure in groundwa-

ter-dependent populations. Although both inhalation and ingestion of Rn-rich water may represent some exposure to the lungs and stomach, respectively, inhalation is believed to be a larger health risk (National Research Council (NRC), 1999). Radon exposure is typically evaluated using a linear, no-threshold model (Samet, 2006) so that all exposures to Rn are potentially significant, and treatment of water supplies containing high Rn activities may be an effective way to reduce Rn exposure in some cases.

Elevated levels of Rn in groundwater are widespread in many areas of the USA (Focazio et al., 2006) and especially in areas underlain by felsic crystalline rocks (Table 1). When high-Rn groundwater is utilized in homes, clothes washing, dishwashing, sink usage and showering can cause 28–98% of dissolved Rn to degas (Partridge et al., 1979), and mixing of this Rn into the volume of the house creates

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Table 1Radon in water activities in selected areas of the United States. Radon activities in Bq L⁻¹. (1 Bq L⁻¹ = 26.67 pCi L⁻¹)

Region	Rock type	Median or geometric mean ²²² Rn, range in parentheses (Bq L ⁻¹)	Reference
Georgia	Granitic gneiss	230 (4.8–31,000)	Dillon et al., 1991
South Carolina	Crystalline rocks	84 (15–2197)	King et al., 1982
North Carolina	Granite	218 (19–1623)*	Loomis, 1987
Virginia	Granite	287 (37–901)	Harris et al., 2006
New Jersey	Granite	107 (<4–463)	Wanty et al., 1991
Pennsylvania	Crystalline rocks	52 (2–1961)	Senior, 1998
Maine	Granite	1208 (22–4514)	Brutsaert et al. 1981
Iowa	Sedimentary rocks	12 (1–87)	Field and Kross, 1998
Area of this study	Meta-intrusive	277 (27–1687)	Campbell, 2006
	Meta-sedimentary	149 (64–529)	Campbell, 2006

* Range is 5th–95th percentiles.

incremental increases in exposure to Rn progeny over the relatively long (multi-day or longer) measurement periods typically employed in residential Rn exposure calculations.

It has been noted previously that this single-cell Rn transfer model of water supply contribution to whole-house average Rn underestimates exposure because occupants are in proximity to water uses and because daily lows in airborne Rn activities occur when residents are not at home (e.g. Gesell and Prichard, 1980). However, the most commonly used basis for exposure estimation is at the scale of the entire house and over relatively long measurement periods (days or longer; NRC, 1999). At those spatial and temporal scales, transfer of Rn into homes is commonly modeled by a transfer coefficient, C_T

$$C_T = \frac{\text{Increment of airborne radon added by water}}{\text{Radon activity in water}} \quad (1)$$

Estimates of C_T at whole-house scales ($C_{T,H}$) have consistently centered on approximately 10^{-4} within a large range of variability (Hess et al., 1982,1990; Nazaroff et al., 1987; NRC, 1999).

Other research has documented the effects of specific water uses, such as showering, at short temporal and spatial scales, before Rn mixes throughout the house (McGregor and Gourgou, 1980; Bernhardt and Hess, 1996; Fitzgerald et al., 1997). Use of Rn-rich water results in short-term exposures to humans that may not be accounted for by multi-day, whole-house measurements. The purpose of this study is to quantify the effect of showering, a routine water use that occurs in enclosed spaces. Details of relevant physical processes, such as effects of shower heads, ventilation, attachment of Rn progeny to particles and deposition of progeny onto surfaces, have been simulated in laboratory (Partridge et al., 1979; Fitzgerald et al., 1997) and modeling (Datye et al., 1997; Nikolopoulos and Vogianis, 2007) experiments. Variations in these parameters can result in several orders-of-magnitude changes in net transfer of Rn from water to an indoor space and mobility of its decay products. Rather than to control the processes involved (e.g. degassing, attachment, deposition and escape, which were beyond the scope of the field measurements), the objective of this paper is to document net transfer under real-world conditions, in which orders-of-magnitude variations in Rn transfer characteristics are expected between houses (e.g. Hess et al., 1990). By examining typical homes, a distribution of net Rn transfer

characteristics may be determined, and the potential exposure to Rn decay products can be estimated.

The selected houses use private wells in fractured crystalline rocks of western North Carolina (Fig. 1), an area known to produce high levels of Rn in indoor air and groundwater. Previous sampling in the study area indicated that Rn in water is strongly associated with rock type. Meta-igneous rocks, including granitic gneiss and granodiorite, exhibit higher median Rn levels than metasedimentary rocks, including garnet-mica schist, mica gneiss, and rocks of the Brevard Fault Zone (Table 1). In contrast to these high Rn activities, U and Ra levels were generally below drinking water standards throughout the study area (Campbell, 2006). Sixteen of the 18 wells selected for this study were included in the previous study, and 15 of those 16 exhibited levels of Rn in water in excess of the EPA-proposed alternate maximum contaminant level of 148 Bq L⁻¹ (Campbell, 2006).

2. Methodology

Eighteen houses with private wells were visited between February and August 2006. Most wells are completed in granitic gneiss, and a few are in other rock types including granodiorite, amphibolite, and mica schist (Table 1). At the well head sampling tap (where present), a water sample was collected for ²²²Rn by submerging a glass vial into a glass beaker receiving a low flow of minimally aerated water, then sealing the submerged vial. Radon-in-water samples were rushed to a commercial laboratory and typically analyzed within 24–36 h of collection by liquid scintillation counting. Radon activities were decay-corrected to the time of collection.

Radon in bathroom air was determined using a Durridge RAD7 alpha counting instrument in 5-min counting intervals, with the sampling point at floor level approximately 1 m from the shower. The instrument operated at a flow rate of 0.7 L min⁻¹ through a 0.45 µm inlet filter into a chamber of internal volume ~1 L. The RAD7 was calibrated by the manufacturer in September 2005 (efficiency 0.006027 ± 0.000241 Bq m⁻³ cpm⁻¹) and September 2006 (efficiency 0.006514 ± 0.000261 Bq m⁻³ cpm⁻¹; counting errors are ±2σ). The RAD7 counts alpha decays of ²¹⁸Po, the daughter of ²²²Rn, generated only by in situ decay, providing a measurement of ²²²Rn gas exclusive of Rn progeny. Under static conditions, the RAD7 equilibrates

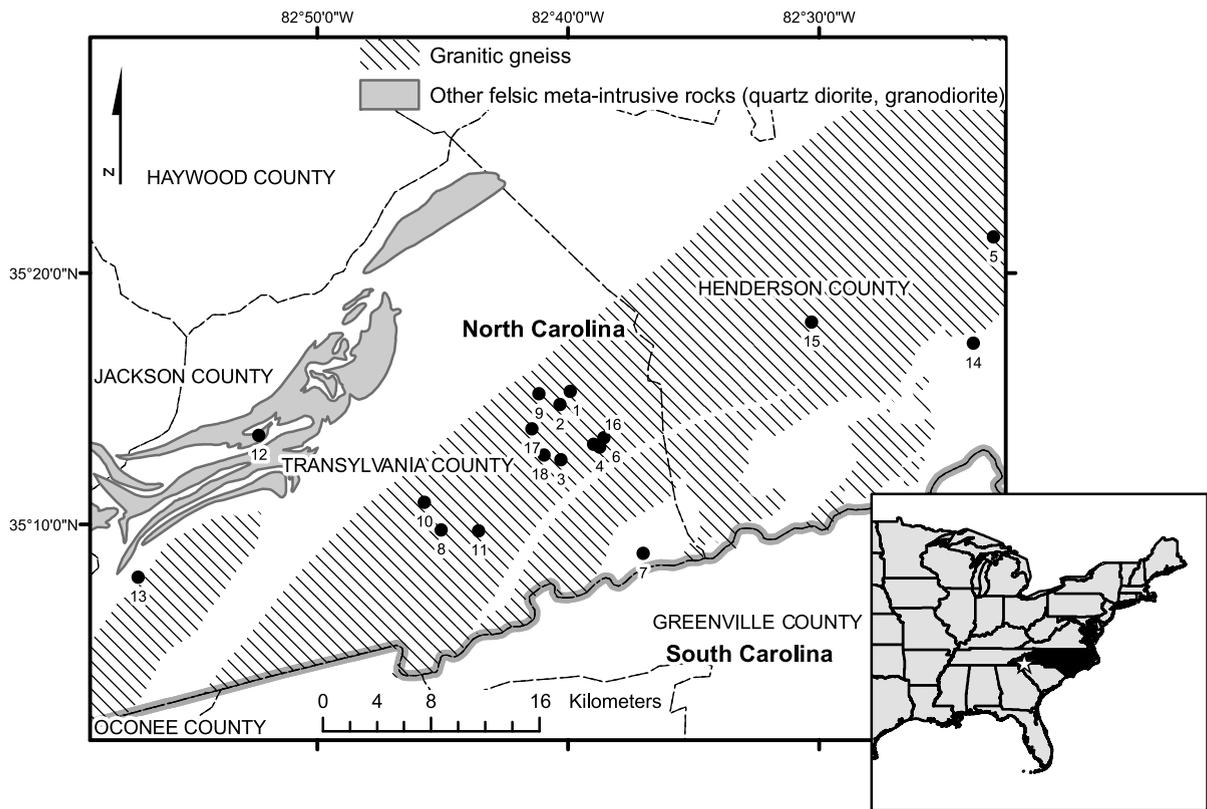


Fig. 1. Map showing the study area (Henderson and Transylvania counties). Geographic layers obtained from www.nationalatlas.gov; geological data modified from North Carolina Geological Survey (1985).

with the room air in approximately 10 min, controlled by the ~ 3 -min half-life of ^{218}Po . Thus, under dynamic conditions, the RAD7 count represents a moving average of the past ~ 10 min of Rn activities. The RAD7 measured ambient Rn levels for 5–15 min before the water was turned on. Background was established as the highest 5-min interval during the pre-shower period because of the possible isotopic disequilibrium inside the RAD7. For each 5-min counting period, the 2σ counting error calculated by the RAD7 was used, and a standard propagation of error equation (Clesceri et al., 1998) was applied for subtraction of the pre-shower airborne Rn measured in the room.

To conduct the experiment, a typical warm shower was operated for 20 min at approximately the same time that the well water samples were collected, and RAD7 measurement continued with the door closed for at least 30 min after the water was turned off. The shower curtain or stall door was left as open as possible to encourage mixing of air in the room, but closed as necessary to avoid spilling water. Bathroom doors were kept closed and only opened as necessary to enter or leave to monitor the experiments. Ventilation fans, where present, were not used, and windows were kept closed. During some tests, water temperature was measured continuously using a Hobo data logger suspended in the stream of water. Typically after the 20-min shower had ended, warm tap water was collected at the sink or bathtub by submerging a glass vial in a beaker filled with minimally aerated tap water, and warm shower water

was collected at that time by holding a glass vial into the shower water and capping without bubbles. Statistical handling of data and construction of a decay simulation were performed using the R software package (R Development Core Team, 2007).

3. Results

Radon in well water ranged from 158 to 811 Bq L^{-1} and had a median of 239 Bq L^{-1} . Radon in warm tap water ranged from 59 to 715 Bq L^{-1} , with a median of 150 Bq L^{-1} , not including one house with a Rn aeration system, in which the indoor tap Rn was less than the laboratory reporting limit of 3 Bq L^{-1} . At most houses, a relatively consistent decline is observed between the cold well water and the warm indoor tap water and thus well water and indoor tap water Rn are well correlated. Linear regression ($R^2 = 0.93$) indicates a slope of 1.05 and an intercept of -102 Bq L^{-1} (Fig. 2). Thus, as well water flows into the household plumbing and hot water system, it experiences an average absolute decline of approximately 100 Bq L^{-1} Rn regardless of the original activity (excluding one house with a Rn treatment system). Radon in warm shower water ranged from 24 to 390 Bq L^{-1} , with a median of 124 Bq L^{-1} . In the experimental showers, peaks of Rn in air were measured that reflect a transition from Rn accumulation in the room as it degasses from water to depletion as it escapes from the room (Fig. 3). Peak recorded levels of Rn in indoor

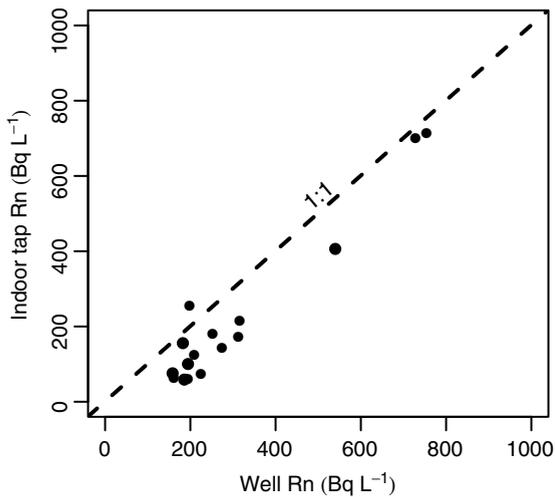


Fig. 2. Comparison of radon in cold well water and warm indoor tap water.

air ranged from 71 to 4420 Bq m⁻³ above background, with a median of 1170 Bq m⁻³. In almost all cases, $\pm 2\sigma$ counting error of the peak Rn level in air was outside the $\pm 2\sigma$ counting error of the pre-shower background level. The 40-min average of the eight 5-min Rn in air counting periods com-

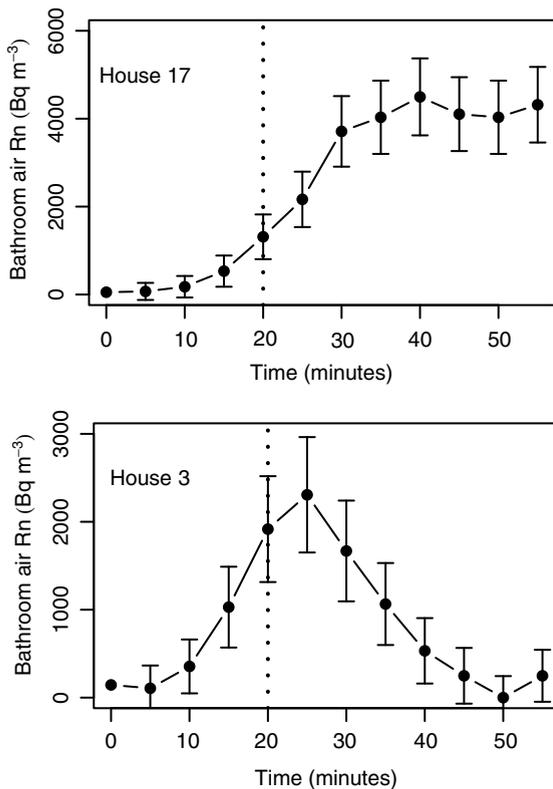


Fig. 3. Results from two shower experiments (prior to background subtraction) illustrating differences in peak size and timing after 20-min showers. Error bars represent $\pm 2\sigma$ counting error.

prising a 20-min shower plus the subsequent 20 min, after background subtraction, ranged from 84 to 1990 Bq m⁻³, with a median of 591 Bq m⁻³, excluding one slightly negative average and one house with a Rn-in-water treatment system (Table 2).

4. Discussion

4.1. Airborne Rn accumulation during and after experimental showers

The consistent decline in Rn from the well to the warm indoor tap sample (Fig. 2) is probably the result of degassing and/or decay losses in the water heating system (McGregor and Gourgon, 1980). Due to the storage capacity of a home hot water system, warm water may require 12–24 h to fully respond to a large change in Rn activity in the water supply (Lowry et al., 1987). Significant changes in Rn activity may be caused by pumping conditions; for example, continuous pumping of an initially stagnant well may cause Rn activity to increase (Cook et al., 1999), or drawdown may shift the well's contributions between fractures of significantly different Rn activities (Lawrence et al., 1991). The cold well sample is an instantaneous sample, but the warm indoor water is time-integrated due to the residence time of the hot water system, and may represent a mix of pumping conditions. That the warm indoor water samples, collected after the well sample and generally after the shower was turned off, exhibit consistently lower Rn activities than the well samples, implies that the difference in Rn activity is mostly due to decay and degassing losses in the plumbing system, rather than large changes in Rn in influent well water. Although shower hardware is expected to release a significant portion of Rn in water, shower water samples exhibited only modestly lower Rn activities, on average, than indoor tap water, and 6 shower samples had higher activity than the corresponding tap water samples (Table 2). Thus, collection methods did not successfully quantify the degree of degassing caused by showers; however, warm indoor tap water contained less Rn than cold well water in most cases.

For Rn carried into the bathroom at a constant rate during the shower, Rn in air should increase steadily until the peak is reached (Fig. 3); thus, the peak value approximates the amount of Rn added to the room by the shower, minus the Rn that escaped the room prior to the peak. Overall, there is no direct relationship between Rn in water and peak Rn activity in air after a shower (Fig. 4). This suggests that other processes, not controlled in these experiments, are significant controls on airborne Rn activities in these bathrooms. These include ventilation (Fitzgerald et al., 1997), differences between shower heads (Partridge et al., 1979) and water flow rates, and mixing into bathrooms of different volumes. Laboratory experiments with shower ventilation indicate that strong ventilation causes peak Rn levels to occur sooner than poorly ventilated rooms, which exhibit later, larger peaks. More ventilation also results in better mixing of Rn throughout the volume of the room (Fitzgerald et al., 1997), so airborne Rn measurements in these rooms should be more representative

Table 2

Radon results for water and air

House number	Rock type	²²² Rn at well (Bq L ⁻¹)	Temp. (°C)	²²² Rn at indoor tap (Bq L ⁻¹)	Temp. (°C)	²²² Rn in shower water (Bq L ⁻¹)	Temp. (°C)	Shower air peak Rn (Bq m ⁻³)	40-min avg air Rn increase (Bq m ⁻³)	Time to peak from start of shower (min)	Experiment water temperature range (°C)	40-min C _{T,B}
1	Granitic gneiss	316	13.1	215 ^a	14.0	229	14.1	550 ± 293	271	45	16–21 ^f	0.00086
2	Granitic gneiss	728	13.3	700 ^c	23.1	382	23.1	603 ± 465	244	40		0.00033
3	Granitic gneiss	540	13.5	406 ^c	30.9	176	30.9	2090 ± 705	909	25		0.00168
4	Granitic gneiss	557	13.2			245	46.0	923 ± 589	510	25		0.00092
5	Granitic gneiss	252	13.8	180 ^c	38.2	150	38.2	1350 ± 690	754	30	28–34	0.00299
6	Granitic gneiss	186	13.5	59 ^c	42.4	24	42.4	284 ± 363	84	25	52	0.00045
7	Garnet-mica schist	195	13.0	62 ^c	38.8	104	38.8	71 ± 301	^d	20	47–51	
8	Granitic gneiss	754	14.4	715	35.9	390	36.7	2170 ± 794	872	35	34–36	0.00116
9	Granitic gneiss	194	15.2	100	36.5	80	39.3	535 ± 404	192 ^e	35	30–34	0.00099
10	Granitic gneiss	158	17.2	76	34.3	94	33.4	678 ± 434	129	55	27–40	0.00082
11	Granitic gneiss	312	15.7	172	25.3	155	31.7	1380 ± 553	697	35		0.00223
12	Quartz diorite/granodiorite	162	16.2	63	31.4	76	30.1	710 ± 439	390	25		0.00241
13	Amphibolite	274	14.4	144	36.9	110	36.2	2000 ± 688	871	40		0.00318
14	Porphyroblastic gneiss	198	14.9	256	22.5	320	22.8	1530 ± 560	568	25	25–30	0.00287
15	Granitic gneiss	811	15.9	<3 ^b	42.0		42.0	106 ± 287	^d	40	31–38	
16	Granitic gneiss	182	13.6	156	33.7	137	33.5	1170 ± 546	617	25	32–34	0.00339
17	Granitic gneiss	225	14.7	73	30.3	139	27.5	4420 ± 897	1990	40	30–44	0.00884
18	Granitic gneiss	209	15.3	123	41.5	98	40.2	1310 ± 621	736	30	22–29	0.00352
	Minimum	158	13.0	59	14.0	24	14.1	71	84	20		0.00033
	Maximum	811	17.2	715	46.0	390	46.0	4420	1990	55		0.00884
	Median	239	14.4	150	35.1	124	34.9	1170	592	33		0.00196
	Geo. mean	295		148		122		928	467			0.00165

Counting errors are ±2σ.

^a Sample collected at kitchen sink.^b Rn in water aeration system in place. This house excluded from median and geometric mean calculations for all except well water.^c One temperature measurement made while sampling indoor tap and shower water. Temperature is assumed to be same for both samples.^d Negative average; excluded from transfer coefficient calculations.^e Room opened prematurely during experiment.^f Experiment used cold water; all other experiments used warm water.

coefficients over the volume of the bathroom, $C_{T,B}$, using the average airborne Rn increase over background during the 20-min shower plus the first 20-min post-shower, divided by the Rn activity in well water as indicated by Eq. (1) (Table 2). Because these transfer coefficients incorporate the average of 40 min of airborne Rn measurement, they are time-integrated in contrast to the peak values of airborne Rn reported in Fig. 4, which are averaged over only 5 min. Well water Rn activity was used rather than warm indoor water activity in order to compare $C_{T,B}$ to $C_{T,H}$ for the same source of water that supplies the entire house. This treats the hot water system as a Rn dilution mechanism similar to ventilation. Use of the warm indoor tap water data would have led to larger estimates of $C_{T,B}$. The approach necessitated removal of two data points: one with a slightly negative, within-counting error negative transfer and another from a house with a Rn-in-water treatment system and no significant airborne Rn increase caused by the shower. In order to generalize across bathrooms, incorporating ventilation and other uncertainty into a probability distribution, it was assumed that the transfer coefficient is lognormally distributed. This is consistent with the lognormal models used for whole-house Rn transfer from water to air (e.g. NRC, 1999). Examination of $\ln C_{T,B}$ on a normal quantile–quantile plot (Dalgaard, 2002) also suggests that this assumption is reasonable given the sample size.

The resulting $C_{T,B}$ values (geometric mean = 1.65×10^{-3} , geometric standard deviation = 0.86) are about one order of magnitude higher than whole-house transfer coefficients (Fig. 5). These high transfer coefficients are the result of high airborne Rn, small spatial scale and short temporal scale. Because a transfer coefficient value is influenced by the spatial and temporal scale of measurement, Rn transfer experiments that do not simulate actual exposure conditions may not accurately quantify exposure. Whole-house transfer coefficients vary because of large variations in house volume, construction methods, and ventilation. The considerable variability observed in $C_{T,B}$

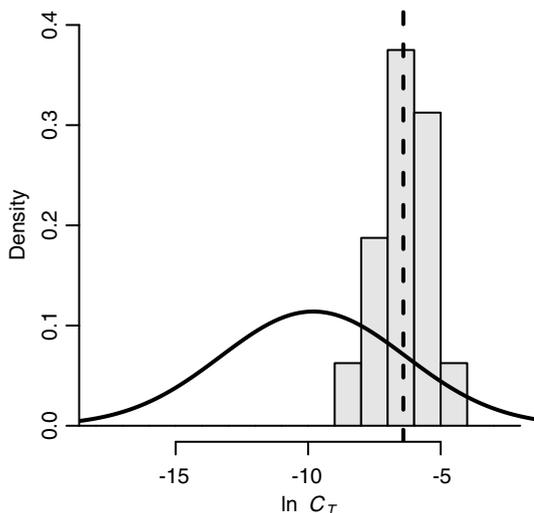


Fig. 5. Histogram of $\ln C_{T,B}$ (this study) in relation to density of $\ln C_{T,H}$ (NRC, 1999).

is less than the variability in $C_{T,H}$ determined by a recent data synthesis (geometric mean = 0.55×10^{-4} , geometric standard deviation = 3.5; NRC, 1999).

4.3. Implications for exposure

Because the health risk from Rn is posed by inhalation of its short-lived alpha-emitting decay products, comparison of transfer coefficients alone does not provide information about alpha energy exposure because high-Rn tap water contains much lower activities of Rn decay products (Bernhardt and Hess, 1996; Fitzgerald et al., 1997). Therefore, decay time is needed for exposure to be significant, that is, for the equilibrium factor (F ; i.e. the alpha energy-weighted activity ratio of airborne Rn progeny to Rn; NRC, 1991) to increase from near zero. Here F is referred to at the scale of the entire house as F_H and at the bathroom scale as F_B . The ingrowth of Rn decay products, depicted in Fig. 6 for a hypothetical sealed room and neglecting escape, attachment, and deposition, indicates that F_B of nuclides delivered by the shower is relatively low during the actual use of shower water, and F_B continues to increase after the water has been turned off. Thus, the highest airborne Rn (at the end of water usage) will not coincide with the highest exposure (after the water is turned off). F_B depends directly on deposition rates to surfaces and indirectly on attachment rates to aerosols, which may differ between the Rn decay products ^{218}Po , ^{214}Pb and ^{214}Bi (Nikolopoulos and Vogianis, 2007). Thus, actual F_B is expected to be lower than depicted in Fig. 6. Published field measurements (calculated from Fig. 6 of Fitzgerald et al., 1997) indicate a F_B range from 0.06 just before the end of a 15-min shower to 0.24 about 28 min after the shower was turned off, reaching values of 0.33–0.43 beyond 1 h. In a laboratory with manipulated ventilation, F_B followed similar trends and increased to 0.41–0.69 after more than 1 h (Fitzgerald et al., 1997). In longer-term measurements corresponding with multi-day exposure periods, F_H at assumed steady state is generally in the range 0.4–0.5 (NRC, 1991; EPA, 2003). Using an average value of F , the equilibrium equivalent concentration

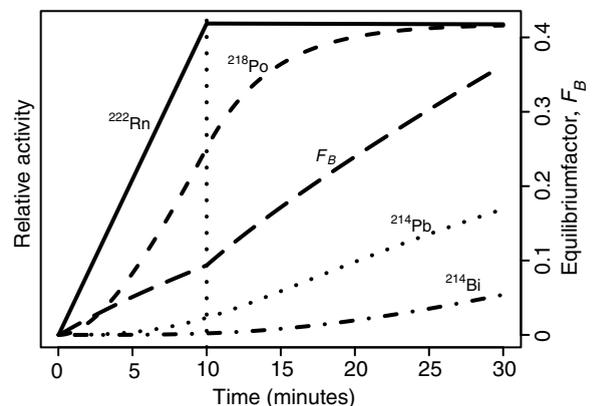


Fig. 6. Relative activities of radon-222 and decay products in a hypothetical sealed room, neglecting effects of ventilation, attachment to aerosols, and deposition onto surfaces.

(International Commission on Radiation Protection (ICRP, 1994) of airborne Rn decay products can be estimated for showering and for whole-house conditions

$$\text{Equilibrium equivalent concentration (Bq m}^{-3}\text{)} = C_T \times F_{\text{average}} \times {}^{222}\text{Rn}_{\text{water}} \text{ activity (Bq m}^{-3}\text{)} \quad (2)$$

If exposure time is known, then the alpha energy contribution from each source may be estimated by multiplying equilibrium equivalent concentration by exposure time and the factor $0.00556 \mu\text{J m}^{-3}$ per Bq m^{-3} of equilibrium equivalent concentration (ICRP, 1994). Although shower exposure varies in a linear fashion with Rn in water activity (Fitzgerald et al., 1997), C_T and F vary in a non-linear fashion with shower length and time prior to opening the bathroom door. If the door is opened quickly after the water is turned off, Rn and its progeny will escape from the room while F_B remains low, even though Rn activity will be near its peak. On the other hand, a long period of exposure before the door is opened (~ 1 h) would allow F_B to approach average steady-state conditions. From data published elsewhere (Fitzgerald et al., 1997), an estimated average F_B between 0.05 and 0.15 (here estimated at 0.1) is consistent with a ~ 10 -min shower followed by ~ 10 additional min before opening the door. It must be emphasized that variability in shower length adds additional uncertainty to exposure estimation, not considered here, in addition to uncertainty related to ventilation and the reactivity of Rn progeny ions. Finally, this simple model only provides estimates of airborne Rn progeny exposure. Parameters necessary for estimating actual dose to the lungs, including aerosol particle size and humidity, are beyond the scope of these field measurements, so the data do not provide a basis for dosimetric calculations (e.g. Fitzgerald et al., 1997).

Nevertheless, it is possible to compare the estimates to the expected whole-house Rn progeny exposure derived from water using $C_{T,H}$ of 0.55×10^{-4} (NRC, 1999), F_H of 0.4 (EPA, 2003), and 70% of time spent at home (EPA, 2003) and treating average inferred whole-house conditions as a background that underlies the entire period of occupancy, including showering. Using an average of 0.98 showers per person per day (Wilkes et al., 2005), estimated additional exposure is presented in Table 3 for a shower length of 7.2 min, which is the median of two

geometric mean shower lengths for United States residents (6.8 and 11.3 min; Wilkes et al., 2005) and an Australian median estimate (7.2 min; Burmaster, 1998), and 10 additional minutes before opening the door. Thus, the 40-min. $C_{T,B}$ values (Table 2) were converted into 17.2-min. transfer coefficients. This was accomplished by (1) assuming that the occupant would be exposed to the maximum level of Rn at the time the water was turned off; (2) modeling the increase of Rn as linear using the median rate of increase during the 20-min shower; and (3) modeling Rn escape from the room as exponential (Hess et al., 1982). The escape constant was determined from the median of the median Rn escape obtained from each room (36% Rn reduction per 5 min). To convert to the 17.2-min scenario using this non-linear approach, the 40-min transfer coefficients were divided by 2.26. Applying this transfer coefficient to the occupancy and equilibrium assumptions described above suggests that the above scenario could result in a geometric mean of approximately 5.6% increased exposure to Rn progeny derived from water relative to the whole-house transfer coefficient (Table 3). This is generally consistent with the 8–9% estimated increase from showering in one experimental house (Datye et al., 1997). Per unit time (Eq. (2)), showering provides 3.3 times as much alpha exposure, on average, from water-derived Rn decay products than overall average conditions in the house. The large two-sigma interval produced by these calculations (Table 3) indicates the significant individual characteristics of homes that influence Rn exposure, described above, as well as the long tail in the upper quantiles of the lognormal distribution.

5. Conclusions

During and after showers, significant airborne Rn elevation is seen in bathrooms receiving water with Rn activities above the alternative maximum contaminant level of 148 Bq L^{-1} (4000 pCi L^{-1}) proposed by the EPA. However, at the one house using a treatment system for Rn in water, there was no significant increase of Rn in bathroom air. Although most water supplies containing Rn activity over 148 Bq L^{-1} exhibited a significant increase in airborne Rn associated with shower use, the level of airborne Rn in

Table 3

Exposure estimation from Rn in water from showering and house-scale exposure. Point estimates are geometric means

Scale of exposure	C_T	$\pm 2\sigma$	F	Annual exposure time (h)	Annual alpha energy contribution per Bq L^{-1} Rn in water ($\mu\text{J h m}^{-3}$)	$\pm 2\sigma$	Increase due to showering (%)	$\pm 2\sigma$
Whole-house	5.50E-05	(5.02E-08–6.03E-02) ^a	0.4 ^b	6132 ^c	0.750	(7.99E-04–712)		
7.2 min shower + 10 min off	7.28E-04	(1.35E-04–3.94E-03)	0.1	103 ^d	0.042	(7.71E-03–0.226)	5.6	(0.03–965) ^e

^a National Research Council, 1999.

^b Environmental Protection Agency, 2003.

^c 70% of time spent at home (EPA, 2003).

^d Based on 7.2 min in shower and 10 additional minutes before opening door.

^e Assuming that bathrooms and their corresponding houses are at the same quantiles on the $C_{T,B}$ and $C_{T,H}$ distributions.

bathrooms is not strongly related to Rn in water activity due to variability in ventilation characteristics between houses, room size and water flow rates. Relative to the contribution from water to whole-house Rn, estimated by the $\sim 10^{-4}$ transfer coefficient, it is suggested that showering contributes a few percent of additional Rn progeny exposure, and is dependent on uncertain estimates of exposure time (shower length and after-shower length in a closed bathroom) and F_B . Relative to Rn-from-water exposure generated at longer temporal and larger spatial scales, short-term shower exposure is highly individualized not only because of variations between houses, but also due to variations in shower length and the time of day that showers are taken, that is the length of time that water used for showering has been stored in pipes, the water heater, or the well bore.

Only one of several common household exposures in which occupants are close to water uses is documented. For example, dishwashing and laundry may also generate additional incremental increases in Rn progeny exposure over that suggested by $C_{T,H}$ that will similarly depend on exposure time, location, ventilation, room volume and water volume. However, it should be emphasized that in spite of these upward-revised estimates of Rn progeny exposure due to water, the findings do not substantively affect existing interpretations that the largest source of Rn progeny exposure for the overall population is from Rn drawn directly into houses from rock and soil rather than Rn from water supplies (NRC, 1999; EPA, 2003). The findings are of greatest relevance to (1) residents for whom most Rn exposure comes from water (i.e. high Rn-from-water contribution, low Rn-from-soil contribution; Folger et al., 1994), and (2) residents exposed to somewhat elevated indoor Rn, such as levels close to the EPA action level of 148 Bq m^{-3} (4 pCi L^{-1}). In the second case, considering short-term Rn exposure from water in addition to whole-house Rn-from-water exposure may cause such residents to exceed a nominal exposure level such as that suggested by the EPA action level.

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References

- Abu-Jarad, F., 1982. Variation in long-term radon and daughters concentration with position inside a room. *Radiat. Protect. Dosim.* 3, 227–231.
- Bernhardt, G.P., Hess, C.T., 1996. Acute exposure from ^{222}Rn and aerosols in drinking water. *Environ. Int.* 22 (1), S753–S759.
- Brutsaert, W.F., Norton, S.A., Hess, C.T., Williams, J.S., 1981. Geologic and hydrologic factors controlling radon-222 in ground water in Maine. *Ground Water* 19, 407–417.
- Burmester, D.E., 1998. A lognormal distribution for time spent showering. *Risk Anal.* 18, 33–35.
- Campbell, T.R., 2006. Radon-222 and other naturally-occurring radionuclides in private drinking water wells and radon in indoor air in Buncombe, Henderson, and Transylvania Counties, North Carolina, 2005. North Carolina Department of Environment and Natural Resources, Ground Water Circular 20.
- Clesceri, L.S., Greenberg, A.E., Eaton, A.D., 1998. Standard methods for the examination of water and wastewater, 20th ed. American Public Health Association/American Water Works Association/Water Environment Federation, Washington.
- Cook, P.G., Love, A.J., Dighton, J.C., 1999. Inferring ground water flow in fractured rock from dissolved radon. *Ground Water* 37, 606–610.
- Dalgaard, P., 2002. *Introductory Statistics with R*. Springer, New York.
- Datye, V.K., Hopke, P.K., Fitzgerald, B., Raunemaa, T.M., 1997. Dynamic model for assessing ^{222}Rn and progeny exposure from showering with radon-bearing water. *Environ. Sci. Technol.* 31, 1589–1596.
- Dillon, M.E., Carter, G.L., Arora, R., Kahn, B., 1991. Radon concentrations in ground water of the Georgia Piedmont. *Health Phys.* 60, 229–236.
- Environmental Protection Agency, 2003. EPA Assessment of risks from radon in homes. Air and Radiation Report EPA 402-R-03-003.
- Field, R.W., Kross, B.C., 1998. Iowa survey of waterborne ^{222}Rn concentrations in private wells. *Health Phys.* 74, 249–252.
- Fisher, E.L., Field, R.W., Smith, B.J., Lynch, C.F., Steck, D.J., Neuberger, J.S., 1998. Spatial variation of residential radon concentrations: the Iowa radon lung cancer study. *Health Phys.* 75, 506–513.
- Fitzgerald, B., Hopke, P.K., Datye, V., Raunemaa, T., Kuuspallo, K., 1997. Experimental assessment of the short- and long-term effects of ^{222}Rn from domestic shower water on the dose burden incurred in normally occupied homes. *Environ. Sci. Technol.* 31, 1822–1829.
- Focazio, M.J., Tipton, D., Dunkle Shapiro, S., Geiger, L.H., 2006. The chemical quality of self-supplied domestic well water in the United States. *Ground Water Monitor. R.* 26, 92–104.
- Folger, P.F., Nyberg, P., Wanty, R.B., Poeter, E., 1994. Relationships between ^{222}Rn dissolved in ground water supplies and indoor ^{222}Rn concentrations in some Colorado Front Range houses. *Health Phys.* 67, 245–253.
- Gesell, T.F., Prichard, H.M., 1980. The contribution of radon in tap water to indoor radon concentrations. In: Gesell, T.F., Lowder, W.M. (Eds.), *Natural Radiation Environment III: Proc. Symp. Houston, Texas, April 23–28, 1978*, US Department of Energy, Symposium Series, vol. 51, pp. 1347–1363.
- Harris, S.A., Billmeyer, E.R., Robinson, M.A., 2006. Evaluation of repeated measurements of radon-222 concentrations in well water sampled from bedrock aquifers of the Piedmont near Richmond, Virginia, USA: effects of lithology and well characteristics. *Environ. Res.* 101, 323–333.
- Hess, C.T., Weiffenbach, C.V., Norton, S.A., 1982. Variations of airborne and waterborne Rn-222 in houses in Maine. *Environ. Int.* 8, 59–66.
- Hess, C.T., Vietti, M.A., Lachapelle, E.B., Guillemette, J.F., 1990. Radon transferred from drinking water into house air. In: Cothorn, C.R., Rebers, P.A. (Eds.), *Radon, Radium, and Uranium in Drinking Water*. Lewis Publishers, Chelsea, Michigan, pp. 51–67 (Chapter 5).
- International Commission on Radiological Protection (ICRP), 1994. Protection against radon-222 at home and at work. ICRP Publication 65 (Annals of the ICRP 23 (2)).
- King, P.T., Michel, J., Moore, W.S., 1982. Ground water geochemistry of ^{228}Ra , ^{226}Ra and ^{222}Rn . *Geochem. Cosmochim. Acta* 46, 1173–1182.
- Krewski, D., Lubin, J.H., Zielinski, J.M., Alavanja, M., Catalan, V.S., Field, R.W., Klotz, J.B., Letourneau, E.G., Lynch, C.F., Lyon, J.L., Sandler, D.P., Schoenberg, J.B., Steck, D.J., Stolwijk, J.A., Weinberg, C., Wilcox, H.B., 2006. A combined analysis of North American case-control studies of residential radon and lung cancer. *J. Toxicol. Environ. Health A* 69, 533–597.
- Lawrence, E.P., Poeter, E., Wanty, R.B., 1991. Geohydrologic, geochemical, and geologic controls on the occurrence of radon in ground water near Conifer, Colorado, USA. *J. Hydrol.* 127, 367–386.
- Loomis, D.P., 1987. Radon-222 concentration and aquifer lithology in North Carolina. *Ground Water Monitor. R.* 7, 33–39.
- Lowry, J.D., Hoxie, D.C., Moreau, E., 1987. Extreme levels of ^{222}Rn and U in a private water supply. In: Graves, B. (Ed.), *Radon, Radium, and Other Radioactivity in Ground Water: Hydrogeologic Impact and Application to Indoor Airborne Contamination*. In: Proc. NWWA Conf., April 7–9, 1987. Somerset, New Jersey, Lewis Publishers, Chelsea, Michigan, pp. 363–375.

- Malanca, A., Gaidolfi, L., Pessina, V., Dallara, G., 1995. What is the “true” radon concentration inside a room? *J. Environ. Sci. Health A30*, 1935–1943.
- McGregor, R.G., Gourgon, L.A., 1980. Radon and radon daughters in homes utilizing deep well water supplies, Halifax County, Nova Scotia. *J. Environ. Sci. Health A15*, 25–35.
- Nazaroff, W.W., Doyle, S.M., Nero, A.V., Sextro, R.G., 1987. Potable water as a source of airborne ^{222}Rn in US dwellings: a review and assessment. *Health Phys.* 32, 281–295.
- Nikolopoulos, D., Vogiannis, E., 2007. Modelling radon progeny concentration variations in thermal spas. *Sci. Total Environ.* 373, 82–93.
- North Carolina Geological Survey, 1985. Geologic map of North Carolina [scale 1:500,000], digital geologic map obtained from <<http://www.geology.enr.state.nc.us>>.
- National Research Council, 1991. Comparative Dosimetry of Radon in Mines and Homes. National Academies Press, Washington.
- National Research Council, 1999. Risk Assessment of Radon in Drinking Water. National Academies Press, Washington.
- Partridge, J.E., Horton, T.R., Sensintaffar, E.L., 1979. A study of radon-222 released from water during typical household activities. Environmental Protection Agency, Technical Note ORP/EERF-79-1.
- Put, L.W., de Meijer, R.J., 1988. Variation of time-averaged indoor and outdoor radon concentrations with time, location and sampling height. *Radiat. Protect. Dosim.* 24, 317–320.
- R Development Core Team, 2007. R: a language and environment for statistical computing [software version 2.5.1]: R Foundation for Statistical Computing, Vienna, <<http://www.R-project.org>>.
- Samet, J.M., 2006. Residential radon and lung cancer: end of the story? *J. Toxicol. Environ. Health A* 69, 527–531.
- Senior, L.A., 1998. Radon-222 in the ground water of Chester County, Pennsylvania. US Geol. Surv. Water-Resour. Invest. Rep. 98-4169.
- Wanty, R.B., Johnson, S.L., Briggs, P.H., 1991. Radon-222 and its parent radionuclides in groundwater from two study areas in New Jersey and Maryland, USA. *Appl. Geochem.* 6, 305–318.
- Wilkes, C.R., Mason, A.D., Hern, S.C., 2005. Probability distributions for showering and bathing water-use behavior for various US subpopulations. *Risk Anal.* 25, 317–337.
- Zhang, Z., Zhang, L., Li, C., Xiao, D., 1993. Investigation of distribution of indoor radon concentration. *Nucl. Tracks Radiat. Meas.* 22, 515–518.