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Contributions of Building Materials to Indoor Radon Levels in Florida Buildings

Final Report

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E. Timothy Oppelt, Director
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ABSTRACT

The Florida Standard for Radon-Resistant Residential Building Construction originally contained a provision to limit the concentration of radium in concrete. The provision was designed to prevent concrete from causing elevated indoor radon concentrations. It was removed from the October 1994 version of the standard, however, because concrete from commercial sources had not been shown to be a major radon contributor in Florida. This report documents follow-up work aimed at identifying one or more Florida buildings whose source of indoor radon is suspected to come from building materials, and at recommending related changes to the building materials radium standard.

A mathematical model is presented to estimate the contributions of building materials to indoor radon concentrations. The model computes radon flux from concrete surfaces using typical Florida concrete properties, and multiplies the flux by corresponding concrete areas to determine their radon contribution to a building. A simplified expression is given that accounts for building ventilation in estimating the radon source to a building from building materials. Published radon data from houses and large buildings are used to calculate indoor radon sources from building materials.

Past and present radium and radon emanation measurements for Florida concretes and their constituents are presented and analyzed to characterize typical Florida concrete properties. Radium distributions in residential floor slabs had a geometric mean of 1.3 pCi g^{-1} and a geometric standard deviation (GSD) of 1.62. Radon emanation coefficients for the slabs averaged 0.10 ± 0.04. Radium measurements in concretes with potentially elevated radon sources had a similar geometric mean of 1.4 pCi g^{-1}, but a much greater GSD of 3.0, owing to occasional elevated-radium samples. Radon emanation coefficients for these samples were also slightly higher and more variable, averaging 0.14 ± 0.07. Radium and radon emanation were also measured in concrete aggregate materials. They showed similar distributions, with occasionally elevated radium concentrations consistent with the concrete measurements.

A concrete and block building in Lake City was found to contain both elevated concrete radium levels and elevated indoor radon. Gamma ray surveys suggested elevated radium levels, and subsequent concrete analyses showed 33 pCi g^{-1} radium in one slab. Indoor radon concentrations averaged 5.0 ± 0.8 pCi L^{-1}, and radon source calculations suggested a ventilation rate of 0.43 h^{-1} during the elevated radon period. The radon source calculations suggested that approximately 93% of the radon came from the ceiling slab, while only 3% came from the floor slab and block walls. The remaining 4% of the radon was estimated to have diffused through the floor slab from foundation soils. The calculated radon source strengths were also consistent with the gamma ray trend identified from published data.

A revised building material radium standard was developed that accounts for the areas and radium concentrations of concretes exposed to building interiors. The standard would limit the indoor radon increment from building materials to no more than 2 pCi L^{-1}. It would limit concrete radium concentrations to 7 to 9 pCi g^{-1} if only a single slab or walls contain elevated radium. However it could limit radium to approximately 3 pCi g^{-1} if floor, ceiling, and walls all utilize concrete with elevated radium.

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1. INTRODUCTION

1.1 BACKGROUND

Radon (^{222}Rn) gas enters buildings primarily from radium (^{226}Ra) in foundation soils. However, significant radon contributions can also come from radium in building materials and from radon dissolved in water if the source strengths in these media are sufficiently elevated. If the total radon entry rate is elevated and the building is not well ventilated, radon can accumulate to levels that can significantly increase the occupants' risks of lung cancer with chronic exposure. The degree of health risk is proportional to the long-term average level of radon exposure. The U.S. Environmental Protection Agency (EPA) attributes 7,000 to 30,000 lung cancer fatalities annually to radon, and recommends remedial action if indoor radon levels average 4 picocuries per liter (pCi L^{-1}) or higher (EPA92a; EPA92b).

The Florida Department of Community Affairs (DCA), under the Florida Radon Research Program (FRRP), has developed radon-protective building standards. These standards are incorporated in proposed rule 9B-52, the Florida Standard for Radon-Resistant Residential Building Construction (DCA94), which is primarily aimed at controlling radon by blocking its entry from foundation soils. The standards require passive radon barriers and active sub-slab ventilation in regions with elevated soil radon potentials, as identified by a statewide radon protection map.

A criterion was developed under the FRRP to limit radon sources in building materials (Rog96). The criterion was included in early drafts of the Florida Standard for Radon-Resistant Residential Building Construction (RAE-9226/4-2, May 1994, Sec. 403.4.1), and required that:

> No material used in concrete for the construction of habitable structures shall have a radium concentration that exceeds 10 pCi g^{-1}, as measured in accordance with approved procedures.

This criterion was removed from the October 1994 version of the standard, when proposed for public hearing, after comments from the Florida Concrete and Products Association indicated that the criterion was unnecessary because: (a) concrete from commercial sources had not been shown to be a major radon contributor in Florida; (b) testing and related cost impacts were not defined; and (c) it appeared that concrete was singled out without considering drywall, lumber, carpets, insulation, and other materials. Related comments from FRRP scientists suggested that inclusion of the radium criterion would encourage suppliers to use higher-radium materials because it was allowed, and that the proposed criterion was three to five times higher than would be expected for a uniform material exposed to the indoor environment.

1.2 OBJECTIVE AND SCOPE

This report presents the findings of a task initiated by DCA under the FRRP to address the first objection to the concrete radium criterion, that concrete from commercial sources had not been shown to be a major radon contributor in Florida. The objective of the task was to identify buildings within Florida whose source of indoor radon was suspected to be building materials. The cause of the problem was also to be examined, and recommendations were solicited for related changes to the standard.

Method development and cost analysis for concrete testing were not included in this study, since funds were limited and the need for concrete testing was still in question. Further study of concrete as a radon source was justified by FRRP scientists, who recognized the potential of concrete to significantly contribute to indoor radon, while the potentials for drywall, lumber, carpets, insulation, and other materials to contribute to indoor radon were judged to be ten to hundreds of times lower, based on literature surveys (Smi80, Ing83, Kah83, Naz88, Obr95). Therefore, this study focused on concrete and concrete products (block).

The following chapter (2) presents a mathematical model to characterize the effects of building material radon sources on indoor radon, and correlates published radon

measurements with gamma ray activity. Chapter 3 summarizes the results of FRRP radium and radon emanation measurements in various concrete and aggregate samples. Chapter 4 presents measurements and analyses linking elevated indoor radon measurements in a concrete building with elevated radium concentrations. Chapter 5 presents a technical basis and draft text for a revised building material radium standard.

2. THEORETICAL EFFECT OF CONCRETE RADON SOURCES

Radon generated by concrete or other building materials cannot be distinguished from soil-generated radon once it has entered a structure and mixed with indoor air. Radon from concrete therefore must be measured directly as a flux exiting a slab or wall surface to characterize it separately from other sources. Although radon fluxes from building materials have been measured in several studies (Smi80; Nie95b; Obr95), the procedures are often difficult and expensive, making alternative approaches such as modeling preferable whenever possible. This chapter describes a simple modeling approach that is used to estimate indoor radon contributions from concrete and other building material sources (Smi80; Nie94). This chapter also compares published indoor radon data with gamma ray measurements to suggest a simple empirical approach for estimating radon source strengths.

2.1 MATHEMATICAL MODEL

Indoor radon concentrations reflect a balance between the rate of radon entry into a structure and the rate of radon loss by decay and dilution by ventilating air. The rate of radon entry is the sum of radon coming from foundation soils, building materials, and in unusual cases, water supplies, natural gas combustion, and any other potential sources. Radon loss rates are invariably dominated by the building ventilation rate, which is commonly expressed in air changes per hour. The simple equation expressing the indoor radon concentration under these conditions is:

$$C_{net} = C_{in} - C_{out} = \frac{\sum_{i}^{n} J_i \cdot A_i}{V(\frac{\lambda}{3{,}600} + \lambda_{Rn})} \qquad (1)$$

where C_{net} = net indoor radon from non-airborne sources (pCi L^{-1})

C_{in} = measured indoor radon concentration (pCi L^{-1})

C_{out} = outdoor radon concentration in ventilating air (pCi L^{-1})

J_i = radon flux from surface i (pCi m^{-2} s^{-1})

A_i = area of radon-emitting surface i (m^2)

V = interior volume of the structure (L)

λ = rate of ventilation by outdoor air (h^{-1})

3,600 = unit conversion ($s\ h^{-1}$)

λ_{Rn} = rate of radon decay ($2.1 \times 10^{-6}\ s^{-1}$).

The expression for indoor radon concentration can be simplified even further by neglecting the C_{out} and λ_{Rn} terms. The outdoor radon concentration, C_{out}, seldom approaches the 4-pCi L^{-1} level at which C_{in} becomes a concern, so C_{out} can be generally ignored. Similarly, the value of λ_{Rn} is only $2.1 \times 10^{-6}\ s^{-1}$, which is less than 8% of the common lower limit of $\lambda/3{,}600 = 2.8 \times 10^{-6}\ s^{-1}$ for ventilation rates in most occupied buildings. With these simplifications, equation (1) can be rearranged by grouping λ with C_{in} (hereafter called C) to isolate the most variable building properties from the more constant ones, giving the following expression:

$$C\lambda = \frac{3{,}600}{V}\sum_{i}^{n} J_i \cdot A_i \qquad (2)$$

The radon flux for a concrete surface can be calculated from the radium concentration, density, emanation coefficient, diffusion coefficient, and thickness of the concrete as:

$$J = 10^4 R \rho E \sqrt{\lambda_{Rn} D}\ \tanh\left(\frac{x}{2}\sqrt{\frac{\lambda_{Rn}}{D}}\right) \qquad (3)$$

where 10^4 = unit conversion ($cm^2\ m^{-2}$)

R = concrete radium concentration ($pCi\ g^{-1}$)

ρ = concrete bulk dry density ($g\ cm^{-3}$)

E = concrete radon emanation coefficient (dimensionless fraction)

D = radon diffusion coefficient for the concrete ($cm^2\ s^{-1}$)

x = concrete thickness (cm).

2.2 COMPARISON WITH GAMMA RAY INTENSITY

Using the simplified relationship in equation (2), published (Kah83) radon concentrations calculated for building materials in houses and large buildings are compared with corresponding calculations of gamma ray intensity. The $C\lambda$ grouping from equation (2) is used to obtain a lumped parameter that is less subject to time and variations caused by changes in building ventilation rate. The radon source strengths ($C\lambda$) are plotted versus gamma ray activity in Figure 2-1 to obtain the following relationship by least-squares linear regression:

$$C\lambda = 0.0127\gamma - 0.081 \tag{4}$$

where γ = gamma ray activity ($\mu R\ h^{-1}$).

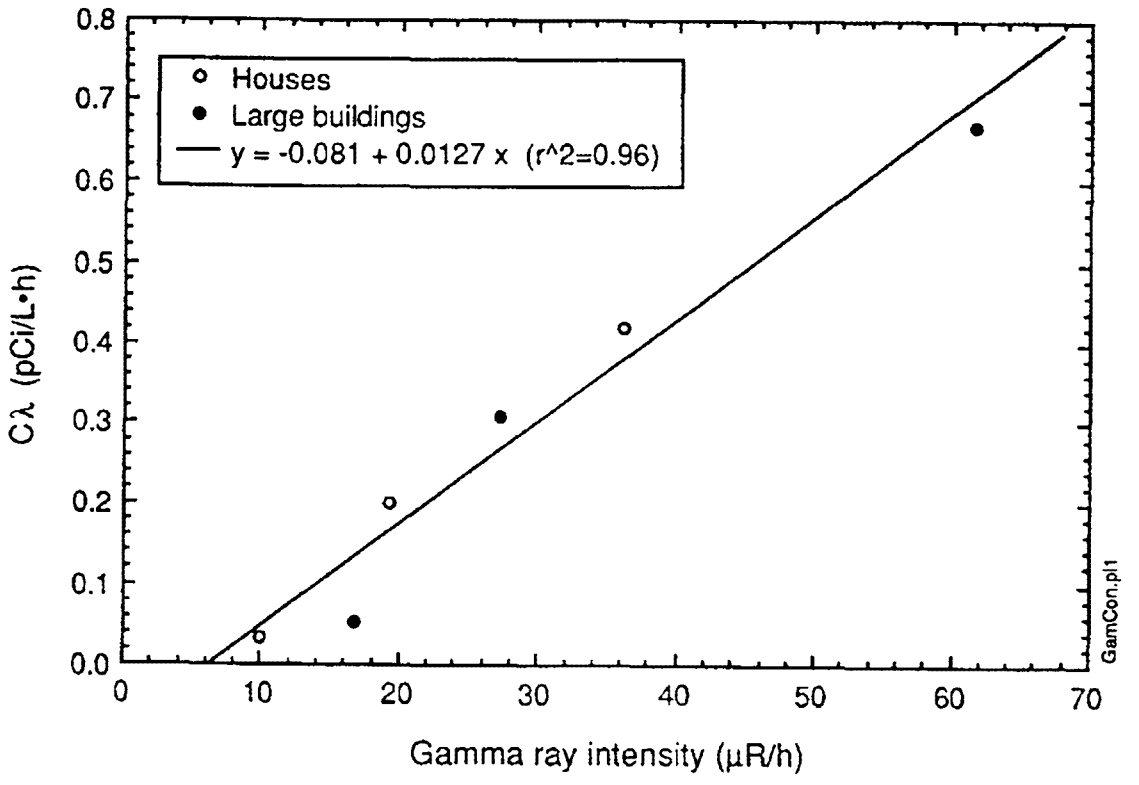

Figure 2-1. Regression of radon sources ($C\lambda$) on gamma activity using data from Kah83.

The empirical correlation of radon source strength with indoor gamma ray intensity in equation (4) could potentially offer a simple, inexpensive test for radon sources in building materials. However, actual gamma ray measurements are subject to several potential biases, including natural background gamma activity, ^{232}Th and ^{40}K gamma activity from the building materials, and source-measurement geometry biases. The effects of background gamma activity should be avoidable by simply subtracting an appropriate background value from the indoor measurements.

Contributions from ^{232}Th activity should generally be small and predictable, since thorium concentrations in most of Florida's natural earthen materials are on the order of 1 pCi g^{-1} or less. Although exceptions in certain mineralized areas could lead to elevated gamma ray measurements, the exceptions would be conservative. This means that the possible ^{230}Th anomalies could lead to unnecessary testing of building materials in a few cases, but that they would not lead to unknowingly incorporating materials with elevated radon sources into new buildings. Contributions from ^{40}K would be similar in nature to those from ^{230}Th, except that they would be much smaller and less frequent.

Possible biases from different source-measurement geometries could generally be made conservative by utilizing maximum readings where the gamma distribution is non-uniform. Although the gamma distribution is relatively uniform if elevated radium levels are present in large concrete floor or ceiling slabs, elevated radium in smaller structures causes a more localized gamma anomaly. Since the smaller structures would cause proportionately less indoor radon, the maximum gamma measurements close to a small structure would overestimate total building radon if attributed to a slab geometry. Therefore, indoor gamma ray measurements could conservatively screen building materials for elevated radon sources. Sampling and laboratory analysis could then be used only where a confirmatory measurement is required.

3. RADIUM AND RADON EMANATION MEASUREMENTS

A review of radium and radon emanation measurements in Florida concretes gives insight into their typical radon source properties. Radium concentrations in concrete floor slabs from Florida houses were directly measured in two previous FRRP studies, one dealing with new houses (Rog94) and the other with older houses (Rog95). Additional concrete analyses were performed in connection with anomaly investigations for the statewide mapping study (Nie95a), and in connection with this study. Together, the concrete analyses give an approximate characterization of the range of radium concentrations and radon emanation coefficients in Florida residential concretes. Additional data on rock aggregate materials are also summarized here from separate FRRP measurements as a possible explanation of the radium distributions observed in Florida concretes.

3.1 MEASUREMENTS IN CONCRETES

In the two previous studies that focused on concrete floor slabs in Florida houses, samples were obtained from cores drilled from the floor slabs of residential structures (Rog94; Rog95). The structures were chosen to represent typical single-family dwellings without regard to indoor radon levels; in fact, indoor radon data were not available for these houses. The radium and emanation measurement procedures and supporting quality assurance (QA) data were reported previously (Rog94). The results of the analyses are presented in Table 3-1.

The data from the first study (first seven rows in Table 3-1) show a geometric mean radium concentration of 1.4 pCi g^{-1} and a geometric standard deviation (GSD) of 1.38, while the data from the second study (remaining rows of Table 3-1) show a geometric mean radium concentration of 1.3 pCi g^{-1} and a GSD of 1.76. Although the variations are larger among the older homes, the means are not significantly different, and both sets are represented here by a single distribution for the 19 slabs with a geometric mean of 1.3 pCi g^{-1} and a GSD of 1.62. Radon emanation averaged 0.069 ± 0.008 in the first study and 0.116 ± 0.042 in the second study, with an overall average of 0.101 ± 0.041 for all 18 slabs in Table 3-1.

Table 3-1. Radium and radon emanation measurements in Florida house concretes.

Sample Identification	Concrete Age (years)	Slab Location	Radium[a] Concentration (pCi g^{-1})	Radon[b] Emanation (fraction)
C002F	1	Jacksonville	1.6 ± 0.1	(not measured)
C003F	1	Jacksonville	1.3 ± 0.1	0.063 ± 0.012
C004C	1	Florida[c]	2.4 ± 0.1	0.057 ± 0.002
C005C	1	Florida[c]	1.7 ± 0.1	0.072 ± 0.003
TC1-1	1	Bartow	1.0 ± 0.1	0.075 ± 0.007
TC1-C	1	Bartow	1.1 ± 0.1	0.078 ± 0.007
TC2-4	1	Bartow	1.0 ± 0.1	0.070 ± 0.007
A-1	12	Boca Raton	1.7	0.085
B-1	1	Boca Raton	2.6	0.13
C-1	25	Pompano Beach	0.9	0.13
D-1	18	Miami	2.1	0.03
E-1	20	Boca Raton	0.9	0.19
F-1	14	Boca Raton	1.5	0.11
G-1	45	Delray Beach	1.0	0.06
H-1	20	Miami	0.6	0.13
I-1	15	Boca Raton	1.8	0.13
J-1	40	Delray Beach	0.4	0.15
K-1	21	Boca Raton	1.3	0.14
L-1	36	Miami	2.2	0.11
First study mean ± s.d.	1 ± 0		1.4 (1.38)[d]	0.069 ± 0.008
Second study mean ± s.d.	22 ± 13		1.3 (1.76)[d]	0.116 ± 0.042
Overall mean ± s.d.	14 ± 14		1.3 (1.62)[d]	0.101 ± 0.041

[a]Dry mass basis mean ± standard deviation (based on Poisson counting statistics).

[b]Mean ± standard deviation (based on Poisson counting statistics).

[c]Florida samples from unspecified locations.

[d]Geometric mean and geometric standard deviation in parentheses.

The radium concentrations in Table 3-1 are 40% to 80% higher than typical U.S. or worldwide concrete radium levels, while the radon emanation coefficients are slightly lower than previously reported values (Rog94). Thus, further insight was sought on radium and radon emanation distributions in Florida concretes from analyses of dry-mix concrete materials sampled from four diverse Florida locations. Portions of these samples were separated by sieving to isolate the aggregate, sand, and cement fractions so that each fraction could be analyzed separately. Additionally, bulk analyses were performed on concretes prepared from the dry mixes. The results of these analyses are presented in Table 3-2.

The geometric mean radium concentration for concretes mixed from the four samples was 0.6 pCi g^{-1} (GSD=2.3), nearly identical to the geometric mean of 0.5 pCi g^{-1} (GSD=2.2) among the mass-weighted component means. Interestingly, the geometric mean radium in the cement components was highest (1.2 pCi g^{-1}, GSD=1.4), followed by the highly variable aggregate radium concentrations (0.5 pCi g^{-1}, GSD=4.1) and the uniformly low sand radium concentrations (0.1 pCi g^{-1}, GSD=1.4). Although the average dry-mix radium concentration is only about half the average for the 19 slabs in Table 3-1, both distributions are so variable that this difference is not statistically significant.

The average radon emanation coefficient for concretes mixed from the four samples was 0.19 ± 0.14, nearly identical to the 0.18 ± 0.09 average of the mass-weighted component means that utilized the moist-paste cement emanation coefficients. The average emanation for the moist cement paste (0.31 ± 0.06) was much greater than for the dry cement powder (0.02 ± 0.01); however, the average 18% composition of cement in the concretes minimizes the effect of this moisture dependence in the mass-weighted means. The average emanation of the sand was lower (0.14 ± 0.05), and that for the aggregate was lower yet (0.07 ± 0.07). The average emanation coefficient for the dry-mix concretes is nearly 90% higher than the average for the slabs in Table 3-1, probably as a result of the higher moisture in the dry-mix samples. The potentially strong moisture dependence of emanation in concretes, as suggested by the cement paste data in Table 3-2, suggests a potential bias in using air-dry concrete samples for laboratory emanation measurements. If concretes, particularly slabs contacting soil surfaces, have elevated moisture, their radon emanation may be significantly higher than would be measured from an air-dry laboratory specimen.

Table 3-2. Radium and radon emanation measurements in dry-mix materials.

Sample	Location	Material	Percent of Mix (wt. %)	Radium[a] Concentration (pCi g^{-1})	Radon Emanation (fraction)
M-1	Lakeland	Mixed Concrete	100	1.2 ± 0.2[b]	0.09 ± 0.01[b]
		Cement (moist paste)	16	1.1	0.32
		Cement (dry powder)			0.02
		Sand	45	0.1	0.14
		Aggregate	39	1.8	0.04
		Calculated Weighted Mean	100	0.9 ± 0.2[c]	0.10[d]
M-2	Tampa	Mixed Concrete	100	1.1 ± 0.1[b]	0.13 ± 0.02[b]
		Cement (moist paste)	18	2.0	0.26
		Cement (dry powder)			0.02
		Sand	45	0.1	0.19
		Aggregate	37	1.3	0.03
		Calculated Weighted Mean	100	0.9 ± 0.3[c]	0.13[d]
M-3	Jacksonville	Mixed Concrete	100	0.5 ± 0.2[b]	0.15 ± 0.05[b]
		Cement (moist paste)	15	0.9	0.39
		Cement (dry powder)			0.04
		Sand	77	0.1	0.16
		Aggregate	8	0.1	0.17
		Calculated Weighted Mean	100	0.2 ± 0.3[c]	0.30[d]
M-4	Pensacola	Mixed Concrete	100	0.2 ± 0.1[b]	0.39 ± 0.26[b]
		Cement (moist paste)	22	1.0	0.29
		Cement (dry powder)			0.01
		Sand	43	0.2	0.08
		Aggregate	35	0.2	0.04
		Calculated Weighted Mean	100	0.3 ± 0.2[c]	0.19[d]
M-1 to M-4	Average[e]	Mixed Concrete	100	0.6 (2.3)	0.19 ± 0.14
		Cement (moist paste)	18 ± 3	1.2 (1.4)	0.32 ± 0.06
		Cement (dry powder)			0.02 ± 0.01
		Sand	52 ± 16		
		Aggregate	30 ± 15	0.5 (4.1)	0.07 ± 0.07
		Calculated Weighted Mean	100	0.5 (2.2)	0.18 ± 0.09

[a]Dry mass basis.
[b]Means of three measurements ± std. deviations calculated from Poisson counting statistics.
[c]Standard deviation calculated from Poisson counting statistic uncertainty of components.
[d]Using moist-paste emanation for cement.
[e]Mean ± s.d. for percents and emanation; geometric mean (geometric s.d.) for radium.

Additional concrete analyses were performed in connection with the radon map anomaly investigations (Nie95a) and with this study. The samples for these analyses were obtained from various locations throughout Florida by commercial concrete suppliers, radon mitigators, and Rogers & Associates Engineering Corp. (RAE) personnel. The samples represented both single-family dwellings and multi-story apartment buildings. Although most samples consisted of cores drilled from floor slabs, some were also taken from foundation footings, poured concrete walls, and concrete blocks. The analyses, summarized in Table 3-3, utilized the laboratory and QA procedures described previously (Rog94).

The measurements in Table 3-3 may be less representative of all Florida concretes than those in Table 3-1 because the Table 3-3 samples were sought from buildings with potentially elevated indoor radon (>4 pCi L^{-1}). The radium concentrations in Table 3-3 have only a slightly higher geometric mean (1.4 pCi g^{-1} compared to 1.3 pCi g^{-1}) than those in Table 3-1, but their GSD is significantly higher (3.0 compared to 1.6). The radon emanation coefficients in Table 3-3 average 0.14 ± 0.07, somewhat higher than the 0.10 ± 0.04 average from Table 3-1, but lower than the average in Table 3-2. Although the radon sources (the product of radium concentration and radon emanation coefficient) in Table 3-3 are expectedly higher, they are not high enough to suggest a consistent correlation of building materials with indoor radon. The comparisons are more consistent with the usual trend of indoor radon concentrations that are dominated by foundation soils rather than by building materials.

Despite the usual trend of soil-dominated radon levels, some of the radium concentrations in Table 3-3 are sufficiently high to contribute to or cause elevated indoor radon if sufficient concrete is used in the buildings. Although radon levels dominated by building materials are expected less frequently than levels dominated by soils, the data in Table 3-3 show the possibility for significant radon problems in buildings where concrete components may contain elevated radon sources.

Table 3-3. Radium and radon emanation measurements in concretes from Florida buildings with potentially elevated radon.

Sample Location	Latitude (North)	Longitude (West)	Sample Material	Radium Concentration (pCi g^{-1})	Radon Emanation (fraction)
Florida[a]	[b]	[b]	Concrete floor slab	1.2 ± 0.4	0.12 ± 0.04
Florida[a]	[b]	[b]	Concrete floor slab	0.4 ± 0.3	[c]
Florida[a]	[b]	[b]	Concrete floor slab	1.3 ± 0.4	0.14 ± 0.05
Florida[a]	[b]	[b]	Concrete floor slab	1.6 ± 0.4	0.09 ± 0.02
Florida[a]	[b]	[b]	Concrete floor slab	1.8 ± 0.4	0.11 ± 0.02
Florida[a]	[b]	[b]	Concrete floor slab	2.0 ± 0.4	0.09 ± 0.02
Florida[a]	[b]	[b]	Concrete floor slab	0.6 ± 0.4	0.31 ± 0.20
Ft. Myers	26.492°	81.820°	Concrete floor slab	3.8 ± 0.3	0.04 ± 0.01
Gainesville	[b]	[b]	Concrete floor slab	0.3 ± 0.4	[c]
Gainesville	[b]	[b]	Concrete floor slab	1.0 ± 0.3	0.15 ± 0.05
Gainesville	[b]	[b]	Concrete floor slab	0.6 ± 0.4	0.02 ± 0.01
Gainesville	[b]	[b]	Concrete floor slab	0.6 ± 0.3	0.14 ± 0.08
Lake City	30.179°	82.692°	Concrete floor slab	32.8 ± 1.7	[c]
Lake City	30.179°	82.692°	Concrete floor slab	0.6 ± 0.4	[c]
Naples	26.18°	81.75°	Concrete floor slab	2.7 ± 0.2	0.11 ± 0.01
Tallahassee	[b]	[b]	Concrete floor slab	0.2 ± 0.2	[c]
Ft. Myers	26.491°	81.820°	Concrete foundation	4.0 ± 0.3	0.16 ± 0.01
Naples	26.234°	81.813°	Concrete foundation	4.8 ± 0.3	[c]
Naples	26.234°	81.813°	Concrete foundation	1.2 ± 0.2	[c]
St. Petersburg	27.720°	82.691°	Concrete foundation	1.1 ± 0.9	[c]
Naples	26.232°	81.813°	Concrete wall	4.0 ± 0.3	0.16 ± 0.01
St. Petersburg	27.720°	82.691°	Concrete wall	0.7 ± 0.2	0.30 ± 0.11
Ft. Myers	26.493°	81.836°	Concrete block	0.6 ± 0.3	0.13 ± 0.06
Lakeland	[b]	[b]	Concrete block	1.2 ± 0.4	0.18 ± 0.05
Naples	26.234°	81.813°	Concrete block	5.1 ± 0.2	[c]
Naples	26.18°	81.75°	Concrete block	4.9 ± 0.2	0.08 ± 0.01
St. Petersburg	27.720°	82.691°	Concrete block	2.1 ± 0.2	0.11 ± 0.03
Mean — poured concrete				1.3 (3.1)[d]	0.14 ± 0.08
Mean — concrete block				2.1 (2.5)[d]	0.13 ± 0.04
Mean — all				1.5 (3.0)[d]	0.14 ± 0.07

[a]Florida samples provided without location details.

[b]Latitude and longitude not measured.

[c]Radon emanation not measured.

[d]Geometric mean and geometric standard deviation in parentheses.

3.2 MEASUREMENTS IN CONCRETE AGGREGATES

The occasionally elevated radon sources in concrete may be caused by any of its constituents. However, the radium and emanation measurements in dry-mix materials (Table 3-2) gave little insight on which constituent dominates, since none of the four samples analyzed contained elevated radon sources. A brief survey of concrete aggregate materials was therefore conducted because aggregate is the least-characterized major concrete constituent. Sand, the other major constituent, is widely distributed throughout most of Florida, and its radium distribution is already characterized by aeroradiometric data and other data summarized by the Florida radon map (Nie95a). Radium distributions in sand are log-normal, extending into ranges that could readily contribute to elevated radon concentrations if sands are not judiciously selected in areas containing elevated-radium soils.

The survey of concrete aggregate materials involved collecting and analyzing aggregate samples from sources throughout Florida. The samples were collected opportunistically during various field investigations and map validation studies. They consisted of aggregate materials from active quarries, rock samples from U.S. Geological Survey investigations in Dade and Broward Counties, and road aggregate samples from various sites. The results of the radium and radon emanation measurements on the aggregate samples are presented in Table 3-4.

Table 3-4. Radium and radon emanation measurements in Florida aggregate materials.

Sample Location (County)	Latitude (North)	Longitude (West)	Sample[a] Material	Radium Concentration (pCi g^{-1})	Radon Emanation (fraction)
Broward	[b]	[b]	Potential aggregate	0.7 ± 0.3	0.10 ± 0.04
Broward	[b]	[b]	Potential aggregate	0.7 ± 0.2	0.14 ± 0.05
Broward	[b]	[b]	Potential aggregate	0.5 ± 0.2	0.13 ± 0.06
Broward	[b]	[b]	Potential aggregate	<0.3	[c]
Dade	[b]	[b]	Potential aggregate	<0.3	[c]
Dade	[b]	[b]	Potential aggregate	1.9 ± 0.5	0.55 ± 0.16
Dade	[b]	[b]	Potential aggregate	4.1 ± 0.3	0.66 ± 0.04
Dade	[b]	[b]	Potential aggregate	4.9 ± 0.3	0.49 ± 0.03
Dade	[b]	[b]	Potential aggregate	1.1 ± 0.3	0.26 ± 0.06
Dade	[b]	[b]	Potential aggregate	3.4 ± 0.3	0.29 ± 0.03
Dade	[b]	[b]	Potential aggregate	1.3 ± 0.3	0.09 ± 0.02
Dade	[b]	[b]	Potential aggregate	3.1 ± 0.3	0.51 ± 0.05
Dade	[b]	[b]	Potential aggregate	0.8 ± 0.3	0.16 ± 0.06
Dade	[b]	[b]	Potential aggregate	2.9 ± 0.4	0.86 ± 0.13
Dade	[b]	[b]	Potential aggregate	1.0 ± 0.3	0.33 ± 0.09
Dade	[b]	[b]	Potential aggregate	1.1 ± 0.3	0.05 ± 0.01
Dade	[b]	[b]	Potential aggregate	11.3 ± 0.4	0.50 ± 0.02
Dade	[b]	[b]	Potential aggregate	2.0 ± 0.3	0.38 ± 0.06
Dade	[b]	[b]	Potential aggregate	<0.2	[c]
Dade	[b]	[b]	Potential aggregate	4.1 ± 0.3	0.62 ± 0.05
Dade	[b]	[b]	Potential aggregate	1.2 ± 0.3	0.23 ± 0.06
Dade	25.690°	80.487°	Aggregate	1.7 ± 0.3	0.02 ± 0.01
Lake	28.814°	81.627°	Road aggregate	0.7 ± 0.5	0.25 ± 0.19
Lee	26.491°	81.820°	Aggregate	3.8 ± 0.3	0.05 ± 0.01
Lee	26.498°	81.694°	Aggregate	5.0 ± 0.3	0.04 ± 0.01
Lee	26.497°	81.825°	Aggregate	5.1 ± 0.3	0.04 ± 0.01
Lee	26.491°	81.760°	Aggregate	3.1 ± 0.3	0.05 ± 0.01
Polk	27.886°	82.022°	Road aggregate	56.9 ± 0.5	0.02 ± 0.01
Collier	26.234°	81.813°	Aggregate	1.3 ± 0.3	[c]
Collier	26.234°	81.813°	Aggregate	3.1 ± 0.3	[c]
Nassau	30.569°	81.445°	Road aggregate	0.3 ± 0.3	[c]
Sumter	28.651°	82.008°	Aggregate	1.5 ± 0.3	0.10 ± 0.02
Hillsborough	27.977°	82.402°	Road aggregate	49.7 ± 0.6	0.20 ± 0.02
Hillsborough	27.982°	82.403°	Road aggregate	43.0 ± 0.6	[c]
Geometric mean (GSD) or mean ± s.d.				2.1 (4.0)	0.26 ± 0.23

[a]Potential aggregate is not from a developed quarry; road aggregate includes asphalt.
[b]Latitude and longitude not measured.
[c]Radon emanation not measured.

Radium measured in the five samples from active gravel quarries was distributed most closely, ranging from 1.7 pCi g^{-1} to 5.1 pCi g^{-1}, and having a geometric mean of 2.7 pCi g^{-1} and a GSD of 1.7. These samples may overestimate the typical radium concentration in Florida aggregates, since they would lead to slightly higher concrete radium concentrations than those listed in Table 3-1. They also fall into the upper range of the radium distribution measured for Florida soils (geometric mean = 0.6 pCi g^{-1}; GSD = 3.5) (Nie95a). Radium in the 21 "potential aggregate" rock samples in Table 3-4 ranged from <0.2 pCi g^{-1} to 11.3 pCi g^{-1}, and had a lower geometric mean of 1.4 pCi g^{-1}, but a higher GSD of 2.8. Radium in the five road aggregate samples ranged from 0.7 pCi g^{-1} to 57 pCi g^{-1}, with a geometric mean of 13 pCi g^{-1} and a GSD of 13.2. The overall geometric mean of the 34 radium measurements in Table 3-4 is 2.1 pCi g^{-1}, and its GSD is 4. Although the rock materials described in Table 3-4 may over-estimate typical radium concentrations in Florida concrete aggregate materials, they show a potential for elevated radium concentrations in concretes.

Radon emanation coefficients for the gravels from active quarries averaged 0.05 ± 0.03, significantly less than the 0.35 ± 0.23 for the potential aggregate rocks and the 0.16 ± 0.12 for the road aggregate samples. These differences are probably dominated by differences in ambient moisture levels, since the emanation measurements were conducted at ambient moisture. Surface samples from gravel piles were dry, while the "potential aggregate" rock samples were collected at significant depths below the soil surface. Road aggregates probably had intermediate moisture, since they were in contact with shallow soils, but were mixed with or covered by asphalt materials. In general, the potential and road aggregate samples suggest emanation coefficients comparable to the "wet paste" values in Table 3-2 unless materials are completely dry.

4. ASSOCIATION OF CONCRETE RADIUM WITH INDOOR RADON

Several radium and radon emanation measurements in Chapter 3 are high enough to associate with elevated indoor radon concentrations using the equations in Chapter 2. However, this study also seeks to determine if actual Florida buildings can be found in which elevated indoor radon levels are caused by building materials. This objective requires measurement of elevated indoor radon in buildings that have elevated radium levels in their building materials.

Measurement opportunities were sought in buildings where elevated concrete radium levels had already been measured. However, access to these buildings was limited because the concrete samples were mostly provided by concrete suppliers or construction workers who could not also provide access for indoor sampling of the completed buildings. Therefore, only one building was studied in sufficient detail to show a link between its concrete radium level and the indoor radon concentration. This chapter describes the measurements made in the study building and the calculated contributions of its concrete radium to the indoor radon level.

4.1 EMPIRICAL MEASUREMENTS

The study building was located at 30.179° N latitude and 82.692° W longitude, in the vicinity of Lake City, Florida, which is entirely within a green (low radon potential) area of the Florida radon protection map (Nie95a). The building was a two-story structure with a concrete floor slab, concrete block walls, and a 20-cm concrete slab separating the first and second stories. The building was initially identified by gamma ray surveys, which showed gamma ray intensities exceeding 60 $\mu R\ h^{-1}$ in some locations. Gamma ray surveys in the vicinity of the building showed no elevated soil radium sources, with typical soil gamma intensities in the 2-$\mu R\ h^{-1}$ to 4-$\mu R\ h^{-1}$ range. Radon flux measurements from the bare surfaces of surrounding soils averaged 0.2 ± 0.1 pCi $m^{-2}\ s^{-1}$, also indicating that the site soils should not contribute to elevated indoor radon concentrations.

A detailed gamma ray survey was conducted in the accessible first-floor portion of the building, as shown in Figure 4-1. The survey was designed to identify the relative radioactivity of different structural parts of the building. As illustrated, the gamma activity near the floor was consistently lower than corresponding gamma ray measurements at the ceiling of the first level. The floor measurements averaged 25.9 ± 3.2 μR h^{-1}, while the ceiling measurements averaged 50.7 ± 4.2 μR h^{-1}. Gamma measurements along the block walls were intermediate, as shown in Figure 4-1, while gamma activity at a single accessible location on the floor of the second level was slightly higher than the measurements from the ceiling of the first level. Because of the relative uniformity of the gamma ray distributions over the survey area, it appeared that the concretes were causing the elevated gamma activity.

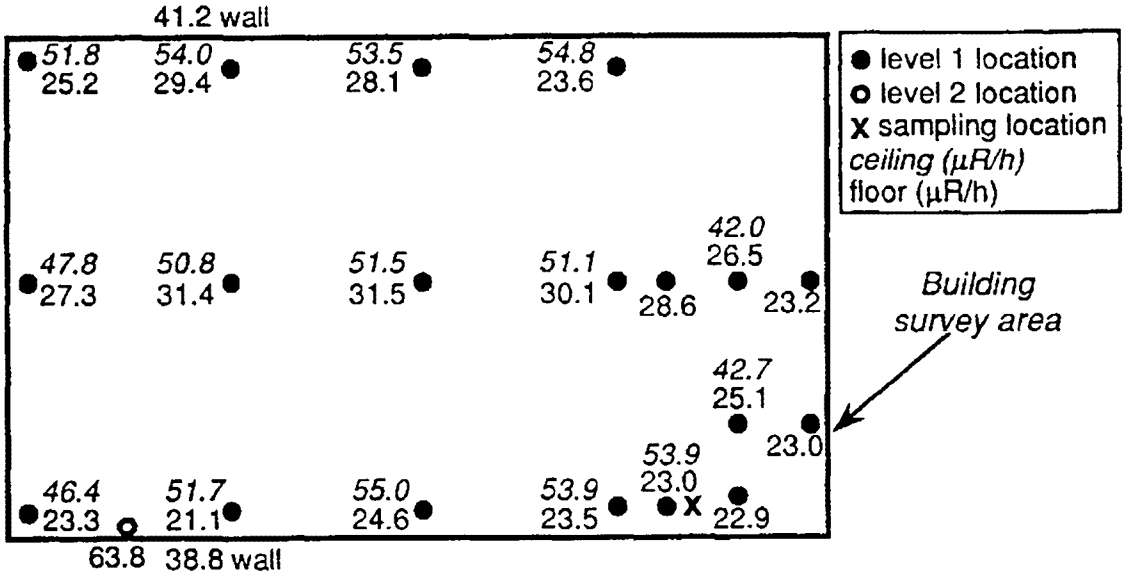

Figure 4-1. Locations of gamma ray measurements in the study building.

Sampling within the building consisted of making triplicate radon flux measurements from the floor slab, taking single concrete samples from the floor slab and the ceiling slab, and making indoor radon measurements in the first level of the building. The radon flux measurements utilized the small charcoal canister method described and used previously for the statewide radon flux sampling (Nie95a). The flux cans were sealed to concrete surfaces with rope caulk. The concrete samples were obtained by drilling several 1.6-cm-diameter, 5-cm-deep holes in the slabs and collecting the drill cuttings on plastic sheets for analysis. The concrete cuttings were analyzed by the same gamma assay procedure used previously for soil samples (Nie95a).

Indoor radon measurements utilized a continuous radon monitor (Model AB-5, Pylon Electronics Inc., Ottawa, Ontario, Canada) that circulated approximately 2 L min^{-1} of room air through its scintillation cell (Pylon, Model 110A) while continually recording alpha activity over 20 min intervals. Radon concentrations were computed from the continuously measured alpha counts using the calibration method and equations of Thomas and Countess (Tho79). The efficiency of the scintillation was determined previously from calibration analyses at the U.S. Department of Energy's Technical Measurement Center radon chamber at Grand Junction, Colorado.

The radon flux measurements from the building floor slab averaged 0.083 ± 0.049 pCi m^{-2} s^{-1}, typical of the range that may be expected from ordinary diffusion of radon through a slab from underlying soils. The concrete radium concentrations were more surprising, however, indicating 0.6 ± 0.4 pCi g^{-1} of radium in the floor slab and 32.8 ± 1.7 pCi g^{-1} in the ceiling slab. Based on these assays, most of the gamma activity at the floor surface was hypothesized to come from the ceiling. The intermediate values along the walls are consistent with this gamma shine interpretation, suggesting that any radium activity in the concrete block walls is too low to significantly affect the gamma measurements.

The indoor radon measurements are presented in Figure 4-2. The concentrations increased at an initial rate of approximately 0.24 pCi L^{-1} h^{-1} during the first 10 h of measurements. The concentrations reached the 3 to 4-pCi L^{-1} range, and then decreased during a period when outdoor gusty winds were observed. The outside door was briefly

opened four times during the measurement period, as shown in Figure 4-2, for entry or exit of personnel. The increased ventilation from door openings may also have contributed to the declines observed during the 10 to 16-h and 22 to 26-h periods.

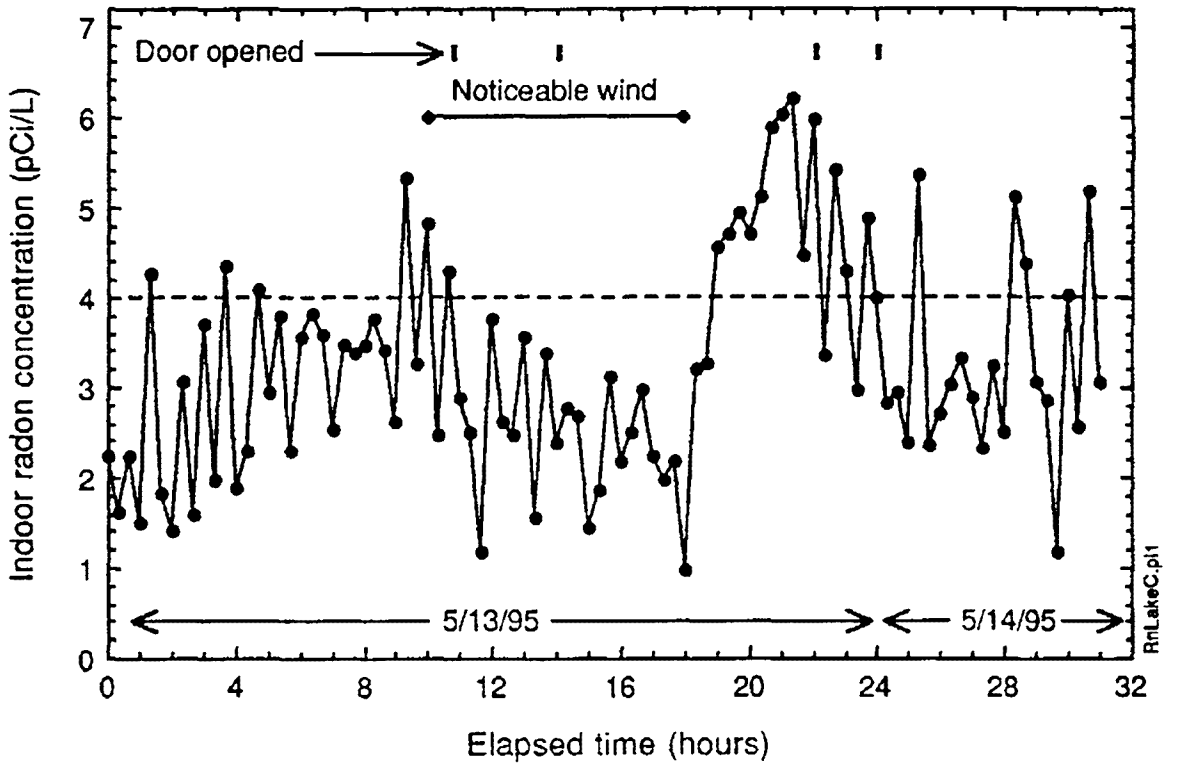

Figure 4-2. Indoor radon measurements in the Lake City study building.

Radon concentrations increased at a higher rate of about 1.2 pCi L^{-1} h^{-1} during the period from 18 to 22 h. They reached the 4 to 6-pCi L^{-1} range and then decreased to levels that were mostly below 4 pCi L^{-1}. The data in Figure 4-2 demonstrate that the building had sufficient radon potential to exceed 4 pCi L^{-1} for sustained periods of several hours when perturbing effects such as winds or mechanical openings were not increasing its natural ventilation rate. For calculation purposes, the indoor radon concentration was estimated from an average of 13 points during the 19 to 23-h period to be 5.0 ± 0.8 pCi L^{-1}.

4.2 CALCULATED EFFECTS

The contributions of various building materials in the study building to indoor radon levels were calculated using the equations presented in Chapter 2. Table 4-1 presents the results of these calculations. Radon fluxes from the ceiling slab were calculated from its 32.8-pCi g^{-1} radium concentration using equation (3), assuming typical density, emanation, and diffusion properties for concretes as measured in the previous studies (Rog94; Rog95). The values used for these parameters are shown in footnotes of Table 4-1. The indoor radon source resulting from this flux was computed from equation (2) using the 25.4-m^2 slab area and 61.9-m^3 volume of the study room. Contributions from the block walls were estimated similarly, assuming a radium concentration equal to that of the floor slab, 0.6 pCi g^{-1}. The wall area used to calculate $C\lambda$ was estimated to be 40.9 m^2. The radon flux and resulting source from radium in the floor slab were calculated from the measured slab radium concentration in the same way as the corresponding values were calculated for the ceiling.

Table 4-1. Calculated contributions of building materials to indoor radon.

Radon Source Material	Radon Flux (pCi m^{-2} s^{-1})	$C\lambda$ Radon Source (pCi L^{-1} h^{-1})	Contribution to Indoor Radon
Ceiling Slab	1.353[a]	1.996	92.9 %
Wall Blocks	0.013[b]	0.031	1.4 %
Floor Slab	0.025[a]	0.037	1.7 %
Foundation Soil	0.058[c]	0.086	4.0 %
Total		2.15	100.0 %

[a]Calculated from measured radium concentration, 10% emanation, 2.1 g cm^{-3} density, and 0.001 cm^2 s^{-1} radon diffusion coefficient.

[b]Same as [a], but assuming 0.6 pCi g^{-1} radium.

[c]Difference between measured flux and floor flux calculated from measured radium.

The flux of radon diffusing through the floor slab from foundation soils was estimated from the difference between the total measured floor flux and the portion that was explained by radium in the slab. The measured floor flux of 0.083 pCi m^{-2} s^{-1} was strongly dominated by underlying soils when compared to the flux of 0.025 pCi m^{-2} s^{-1} calculated to result from radium in the concrete. The soil contribution to the total radon source strength was also estimated using equation (2). The last column in Table 4-1 shows the relative contributions of each of the four components to the total indoor radon concentration.

The indoor radon concentration expected from the calculations in this section is equal to the total value of $C\lambda$ =2.15 pCi L^{-1} h^{-1} from Table 4-1 divided by the ventilation rate of the room. Although the ventilation rate was not directly measured, previous estimates of ventilation in Florida residential structures have usually been in the 0.25-h^{-1} to 0.50-h^{-1} range (Nie94). This range of ventilation rates corresponds to a radon concentration range of 4.3 pCi L^{-1} to 8.6 pCi L^{-1} for the calculated radon source potential. The measured concentration of 5.0 ± 0.8 pCi L^{-1} is within this range, and corresponds to a ventilation rate of λ = 0.43 h^{-1}. This ventilation rate is higher than values estimated for many Florida buildings, suggesting that the measured radon source could potentially cause higher indoor radon levels in a more tightly sealed building. Ventilation rates as low as 0.1 h^{-1} have been measured in Florida (Nie94), and rates as low as 0.04 h^{-1} have been reported for unoccupied buildings when ventilation systems were not operating (Smi80).

The indoor radon source strength was also estimated independently, using the empirical relationship in equation (4). The average gamma ray intensity of 50.7 μR h^{-1} measured near the ceiling gives a radon source estimate of 0.56 pCi L^{-1} h^{-1}, which is within the measurement uncertainty of the 0.52-pCi L^{-1} h^{-1} value estimated in Table 4-1.

The study building satisfies the objective of identifying a Florida building whose source of indoor radon is suspected to be from building materials. Based on the building material contributions demonstrated in Table 4-1, the indoor radon is clearly dominated by radium in the ceiling slab. The long-term average radon concentration in the study building remains unclear because of the short duration of the radon measurements and the lack of

information on its average ventilation rate. However, the short-term radon measurements and ventilation estimates for Florida buildings ($\lambda \approx 0.25 - 0.50\ h^{-1}$) both suggest the potential for long-term radon concentrations exceeding 4 pCi L^{-1}. The consistency of the calculated radon potential with that estimated from the gamma ray correlation in equation (4) suggests a potential for screening buildings for building-material radon sources using gamma ray surveys.

5. BUILDING MATERIALS RADIUM STANDARD

5.1 TECHNICAL BASIS

The present empirical measurements and model analyses show that building materials can and do contribute significantly to indoor radon concentrations in some instances. To protect the public against unknowingly incorporating harmful radon sources into building materials, a standard is proposed for limiting radium concentrations in the building materials. The standard is based on the typical concrete properties used in the analyses in Table 4-1, from which equation (3) gives the following relationship between concrete radium concentration (R in pCi g^{-1}) and radon flux (J in pCi m^{-2} s^{-1}) for a 20 cm concrete wall:

$$J = 0.041R. \tag{5}$$

Substituting equation (5) into equation (2) then gives a relationship that expresses indoor radon concentration as a function of concrete radium concentration, concrete area, ventilation rate, and occupied volume. Assuming a ventilation rate of $\lambda = 0.25\ h^{-1}$, as in previous modeling of Florida residences (Nie95a), the resulting equation can be simplified to give:

$$C = \frac{600}{V}\sum_{i}^{n} R_i A_i \tag{6}$$

where C = indoor radon concentration caused by concrete materials (pCi L^{-1})

R_i = concrete radium concentration in slab i (pCi g^{-1})

A_i = area of interior concrete surface i (m^2)

V = interior occupied volume (L).

Equation (6) can be used to predict indoor radon contributions from concrete building materials under various construction scenarios. For example, a 140-m^2 (1,500-ft^2) residence could have 140 m^2 of floor slab area plus another 140 m^2 of ceiling slab area if it were part of a multi-story building separated by concrete slabs. In addition, concrete or block perimeter walls could comprise an additional 115 m^2 of concrete area exposed to the occupied space.

If all of the concrete contained background radium at the 0.5-pCi g^{-1} level, the concrete would contribute a total of only 0.35 pCi L^{-1} to the indoor radon concentration. However, if the concrete contained elevated radium concentrations, it would cause higher radon levels, as shown by the limiting radium concentrations in Table 5-1. These concentrations are the calculated limits for the total concrete to contribute no more than 2 pCi L^{-1} to the indoor radon levels.

Table 5-1. Limiting concrete radium concentrations for contributing 2 pCi L^{-1} of radon to a 140-m^2 residence using equation (6).

Concrete Structures with a Background Radium Concentration of 0.5 pCi g^{-1}	Concrete Structures with Elevated Radium Concentrations	Limiting Elevated Radium Concentration (pCi g^{-1})
2 Slabs	Walls	8.6
Walls + 1 Slab[a]	1 Slab[a]	7.2
Walls	2 Slabs	3.8
None	2 Slabs + Walls	2.9

[a]Either floor or ceiling slab.

5.2 PROPOSED STANDARD

The standard proposed for limiting radium concentrations in building materials is designed to permit no more than 2 pCi L^{-1} of indoor radon to be caused by the building materials. The 2-pCi L^{-1} limit is purposely defined lower than the 4-pCi L^{-1} standard addressed by the Florida legislature to accommodate radon contributions from other sources, such as soil gas from foundation soils. The proposed standard gives specific guidance for concrete products, since concrete presently appears to be the dominant building material contributing to indoor radon levels. The standard is also formulated to give credit for different occupied volumes, for different surface areas of concrete components, and for different radium concentrations in concrete components. The standard is based on equation

(6), which is explicitly stated in the standard for clarity. Radium concentrations specified by the standard and by equation (6) are intended to be measured by protocols accepted by the FRRP (Wil91). The following standard is therefore proposed for avoiding elevated indoor radon concentrations caused by radium in building materials:

> Building materials used in the construction of habitable structures shall not contain quantities of radium that increase the indoor radon concentration by more than 2 pCi L^{-1}. The contribution of concrete materials toward the 2-pCi L^{-1} limit shall be defined as:
>
> $$C = \frac{600}{V}(R_f A_f + R_c A_c + R_w A_w) \qquad (7)$$
>
> where C = radon concentration from concrete materials (pCi L^{-1})
> V = volume of the habitable space (L)
> R_f = radium concentration in the floor slab(s) (pCi g^{-1})
> A_f = area of the concrete floor slab(s) (m^2)
> R_c = radium concentration in the ceiling slab(s) (pCi g^{-1})
> A_c = area of the concrete ceiling slab(s) (m^2)
> R_w = radium concentration in the concrete walls (pCi g^{-1})
> A_w = area of concrete walls facing the interior volume (m^2).
>
> Radium concentrations used to compute radon contributions shall be measured in accordance with "Standard Measurement Protocols, Florida Radon Research Program" (Wil91), or other procedures accepted by the Department.

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16. ABSTRACT The report documents work to characterize potential radon sources in concretes and recommend related chages to Florida's building materials radium standard. (NOTE: The Florida Standard for Radon-resistant Residential Building Construction originally contained a provision to limit the concentration of radium in concrete. The provision was designed to prevent concrete from causing elevated indoor radon concentrations. It was removed from the October 1994 version of the standard, however, because concrete from commercial sources had not been shown to be a major radon contributor in Florida.) A mathematical model is presented to estimate the contributions of building materials to indoor radon levels. The model computes radon flux from concrete surfaces using typical Florida concrete properties, and multiplies the flux by concrete surface areas to estimate their contribution to indoor radon. The model also accounts for building ventilation by outdoor air. A revised building material radium standard was developed to account for the areas and radium concentrations of concretes exposed to building interiors. The standard would limit the indoor radon increment from building materials to 2 pCi/L. It would limit concrete radium concentrations to 7-9 pCi/g if only a single slab or walls contain elevated radium. However, it could limit radium to about 3 pCi/g if floor, ceiling, and walls are high.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Slabs	Pollution Control	13B	13M
Radon	Soils	Stationary Sources	07B	08G, 08M
Construction Materials		Indoor Air	13C	
Concretes	Residential Buildings			
Radium	Mathematical Models			12A
Ventilation			13A	

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