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**RADON GENERATION AND TRANSPORT  
THROUGH CONCRETE FOUNDATIONS**

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16. ABSTRACT The report gives results of an examination of radon generation and transport through Florida residential concretes for their contribution to indoor radon concentrations. Radium concentrations in the 11 concretes tested were all < 2.5 pCi/g, and radon emanation coefficients were all < 0.08. Measurements on the constituents of four of the concretes revealed that radium concentrations > 1 pCi/g were generally due to elevated radium in the aggregate, but occurred occasionally from radium in the cement. Because the aggregates tested generally had very low emanation coefficients, elevated radium in the aggregate had a lesser impact on indoor radon than elevated radium in the cement component. Diffusion coefficients for Florida concrete samples range from 0.00018 sq cm/sec to 0.0046 sq cm/sec, but air permeability coefficients are generally < 10 to the minus 11th power sq cm. Thus, advection through a concrete slab is negligible compared to diffusion. Finally, the report presents simple correlations for diffusion and permeability coefficients and for radon generation in concrete and entry into dwellings.		
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## **ABSTRACT**

Radon generation and transport through Florida residential concretes are examined for their contribution to indoor radon concentrations. Radium concentrations in the 11 concretes tested are all less than 2.5 pCi/g, and radon emanation coefficients are all less than 0.08. Measurements on the constituents of four of the concretes reveal that when the radium concentrations are greater than 1 pCi/g, it is generally due to elevated radium in the aggregate, but may occasionally occur from radium in the cement. Because the aggregates tested generally have very low emanation coefficients, elevated radium in the aggregate has a lesser impact on indoor radon than elevated radium in the cement component.

Diffusion coefficients for Florida concrete samples generally range from  $1.8 \times 10^{-4} \text{ cm}^2 \text{ sec}^{-1}$  to  $4.6 \times 10^{-3} \text{ cm}^2 \text{ sec}^{-1}$ . Air permeability coefficients are less than  $10^{-11} \text{ cm}^2$ . Thus, advection through a concrete slab is negligible compared to diffusion. Finally, simple correlations are presented for diffusion and permeability coefficients and for radon generation in concrete and entry into dwellings.

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## Section 1

### INTRODUCTION

Indoor radon entry has been modeled most commonly as advective transport by pressure-driven air flow from the soil through foundation openings or cracks. The flow is caused by the typically-negative indoor pressure compared with that in the soil and the outdoor atmosphere. Radon generated in the concrete floor and radon diffusion from the soil through the concrete floor has generally been ignored. Recently, attention has been directed toward the importance of diffusion as a significant mechanism for radon entry. In particular, Scott and Gordon (1) have identified radon diffusion through concrete as a possible significant source of indoor radon, and Tanner (2) identified radon diffusion as the dominant entry mechanism when foundation soil permeabilities are less than  $7 \times 10^{-12}$  m<sup>2</sup>. Rogers and Nielson (3) also identified diffusion through concrete floors and the contiguous soil as a significant mechanism for radon entry for many soils under typical long-term average foundation pressure gradients. Loureiro et al. (4) have compared theoretical diffusive and advective radon transport in soils to estimate conditions when diffusion is insignificant.

While the diffusive radon flux through concrete floors is much smaller than the advective flux through cracks in the floor, the predominance of the intact floor area over the crack area may compensate for the difference in fluxes. Thus, it is desirable to examine the diffusive properties of concretes used in dwelling floors to better assess this mode of radon entry. It also is instructive to characterize the relative importance of radon generated within the concrete to determine whether aggregates or other concrete components may contribute significantly to indoor radon concentrations. Very little relevant data on concrete exist in the general literature.

#### 1.1 SCOPE

This report characterizes the radon generating properties of Florida concretes. The work was conducted by Rogers & Associates Engineering Corporation as part of the Florida Radon Research Program (FRRP) cosponsored by the Florida State Department of Community Affairs and the U.S. Environmental Protection Agency. The parameters measured are the radium concentrations and emanation coefficients of Florida concretes and their constituents. The report also identifies the main properties of concrete that influence radon migration from the subsoil into dwellings. The parameters characterizing radon transport through concrete are the diffusion coefficient, the porosity, and the permeability coefficient. The report then examines the relation of the measured properties to other physical properties of the concretes. Finally, it examines the relative importance of the concrete properties, including radium concentrations, to radon entry into dwellings. The radon entry correlations are based on the laboratory data, on a simple indoor radon balance equation, and on a complete numerical analysis of combined diffusive-advective radon entry.

#### 1.2 BACKGROUND

The literature contains several references for the radium concentration (Ra) and radon emanation coefficient (E) in concretes. In 1981, a group at the National Bureau of Standards published a comprehensive review of relevant data prior to that time (5). Table 1-1 contains representative values of Ra and E from the literature. The radium-226 concentration is generally less than 1 pCi/g and the radon emanation coefficient is around 15 percent. Only the data in references 10 and 11 are for concretes in the United States. References 10 and 17 also report Ra and E values for concrete which contains phosphogypsum or phosphate slag. The last two entries of Table 1-1 contain the data for this type of



concrete. Although Ra contents were not reported for the slag in Reference 10, they must be less than 1 pCi/g. Reference 17 reports the same Ra concentration in the phosphogypsum as the concrete, even though the phosphogypsum comprises only 47 percent of the concrete.

**TABLE 1-1. PREVIOUS RADIUM CONCENTRATION AND RADON EMANATION COEFFICIENT MEASUREMENTS IN CONCRETE**

Year	Ra (pCi/g)	E (percent)	Comment	References
1971	2.4			6
1971	2.0			7
1974	0.4			8
1979		5-25	No measurement details given	9
1983	0.49 ± 0.19 <sup>a</sup>	21 ± 5 <sup>a</sup>	114 measurements	10
1985	1.0		Compilation of U.S. values	11
1986	0.54	20		12
1988	0.3-2.2	10-40	Only ranges are given	13
1989	0.78 ± 0.40 <sup>a</sup>		Compilation of worldwide values	14
1990	0.53-1.4	5.2-8.8		15
1991	0.54, 0.50	15, 4.2		16
1991	0.61 ± 0.50 <sup>a</sup>	14 ± 10 <sup>a</sup>	Concretes from several portland cement mixes	17
1983	0.20	16	Phosphate slag concrete	10
1991	19.2	9	Phosphogypsum concrete	17

a. Standard deviation of reported means.

Previously reported values for the radon diffusion coefficients,  $D$ , of concretes are listed in Table 1-2. Most of the data are reported as a diffusion length,  $L$ , where

$$L = \sqrt{D/\lambda} \quad (1-1)$$

and

- $L$  = diffusion length (cm)
- $D$  = diffusion coefficient ( $\text{cm}^2 \text{s}^{-1}$ )
- $\lambda$  = radon decay constant ( $2.06 \times 10^{-6} \text{s}^{-1}$ ).

Equation (1-1) was used to obtain the value of  $D$  when  $L$  was given in the literature. When available, the porosity,  $p$ , of the samples is also presented in Table 1-2.

**TABLE 1-2. PREVIOUS DIFFUSION COEFFICIENT MEASUREMENTS IN CONCRETE**

Year	Diffusion Coefficient $D$ ( $\times 10^4 \text{ cm}^2/\text{s}$ )	Porosity $p$ (percent)	Comments	Reference
1976	1.2-3.3	5-25	One combined $pD$ value measured, $p$ given as a range	18
1978	1.1	26	High density concrete	19
1983	3.3, 6.0	7, 32	High density concrete	20
1988	0.7-8.2		General review article	13
1988	0.7, 1.7		Low $w/c$ concrete for long-term vaults	21

The mean value of the measurements is  $2.8 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$ . This is probably lower than for typical concretes presently used in U.S. construction.

Data for the permeability coefficient of air in concrete,  $K$ , are available mainly because many types of concrete structures must be airtight under a specified internal pressure. Typical values of  $K$  for intact concrete range from  $10^{-14}$  to  $10^{-12} \text{ cm}^2$  (22,23).

### **1.3 REPORT CONTENTS**

Section 2 of this report contains the diffusion coefficient and permeability coefficient results as well as a description of the concretes tested. This section also contains D and K measurements made by Acurex (24) as part of the FRRP. Section 3 contains the radium concentration (Ra) and radon emanation coefficient (E) measurements and results for the concrete and for concrete constituents. Measurements on phosphogypsum concrete are reported in Section 4. The concrete correlations and supporting model analyses are presented in Section 5. The quality assurance of data for all analysis is presented in Section 6, and Section 7 contains a summary and conclusions.

## Section 2

### LABORATORY TESTS OF RADON TRANSPORT PROPERTIES

Radon diffusion (D) and permeability (K) coefficient tests were performed on samples from thirteen cylindrical concrete test specimens from Florida. Duplicate concrete samples were obtained from two of the test specimens, and two samples, M-5 and M-6, were made from a concrete mix from Florida, giving a total of 17 samples on which D and K measurements were made. Samples M-5 and M-6 have a processed gypsum pozzolan additive. The processed gypsum comprises 8 and 15 weight-percent of the mix, respectively. A detailed description of the preparation of samples M-5 and M-6 is given in Section 3.3.

#### 2.1 SAMPLE DESCRIPTION AND PREPARATION

The physical properties of the samples are given in Table 2-1.

TABLE 2-1. PHYSICAL PROPERTIES OF CONCRETE SAMPLES

Sample ID	Water/Cement Ratio	Density (g cm <sup>-3</sup> )	Porosity
F-1	0.60	2.15	0.20
F-2	0.60	2.14	0.21
F-3	0.61	1.99	0.26
F-4	0.61	2.00	0.26
M-5	0.53	2.15	0.20
M-6	0.36	2.30	0.15
C002F	0.53	2.11	0.22
C003F	0.67	2.00	0.26
C004C	0.66	2.08	0.23
C005C	0.58	2.06	0.24
TC1-C	0.60	2.19	0.17
TC1-1	0.60	2.17	0.18
TC2-4	0.60	2.18	0.17
CC008	0.60	2.12	0.26
CC013	0.56	1.94	0.28
CC015	0.56	1.97	0.27
CC033	est 0.57	2.12	0.21

The water-to-cement ratios (W/C) for the F, C, and CC series samples were supplied by concrete plants in Florida (24). The T series samples are from the radon entry test cells constructed and operated on the property of the Florida Institute for Phosphate Research in Bartow. The CC series are samples from

concrete slabs in Florida. Water-cement ratios were estimated for samples missing this information using the correlation (25):

$$W/C = 1.93 - 0.64d \quad , \quad (2-1)$$

where

$$d = \text{concrete density (g/cm}^3\text{)}.$$

If density values were not available, they were obtained from laboratory measurements of the mass and volume of the samples. The porosities were calculated from the relationship

$$p = 1 - d/G \quad , \quad (2-2)$$

where

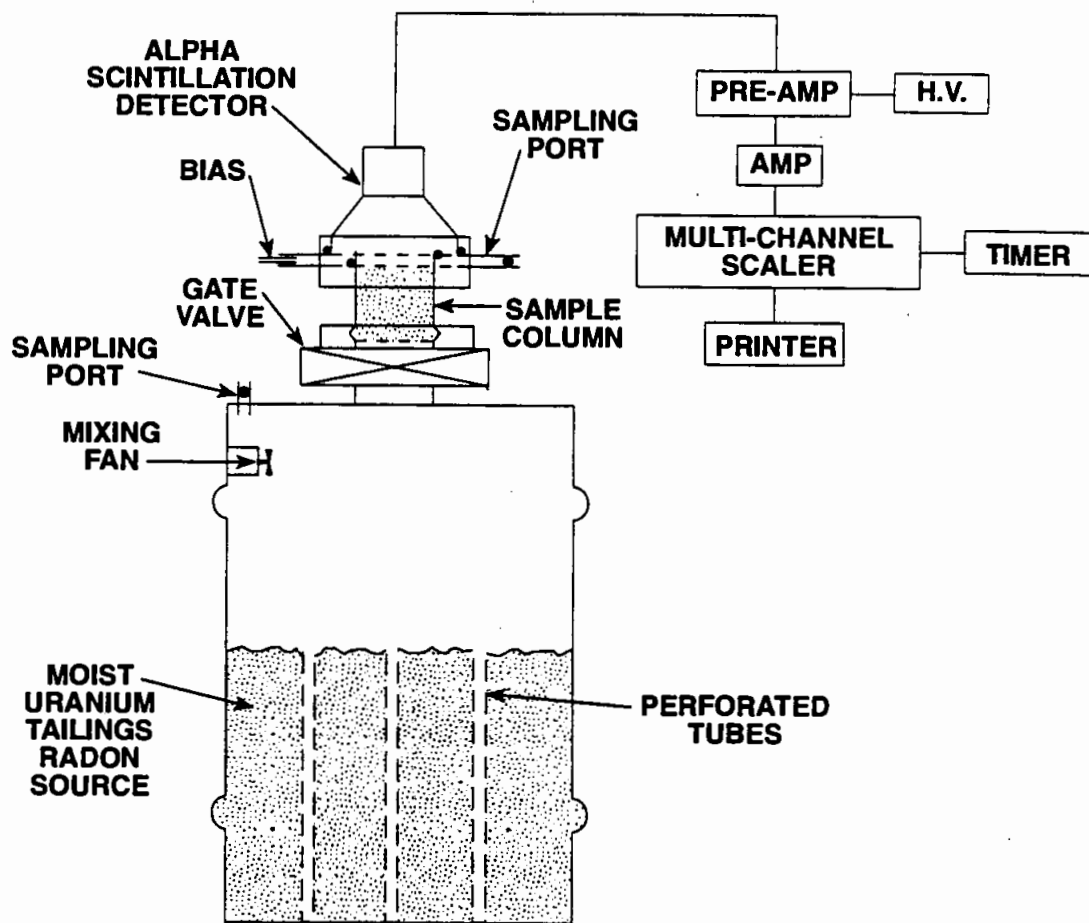
$$\begin{aligned} p &= \text{total sample porosity (cm}^3\text{/cm}^3\text{)} \\ G &= \text{solids density (assumed to be 2.7 g/cm}^3\text{)}. \end{aligned}$$

The concrete samples were 10 cm in diameter and ranged from about 5 to 10 cm in thickness.

To prepare the samples for the diffusion and permeability measurements, they were epoxied into standard diffusion sample holders (26) using an epoxy which has negligibly low diffusion and permeability coefficients. The air permeability measurements were also made with the concrete samples in the diffusion sample holder to minimize disruptive handling of the samples.

## 2.2 DIFFUSION COEFFICIENT MEASUREMENTS

Figure 2-1 contains a sketch of the equipment. The sample is placed on a large radon source and an alpha detector is placed on the top end of the sample. At zero time, the valve of the radon source is opened and the radon diffuses upward through the concrete sample and into the detector. The time dependence of the detector counts indicates the time dependence of the radon diffusion through the sample. The resulting measured diffusion coefficients are given in Table 2-2. The diffusion coefficients range from  $1.8 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1}$  to about  $4.6 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ . Uncertainties associated with the measurements range from 20 to 30 percent. Except for sample C002F, the D generally increases with increasing W/C ratio.



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**Figure 2-1. Time dependent radon diffusion apparatus.**

**TABLE 2-2. RADON DIFFUSION COEFFICIENTS OF FLORIDA  
CONCRETE SAMPLES**

Sample No.	D (cm <sup>2</sup> sec <sup>-1</sup> )	W/C
F-1	1.0x10 <sup>-3</sup>	0.60
F-2	2.9x10 <sup>-3</sup>	0.60
F-3	1.3x10 <sup>-3</sup>	0.61
F-4	1.2x10 <sup>-3</sup>	0.61
M-5	5.5x10 <sup>-4</sup>	0.53
M-6	2.9x10 <sup>-4</sup>	0.36
TC1-C	6.3x10 <sup>-4</sup>	0.60
TC1-1	2.2x10 <sup>-4</sup>	0.60
TC2-4	1.8x10 <sup>-4</sup>	0.60
C002 F	4.6x10 <sup>-3</sup>	0.53
C003 F	3.3x10 <sup>-3</sup>	0.67
C004 C	3.9x10 <sup>-3</sup>	0.66
C005 C	3.5x10 <sup>-3</sup>	0.58
CC008	3.1x10 <sup>-4</sup>	0.60
CC013	1.0x10 <sup>-3</sup>	0.56
CC015	1.0x10 <sup>-3</sup>	0.56
CC033	8.6x10 <sup>-4</sup>	0.57

### 2.3 PERMEABILITY COEFFICIENT MEASUREMENTS

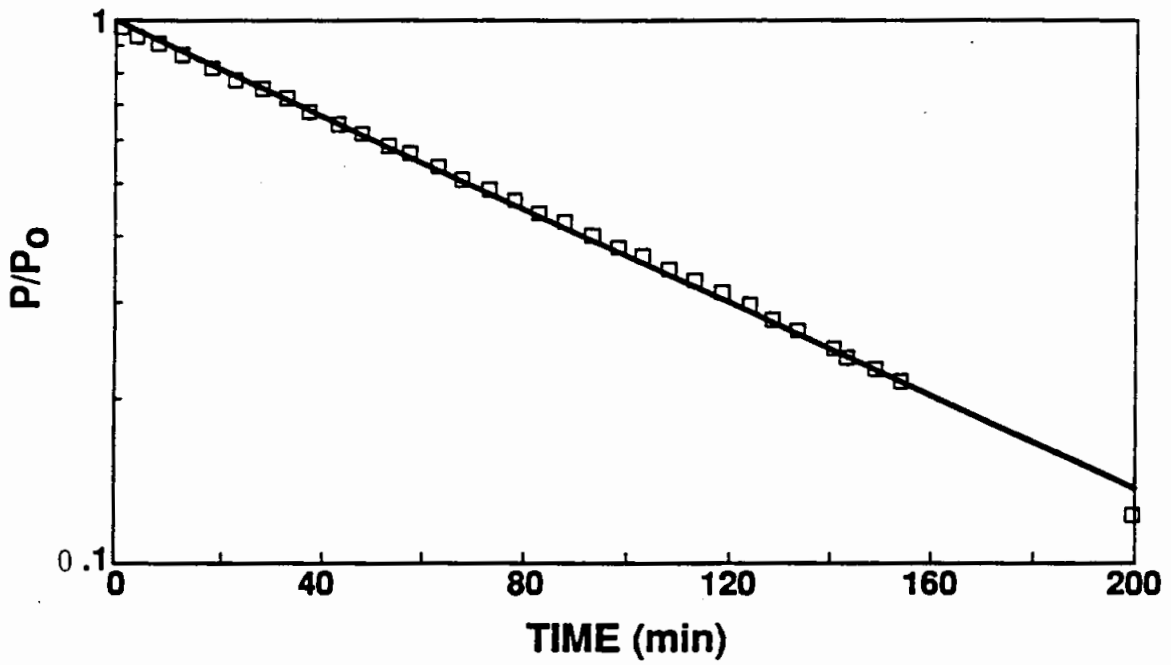
The permeability coefficients were measured using a procedure and equipment similar to that developed by Snoddy et al. (24). The technique measures the decrease in pressure with time in a pressurized air chamber due to air leakage through the concrete sample. The equipment consists of a small pressure vessel with the sample in its holder forming one end of the pressure chamber. Pressures in the pressure chamber were measured with a PX182-060GI pressure transducer and a DM160 data-logger with compatible accessories.

As shown in Figure 2-2, the pressure decreases with time according to the expression

$$P(t) = P_i \exp(-t/T) \quad , \quad (2-3)$$

where

- P<sub>i</sub> = initial pressure
- P(t) = pressure at time, t
- T = relaxation time.



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**Figure 2-2. Plot of pressure decrease with time for sample COO2F  
Resultant Permeability is  $3.4 \times 10^{12} \text{ cm}^2$ .**



Solution of the pressure balance equation reveals that (24):

$$T = \frac{\mu V_c x}{P_a K A} \quad (2-4)$$

or

$$K = \frac{\mu V_c x}{P_a T A} \quad (2-5)$$

where

- $\mu$  = viscosity of air
- $P_a$  = atmospheric pressure
- $V_c$  = enclosed volume under pressure
- $A$  = area of sample
- $x$  = concrete sample thickness.

This approach assumes a log-linear pressure drop throughout the concrete sample. Deviations from log-linearity increase the uncertainty associated with the reported K values.

The results of the permeability measurements are given in Table 2-3. In general the Florida concrete samples had very low permeabilities, with none having a value greater than  $7 \times 10^{-12} \text{ cm}^2$ . These values are consistent with the  $1.0 \times 10^{-14} \text{ cm}^2$  to  $3.0 \times 10^{-12} \text{ cm}^2$  values reported by Hansen et al. (23).

**TABLE 2-3. AIR PERMEABILITY COEFFICIENTS OF FLORIDA CONCRETE SAMPLES**

Sample No.	K (cm <sup>2</sup> )
F-3	$4.4 \times 10^{-12}$
M-5	$5.0 \times 10^{-13}$
M-6	$7.9 \times 10^{-13}$
TC1-C	$6.5 \times 10^{-12}$
TC1-1	$8.0 \times 10^{-14}$
TC2-4	$8.7 \times 10^{-14}$
C002 F	$3.4 \times 10^{-12}$
C003 F	$3.6 \times 10^{-12}$
C004 C	$4.0 \times 10^{-12}$
C005 C	$4.1 \times 10^{-12}$
CC008	$2.1 \times 10^{-13}$
CC013	$1.7 \times 10^{-12}$
CC015	$3.3 \times 10^{-12}$
CC033	$8.8 \times 10^{-13}$

## 2.4 RELATED DIFFUSION AND PERMEABILITY COEFFICIENT MEASUREMENTS

The D and K measurements by Snoddy (24), also part of the FRRP, were made with equipment similar to that used in the measurements reported here. The results of Snoddy's measurements are given in Table 2-4. In general, Snoddy's values for D and K are within a factor of two of the present results, which is within the experimental uncertainties of the measurements.

**TABLE 2-4. D AND K MEASUREMENTS BY SNODDY AS PART OF FRRP**

Sample No.	W/C	d (g/cm <sup>3</sup> )	p	pD (x10 <sup>4</sup> cm <sup>2</sup> /s) <sup>a</sup>	K (x10 <sup>13</sup> cm <sup>2</sup> )
C000	0.66		0.12	1.8	21
C001	0.61		0.20	3.8	71
C002A	0.53	2.09	0.19	3.1	8.8x10 <sup>+3b</sup>
C002B	0.53	2.09	0.20	3.1	4.6x10 <sup>+3b</sup>
C003A	0.67	2.04	0.15	2.6	20
C003B	0.67	2.07	0.17	3.8	20
C004A	0.66	2.14	0.14	3.8	13
C004B	0.66	2.15	0.14	3.8	11
C005A	0.58	1.96	0.17	2.6	21
C005B	0.58	2.04	0.14	2.1	9.4
C006	0.60			3.8	11
C007	0.60			4.6	6.9
C008	0.60			0.82	1.3
C010	0.56			4.6	10
C011	0.56			4.6	11
C012	0.56			9.9	25
C013	0.56			9.9	24
C014	0.56			3.8	11
C015	0.56			4.6	13
C016	0.56			6.6	14
C017	0.56			4.6	11
C018	0.56			5.6	7.2
C019	0.56			1.8	5.7
C020	0.56			4.6	9.1
C021	0.56			4.6	27
C022	0.56			3.8	23
C023				3.8	17
C024				8.2	16
C025				4.6	27
C026				4.6	23
C027				3.8	16

(continued)

**TABLE 2-4. (continued).**

<b>Sample No.</b>	<b>W/C</b>	<b>d (g/cm<sup>3</sup>)</b>	<b>p</b>	<b>pD (x10<sup>4</sup> cm<sup>2</sup>/s)<sup>a</sup></b>	<b>K (x10<sup>13</sup> cm<sup>2</sup>)</b>
C028				5.6	23
C030				3.8	10
C031				3.8	11
C032				2.6	11
C033				2.1	13
C034				6.8	17

- a. Snoddy reports only pD for samples C006-C034, and only p,d for samples C000 through C005B.
- b. Air passageways through these samples were sufficiently large that the airflow was audible.

## Section 3

### LABORATORY MEASUREMENTS OF RADIUM AND RADON EMANATION

Concrete floors in buildings generate radon that can enter the dwelling in addition to the radon transmitted from the underlying soils into the dwelling. The importance of the concrete floor and walls as an indoor radon source depends mainly on the radium concentration and the radon emanation coefficient (E) in the concrete. This section presents the results of radium-226 measurements and radon emanation coefficient measurements in Florida concretes and in concrete constituents.

#### 3.1 RADIUM AND EMANATION MEASUREMENT METHODS

Florida concrete and concrete constituents have relatively low Ra concentration and E. While the methods in the FRRP Procedures Manual (26) may be sufficient for the low Ra values expected if long counting times are used, they do not have sufficient sensitivity for the low E measurements. Consequently, new methods were used to measure the Ra and E of the Florida concretes and their constituents. These methods were developed for Ra and E measurements on the soils tested in the Mapping Project and are documented in Reference 27.

Basically, the Ra is determined using the sealed can gamma method with a large 7.6 cm by 12.7 cm NaI detector to increase counting efficiency and decrease sample counting time. The gamma counts under the radium decay-chain peaks are corrected for interferences from the thorium decay-chain gammas.

The emanation coefficient is determined by extracting the free radon from the sealed can into a Lucas cell and counting to determine the radon-222 concentration. Adjustments are made for pressure differences that occur with this method. Comparison tests demonstrate the significantly improved sensitivity of this method over the dual gamma counting method for measuring E.

#### 3.2 RADIUM AND EMANATION FOR CONCRETE SAMPLES

The Ra and E measurements were made on some of the Florida concrete samples used for the D and K measurements. The results are shown in Table 3-1. All Ra values are given in terms of dry weight. The Ra ranges from 1.0 pCi/g to about 2.4 pCi/g. These values are consistent with the values reported in Table 1-1.

The E values are less than 0.08 which is less than the average values given in Table 1-1. One difficulty occurred in measuring the E's for some of concrete samples, which calls into question the reported result for sample C002F. The air extraction needle used to penetrate the sealed can would occasionally become blocked by the surface of the concrete sample, thus restricting the air and radon flow into the Lucas flask. This could potentially decrease the measured value of E, and it is likely the cause of the very low E value for C002F.

**TABLE 3-1. Ra AND E VALUES FOR FLORIDA CONCRETE SAMPLES**

Sample No.	Ra (pCi/g)	E
C002F	1.60 ± 0.24	0.019
C003F	1.60 ± 0.06	
	1.18 ± 0.23	0.063 ± 0.012
	1.44 ± 0.05	
C004C	2.36 ± 0.06	0.057 ± 0.002
	2.25 ± 0.22	
C005C	1.69 ± 0.06	0.072 ± 0.003
	1.56 ± 0.24	
TC1-1	1.00 ± 0.10	0.075 ± 0.007
TC1-C	1.08 ± 0.09	0.078 ± 0.007
TC2-4	0.96 ± 0.10	0.070 ± 0.007

### 3.3 RADIUM AND EMANATION MEASUREMENTS FOR FLORIDA CONCRETE CONSTITUENTS

Dry concrete mixes were obtained from manufacturing facilities in the Jacksonville, Lakeland, Tampa, and Pensacola areas in order to measure the Ra and E of the constituents and compare them to the values for the mixed concrete. The concrete mixes were sieved to separate the aggregate, sand, and cement components. The relative compositions of the dry mixes are given in Table 3-2. Each component was then sealed for the Ra and E measurements. In addition, water was added to the samples M-1 through M-4 to form solid concrete samples with water-to-cement ratios of 0.50. Radium and radon emanation measurements were also made on these samples.

**TABLE 3-2. RELATIVE AMOUNTS OF CONSTITUENTS OF DRY FLORIDA CONCRETE MIXES**

Sample No.	Location	Percentage		
		Cement	Sand	Aggregate
M-1	Lakeland	16.4	45.0	38.6
M-2	Tampa	17.6	45.5	36.9
M-3	Jacksonville	14.6	77.3	8.1
M-4	Pensacola	21.7	43.5	34.8

The effect of a processed gypsum pozzolan was investigated using the concrete constituents. The relative amounts (weight percent) of constituents using the processed gypsum are given in Table 3-3. The Ra and E values for the concrete constituents are given in Table 3-4. The cement powders have the highest emanation coefficients, and, except for sample 1, the highest radium concentrations. Aggregates for samples M-1 and M-2 have radium contents exceeding 1 pCi/g; however, except for sample M-3, the aggregates have very low E values. The low E's decrease the importance of the radium as an indoor radon contributor. All the sands have low Ra and relatively low E.

**TABLE 3-3. CONSTITUENTS FOR POZZOLAN CONCRETES**

Constituents for Pozzolan Concretes (weight percent)					
Sample No.	Cement	Processed Gypsum	Sand	Aggregate	Water
M-5	10.8	7.2	45.1	27.0	9.9
M-6	13.5	13.5	31.5	31.5	9.9

The E values in Table 3-4 are reported for moist cement paste. This matches more closely the environment in the concrete. Emanation coefficients were also measured for dry cement powder. These measured emanation coefficients, given in the footnote to Table 3-4, also include the effects of radon adsorption on the cement powder, causing the E values to be extremely low (8). For example, if the adsorption coefficient for the dry cements is  $5 \text{ cm}^3 \text{ g}^{-1}$ , then the actual  $E_{dry}$  would increase by about a factor of 6 over the reported values.

**TABLE 3-4. RADIUM CONCENTRATIONS AND EMANATION COEFFICIENT MEASUREMENTS ON CONCRETE CONSTITUENTS**

Sample No.	Ra / (E) <sup>(a)</sup>		
	Cement	Sand	Aggregate
M-1	1.07 / (0.32) <sup>(b)</sup>	0.14 / (0.14)	1.82 / (0.04)
M-2	2.02 / (0.26) <sup>(b)</sup>	0.14 / (0.19)	1.29 / (0.03)
M-3	0.87 / (0.39) <sup>(b)</sup>	0.08 / (0.16)	0.10 / (0.17)
M-4	0.98 / (0.29) <sup>(b)</sup>	0.20 / (0.08)	0.17 / (0.04)

- a. Radium 226 concentrations in pCi/g, emanation coefficient given in parentheses.
- b. E values for moist cement paste. The respective E values for dry cement powder in samples M1, M2, M3, and M4 are 0.024, 0.020, 0.041, and 0.011.

The Ra and E values for the constituents of samples M-5 and M-6 are given in Table 3-5.

**TABLE 3-5. RADIUM AND EMANATION MEASUREMENTS FOR SAMPLES M-5 AND M-6**

Sample No.	Ra/(E) <sup>a</sup>			
	Cement	Processed Gypsum	Sand	Aggregate
M-5	1.3 (0.32)	0.55 (0.18)	0.14 (0.14)	0.52 (0.08)
M-6	1.3 (0.32)	0.55 (0.18)	0.14 (0.14)	0.52 (0.08)

a. Ra-226 concentrations in pCi/g, emanation coefficient given in parenthesis.

The Ra and E for the solid concrete samples are given in Table 3-6. Also shown in Table 3-6 are derived Ra and E values based on the Ra and E values of the constituents. All Ra values are expressed in units of pCi per gram dry weight. However, in obtaining the derived values, the bound water in the solid concrete samples is included in the computations. In general, the derived Ra values agree to within the experimental uncertainties with the measured values; however, they average about 13 percent lower. The E values also agree with the measured values to within experimental uncertainties. Except for M-5 and M-6 the derived values average about 17 percent higher. It is expected that the emanation values derived from constituents would be higher than the solid concrete sample values due to the chemical reactions and binding processes that decrease the porosity and increase the fraction of radon recoil atoms that are retained in adjacent particles.

**TABLE 3-6. RADIUM CONCENTRATION AND EMANATION COEFFICIENT  
VALUES FOR SOLID CONCRETE SAMPLES FROM DRY MIXES**

Sample No.	Ra (pCi/g)	E	Derived	
			Ra	E
M-1 A	1.24 ± 0.17 <sup>a</sup>	0.087 ± 0.012 <sup>a</sup>		
B	1.29 ± 0.16	0.087 ± 0.011		
C	1.18 ± 0.17	0.097 ± 0.014		
M-1 Ave.	1.24 ± 0.17	0.090 ± 0.012	0.93 ± 0.24	0.098
M-2 A	1.37 ± 0.17	0.081 ± 0.016		
B	0.94 ± 0.11	0.117 ± 0.014		
C	0.86 ± 0.12	0.178 ± 0.025		
M-2 Ave.	1.06 ± 0.14	0.125 ± 0.019	0.89 ± 0.27	0.132
M-3 A	0.47 ± 0.19	0.151 ± 0.061		
B	0.41 ± 0.15	0.169 ± 0.063		
C	0.61 ± 0.13	0.120 ± 0.026		
M-3 Ave.	0.50 ± 0.16	0.147 ± 0.053	0.20 ± 0.27	0.303
M-4 A	0.19 ± 0.14	0.407 ± 0.291		
B	0.36 ± 0.17	0.201 ± 0.094		
C	0.15 ± 0.09	0.562 ± 0.326		
M-4 Ave.	0.23 ± 0.14	0.390 ± 0.258	0.35 ± 0.24	0.191
M-5	0.52 ± 0.11	0.11 ± 0.21	0.43	0.15
M-6	0.63 ± 0.11	0.17 ± 0.02	0.51	0.15

a. Quoted uncertainties are one-standard deviation from Poisson counting statistics.



## Section 4

### **RADON TRANSPORT PROPERTIES OF CONCRETE CONTAINING PHOSPHOGYPSUM**

One of the project's objectives is to determine the properties of and impacts from concretes that have constituents elevated in radium. Phosphogypsum was selected as an additive to concrete constituents to investigate this effect.

Phosphogypsum is a by-product of the phosphate fertilizer industry's wet acid production of phosphoric acid. The main component of phosphogypsum is calcium sulfate dihydrate. According to Chang (28), recent studies have concluded that central Florida has in excess of 500-million tons\* of phosphogypsum currently stockpiled, and that by the year 2000 the total may exceed 1-billion tons. In recent years, efforts have been underway to find useful applications for this readily available and abundant resource.

One of the more promising potential uses for phosphogypsum is a cement additive. Preliminary research reported by Chang & Mantell (29) has shown that concretes made with phosphogypsum exhibit strength properties that are sufficient for many applications (such as road construction). This past research has focused primarily on the mechanical properties of phosphogypsum containing concretes, but relatively little work has been performed to examine the radiological properties of these concretes. Thus the properties of concrete which affect gaseous migration through concrete, permeability and diffusion coefficients, need to be characterized for concretes containing phosphogypsum. With the measured levels of activity in phosphogypsum, reported by Chang & Mantell (29), between 17 pCi/g - 25 pCi/g, radon generation and release from the concrete itself also needs to be addressed.

#### **4.1 BACKGROUND INFORMATION**

Research reported by Chang & Mantell (29) has shown that concretes containing phosphogypsum exhibit very good mechanical properties when produced under high compaction pressures, and when the correct amount of overall moisture is present. In order to test concretes which would be representative of the real world, selections were based on data reported by Chang & Mantell (29), and on conversations with Dr. W. F. Chang (30). Dr. Chang has conducted much of the initial research on phosphogypsum concretes at the University of Miami. The percentages of sand, aggregate, cement, and phosphogypsum were selected to yield a concrete with a compressive strength greater than 3,000 psi.\* The selections also provided concretes with different levels of phosphogypsum that will allow any differences in properties due to phosphogypsum to be noted and addressed.

The constituents used for the phosphogypsum concrete are the same as those given in Table 3-3, except the processed gypsum was replaced by phosphogypsum. The sets were chosen to represent realistic percentages that could be used by industry to provide a concrete with good properties. The phosphogypsum was supplied from the Southern Research Institute.

In order to produce a useful concrete containing phosphogypsum, the mixture must be compacted. The typical method for preparing impact compacted concrete is the Modified Proctor Method (30). The modified proctor method has been used in most of the initial characterization work on phosphogypsum concrete, and was used to produce samples for this study.

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(\*) 1TM = 907 Kg; 1Psi = 6.89 kPa.

## 4.2 SAMPLE PREPARATION AND CURING

Table 4-1 shows the radium concentrations, and emanation coefficients for the concrete constituents.

**TABLE 4-1. RADIOLOGICAL PROPERTIES OF PHOSPHOGYPSUM  
CONCRETE CONSTITUENTS**

Constituent	Ra (pCi/g)	E
Cement	1.32	0.32
Sand	0.14	0.14
Aggregate	0.52	0.08
Phosphogypsum	23.7	0.22

The phosphogypsum was tested for free moisture content. To prevent calcination of the phosphogypsum, a sample was dried at 85°C for two days to drive off any free moisture. The free moisture content of the phosphogypsum is 18.6 percent. The sand mixture was also tested for free moisture, and the results indicated that there was no free moisture in the sand.

The following procedure, prescribed by Chang, was used to mix the concrete specimens:

1. The phosphogypsum should be ground to minimize lumps.
2. Combine the appropriate amount of phosphogypsum with sand. The sand will help prevent the phosphogypsum from reforming lumps.
3. Mix in the correct amount of gravel to the mixture.
4. Add the cement powder to the mixture.
5. Add the correct amount of water to the mixture. The water should be added slowly to ensure that the entire mix is uniformly wetted. Be sure to include the free water and crystalline water in the calculation of the optimum moisture.
6. Compact the mixture according to the Modified Proctor Method (30).
7. Upon completion, wrap the samples in a plastic membrane to prevent water loss and cure at room temperature (approximately 25°C).

Four samples from each set of parameters were produced. Three of the four samples were for diffusion and permeability testing, and the fourth sample was used for radium content and emanation testing.

### 4.3 ANALYTICAL TESTS PERFORMED

#### 4.3.1 Diffusion Coefficients

The samples, three from each set, were tested to determine the radon diffusion coefficient of the concrete.

The results of the diffusion coefficients measurements completed to date are presented in Table 4-2.

**TABLE 4-2. DIFFUSION COEFFICIENT MEASUREMENT OF PHOSPHOGYPSUM CONCRETE**

Sample Identification	Diffusion Coefficient ( $\times 10^4$ $\text{cm}^2/\text{s}$ )	Permeability Coefficient ( $\times 10^{13}$ $\text{cm}^2$ )
SET 1, SAMPLE #1	3.5	7
SET 1, SAMPLE #2	8.6	7
SET 1, SAMPLE #3	4.9	9
SET 2, SAMPLE #1	18	200
SET 2, SAMPLE #2	35	210
SET 2, SAMPLE #3	10	200

The results for D from the phosphogypsum concrete fall within the range of measurements on regular Florida concretes, given in Table 2-2. The phosphogypsum does not appear to have a significant impact on the concrete's ability to hinder radon migration via diffusion.

#### 4.3.2 Permeability Tests

The permeability tests were performed on three samples from each parameter set. The results of the permeability tests are also given in Table 4-2.

The results for K from phosphogypsum concrete set 1 fall within the range of the previous tests. However, the results for set 2 are about a factor of five higher than the upper range of the K values for regular concretes given in Table 2-3. This is believed to be caused by small phosphogypsum clumps in the concrete.

### 4.3.3 Radium Content and Emanation

The radium content and emanation coefficient of the phosphogypsum concrete were measured. The results of the measurements are provided in Table 4-3.

**TABLE 4-3. RADIOLOGICAL PROPERTIES OF PHOSPHOGYPSUM CONCRETE**

<b>Sample</b>	<b>Radium Content (pCi/g)</b>	<b>Emanation Coefficient</b>
Set 1	2.3	0.18
Set 2	4.0	0.087

The radium content of the concrete is identical to the predicted values based on the constituent data of 2.3 pCi/g and 4.0 pCi/g for sets 1 and 2, respectively.

Therefore, the measurements for the concrete containing phosphogypsum yield radium concentrations slightly higher than for normal concretes, D values that are consistent with and K values that slightly exceed the ranges of values for normal concretes.

## Section 5

### DATA INTERPRETATION AND MODELING

Several correlations and simple models can be obtained from application of the measured data. This section identifies correlations for the water-to-cement ratio, the diffusion and permeability coefficients, and the radon entry from concrete floors into structures. The radon entry correlation is compared to radon entry from a concrete floor as calculated with the RAETRAD code (3).

#### 5.1 ESTIMATE OF WATER-TO-CEMENT RATIO

Frequently W/C of concrete is not known. For concretes that have about the same sand plus aggregate fraction, W/C should be related to the dry density.

The following linear least-squares regression resulting W/C with  $d$  was presented previously (25):

$$W/C = 1.93 - 0.64 d \quad , \quad (5-1)$$

where

$$\begin{aligned} W/C &= \text{water-cement ratio} \\ d &= \text{bulk dry density of concrete (g/cm}^3\text{)}. \end{aligned}$$

The correlation coefficient associated with this expression is  $r=0.90$ . Equation (5-1) was used to estimate the W/C for samples TC1-C, TC1-1, TC2-4, and CC033.

#### 5.2 ESTIMATE OF D FOR FLORIDA CONCRETES

Nielson and Rogers (25) have previously given a correlation between the measured values of  $D$  for concrete and then W/C ratio. The regression equation is

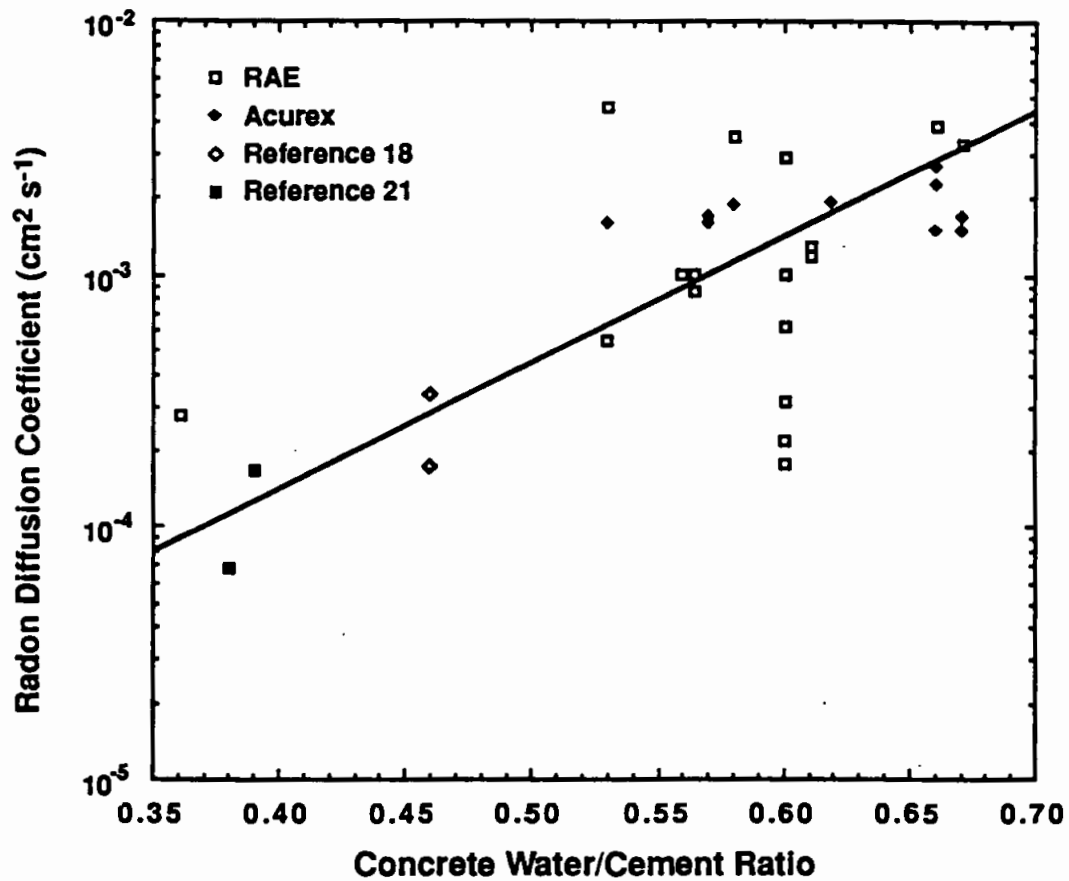
$$D = 1.5 \times 10^{-6} \exp(11.4 W/C) \quad . \quad (5-2)$$

The data and correlation are shown in Figure 5-1. The correlation coefficient for the  $\ln(D)$  vs. W/C regression is  $r=0.82$ . Data from the literature (18,12) are also included in Figure 5-1.

The linear relationship between W/C and  $d$ , given in Equation (5-1) suggests that a useful correlation also exists between  $D$  and  $d$ . Least squares regression correlating these variables gives (25):

$$D = 2.6 \times 10^4 \exp(-8 d) \quad , \quad (5-3)$$

with  $r=0.77$ , where as before the  $r$  applies to  $\ln(D)$  vs.  $d$  regression.



RAE-104531

**Figure 5-1. Regression of ambient-moisture radon diffusion measurements on the water/cement ratio of concrete.**

### 5.3 ESTIMATE OF K FOR FLORIDA CONCRETES

The permeability data do not exhibit the same definite trends with W/C as do the diffusion coefficient data. Much of the scatter in the data are due to experimental errors and uncertainties. Nevertheless, some benefit can be gained from a correlation between K and W/C. The least squares analysis gives (25)

$$K = 1.6 \times 10^{-16} \exp(15 W/C) \quad (5-4)$$

The correlation coefficient associated with this fit is  $r=0.75$ .

A slightly better fit is obtained for the correlation between K and d. This expression is

$$K = 0.22 \exp(-12.4 d) \quad (5-5)$$

with  $r=0.80$ . This correlation and associated data are shown in Figure 5-2.

### 5.4 INDOOR RADON ENTRY FROM FLORIDA CONCRETES

In general, the calculation of radon generation and transport through soil and concrete into dwellings is complex and involves multidimensional models such as RAETRAD (3). However, for Florida concretes, advection is negligible and the total radon generation rate per unit area is small compared to the radon generation rate per unit area in the subsoil. Under these conditions, the radon flux from the concrete floor can be estimated separately and can be added to the diffusive indoor flux from the subsoil (32). The flux from radon generated in the concrete is estimated by

$$F = Ra d E \sqrt{\lambda D} \tanh \left( \sqrt{\frac{\lambda}{D}} x_c \right) \quad (5-6)$$

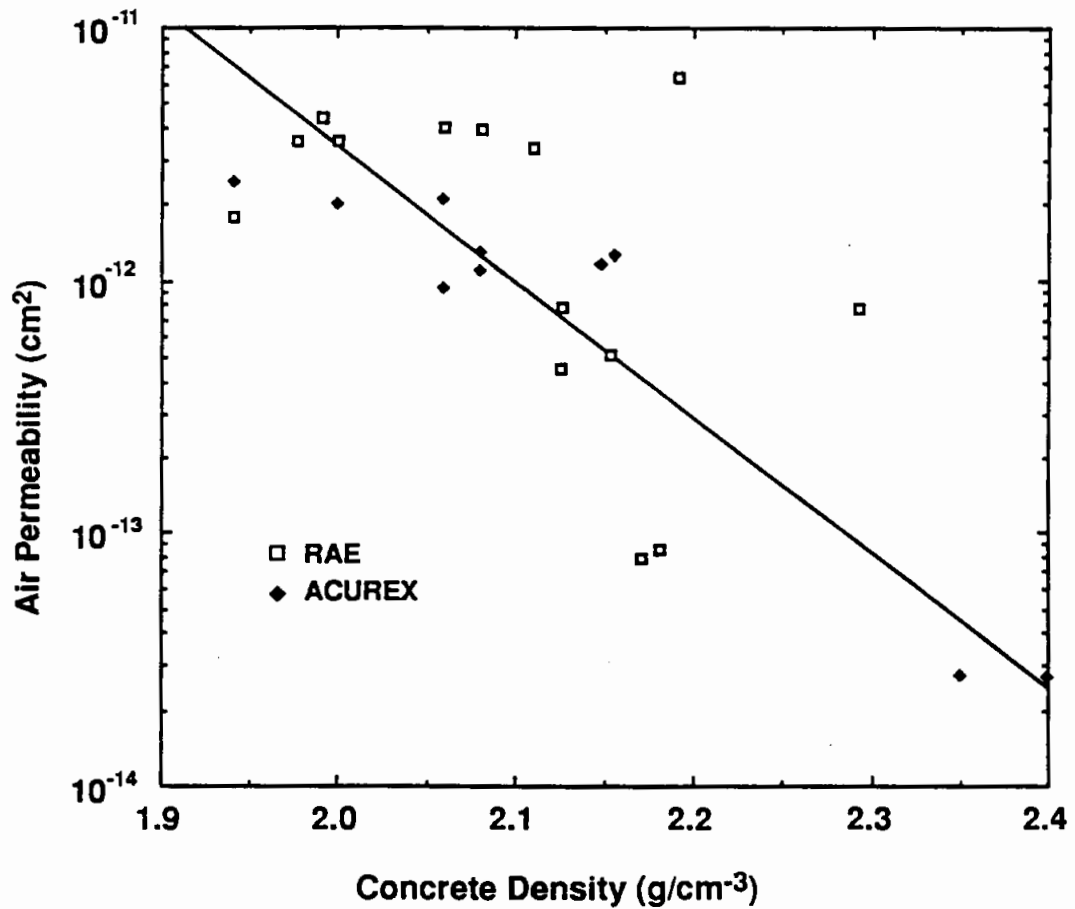
where

- Ra = Ra-226 concentration (pCi/g)
- $\lambda$  = radon decay constant ( $s^{-1}$ )
- $x_c$  = thickness of concrete slab (cm).

For the range of Florida concretes studied in the present work, d, E, D and  $x_c$  can be approximated by average values, so that Equation (5-6) can be expressed as

$$Q_c = Ra A_s / 28 \quad (5-7)$$

where



RAE-104532

**Figure 5-2. Regression of ambient-moisture air permeability measurements on the dry bulk density of concrete.**



- $Q_c$  = radon entry rate from concrete slab (pCi/s)  
 $A_s$  = area of concrete slab (m<sup>2</sup>)  
 28 = units conversion factor and constants (m<sup>2</sup> s/g).

Use of Equation (5-7) is illustrated for the concrete sample C004C. For Ra=2.31 pCi/g, and a house area of 141 m<sup>2</sup>, the radon entry rate from radon generation in the concrete is 13 pCi/s. For comparison, comprehensive RAETRAD calculations yield an entry rate of 13 pCi/s for radon generated in the concrete slab. This value is about 6 percent of the radon entry rate from the subsoil, where the subsoil is a loamy sand with a radium concentration of 2 pCi/g.

A simple estimate of the radon diffusion through the slab from the subslab radon is obtained in a similar manner as Equation (5-7)

$$Q_s = C_{ss} A_s / 2000 \quad , \quad (5-8)$$

where

- $Q_s$  = radon entry rate through concrete from subslab soil gas radon (pCi/s)  
 $C_{ss}$  = subslab radon concentration (pCi/L)  
 2000 = units conversion factor and constants (s L<sup>-1</sup> m<sup>+2</sup>).

The significance of indoor radon entry by diffusion through concrete floors can be estimated from a simplified approximation of the indoor radon balance equation. The approximation assumes that all indoor radon enters via the concrete foundation area, and that the indoor volume is uniformly diluted at the continuous rate of  $\lambda_v$  with clean air having an insignificant radon concentration:

$$(Q_c + Q_s) = C_{in} V \lambda_v \quad (5-9)$$

where

- $C_{in}$  = steady-state indoor radon concentration (pCi/L )  
 $V$  = indoor volume (L)  
 $\lambda_v$  = ventilation rate of indoor volume (s<sup>-1</sup>).

For a simple slab-on-grade house geometry typical of Florida construction, Equation (5-9) can be simplified further by introducing Equations (5-7) and (5-8) and setting the height of indoor volume equal to 2.3 m. This leads to the expression:

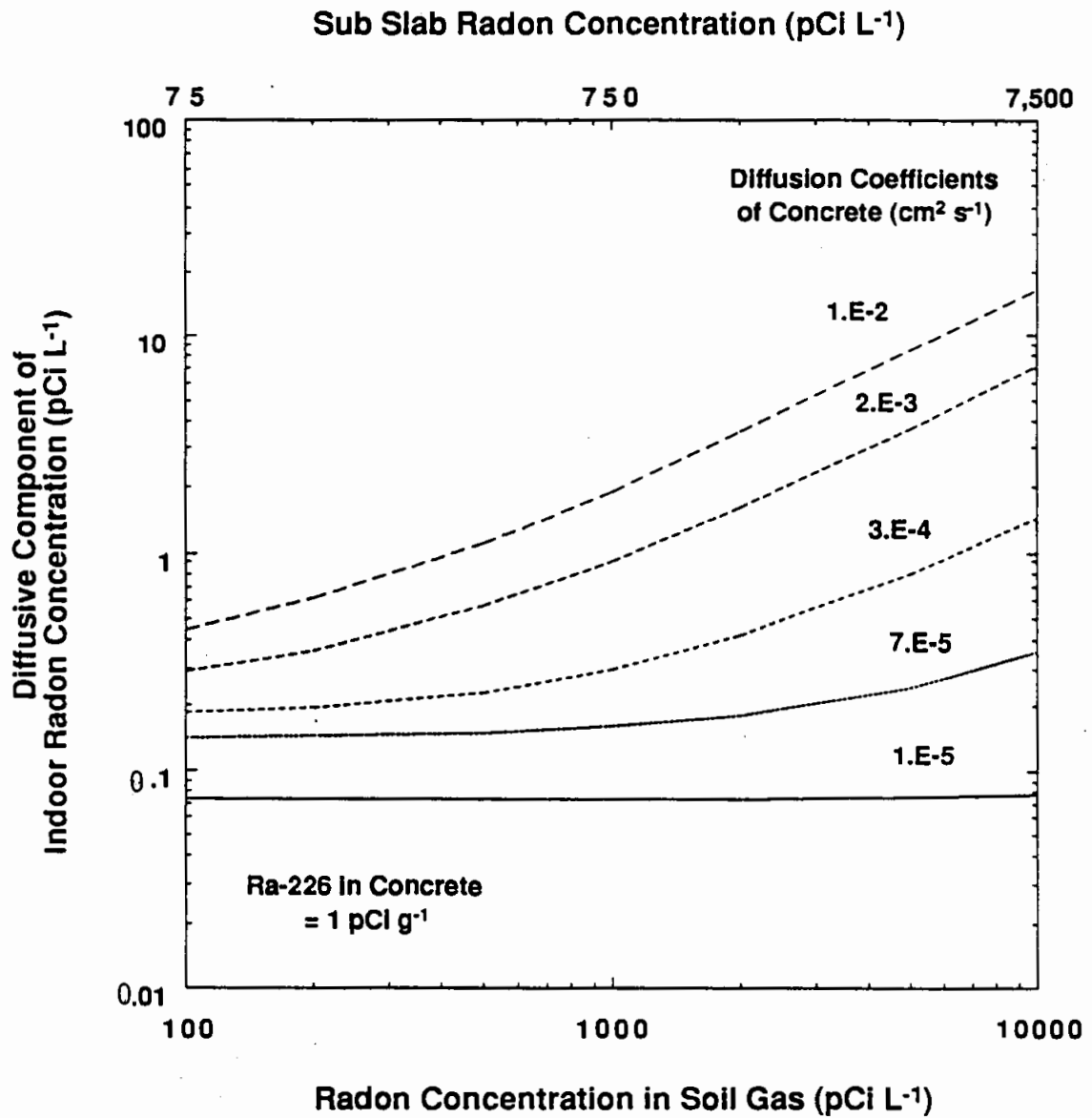
$$C_{in} = [15.5 Ra + 0.22 C_{ss}] / (1000\lambda_v) \quad (5-10)$$

where

- $\lambda_v$  = ventilation rate of indoor volume (ach).

Based on the approximate separability of diffusive and advective radon entry into dwellings, Equation (5-10) can be used directly to estimate the component of the indoor radon concentration that results from diffusion through and from the concrete floor slab. The diffusive radon flux through the slab was estimated by repeated analyses with the RAETTRAN code (33) in which a 10-cm slab separated an indoor radon concentration of 2 pCi/L from 5 m of sandy foundation soil that had varying source strengths corresponding to deep-soil radon concentrations of 100 pCi/L to 10,000 pCi/L ( $C_{ss} = 75$  pCi/L to 7,500 pCi/L). Various diffusion coefficients also were used for the slab, which had a fixed porosity of  $p=0.23$ . The radium concentration in the concrete first was assumed to be 1 pCi/g, and to have a radon emanation coefficient of 0.25.

The resulting diffusive component of the indoor concentrations, from the RAETTRAN code calculations, shown in Figure 5-3, start to exceed the 1 pCi/L level for elevated subslab radon concentrations (several thousand pCi/L) when concrete diffusion coefficients exceed about  $1 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$  or higher. The assumption of separability of diffusive and advective entry generally made a difference of about 5 percent in the entry rates. Even if a 1-cm perimeter floor crack is assumed for the house, diffusion rates through the intact part of the slab are affected by less than 25 percent, mainly from altered gradients near the perimeter.



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**Figure 5-3. Diffusive contributions to indoor radon concentrations for varying soil radon sources and five different radon diffusion coefficients.**

## Section 6

### QUALITY ASSURANCE FOR CONCRETE ANALYSIS

The quality assurance of all analyses is determined by three data quality parameters: precision, accuracy, and completeness. The following sections present the summary statistics of the analytical results in terms of these data quality parameters for radium assays, radon emanation, radon diffusion, and permeability coefficients. Completeness of the laboratory tests is estimated from the total number of measurements compared to the total number of samples available for testing. On this basis, the completeness percentage for all analyses was 100 percent.

#### 6.1 RADIUM CONCENTRATION MEASUREMENTS

There are no numerical data quality indicators for radium concentration measurements in concrete due to the lack of prior testing and of radium levels and variability in concrete. However, comparisons between the data quality indicators for soils (27) and concrete illustrate the precision of this method. The precision of the radium determinations is defined as the relative measurement uncertainty. Figure 6-1 presents all of the radium concentration measurements with their associated relative uncertainties. The relative uncertainties are computed from one standard deviation Poisson gamma-ray counting statistics and then divided by the measured radium concentration in order to express them in relative, dimensionless units for plotting. Thus  $10^{-1}$  in Figure 6-1 represents 10 percent standard deviation.

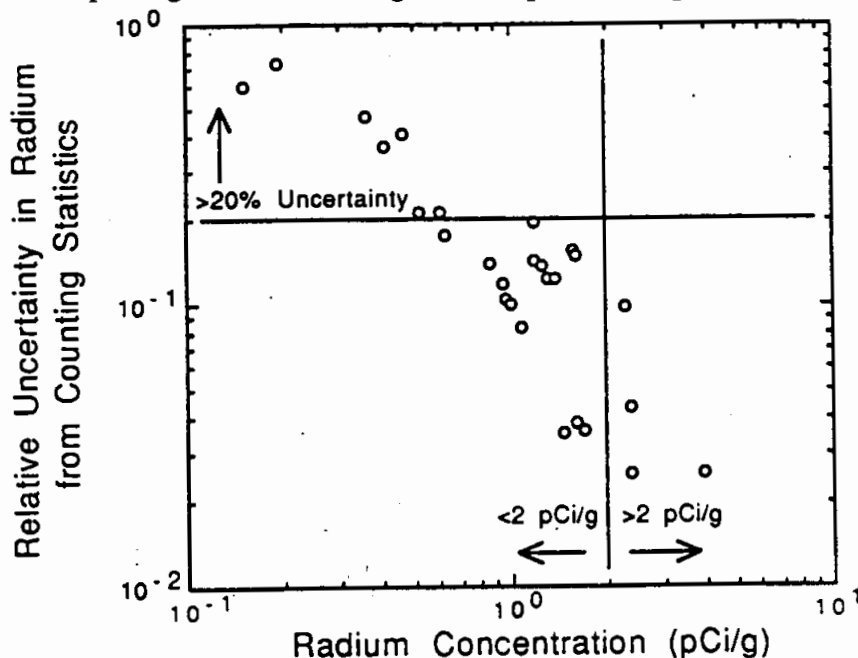


Figure 6-1. Relative uncertainties in radium determinations computed from gamma ray counting statistics as a function of radium concentration.

As Figure 6-1 illustrates, all the uncertainties fall below an uncertainty line of  $\pm 20$  percent at  $> 2$  pCi  $g^{-1}$ . As expected, numerous measurements are associated with higher uncertainties, but these are all in a radium range low enough to approach the detection limit.

A second estimate of the precision of the radium determinations is based on comparing the results of duplicate assays for a selected number of samples. Almost 34 percent duplicates were counted in the

radium determinations, which corresponds to 12 samples with duplicate counts. Table 6-1 summarizes these results. The final column lists the differences between the duplicate assay results and those given in the report. The average difference, indicating net bias, is -0.06 with a sample standard deviation of 0.22, using a Student's t test. Thus, the data are consistent with the hypothesis of a zero bias. The average absolute difference is 0.16 pCi g<sup>-1</sup>. The relative standard deviation between the duplicate measurements is 14.6 percent if the entire set of measurements above the detection limit is included. However, if only measurements above 1 pCi g<sup>-1</sup> range are included, the relative standard deviation between the duplicate pairs of measurements is only 6.0 percent. The relative standard deviation is computed as (34):

$$RSD_{dup} = \sqrt{2n \Sigma (x_1 - x_2)^2} / \Sigma (x_1 + x_2) \quad (6-1)$$

where

- RSD<sub>dup</sub> = relative standard deviation among duplicates
- x<sub>1</sub> = first observation
- x<sub>2</sub> = second observation
- n = number of pairs being compared.

**TABLE 6-1. COMPARISON OF DUPLICATE RADIUM ASSAYS TO ESTIMATE ANALYTICAL PRECISION**

Sample No.	Duplicate Assay Radium ± unc. <sup>a</sup> (pCi g <sup>-1</sup> )	Reference Radium (pCi g <sup>-1</sup> )	Difference (pCi g <sup>-1</sup> )
C002F	1.60 ± 0.24	1.60	0.00
C003F	1.18 ± 0.23	1.44	-0.26
C004C	2.36 ± 0.06	2.25	0.11
C005C	1.69 ± 0.06	1.56	0.13
M-1B	1.29 ± 0.16	1.24	0.05
M-1C	1.18 ± 0.17	1.24	-0.06
M-2B	0.94 ± 0.11	1.37	-0.43
M-2C	0.86 ± 0.12	1.37	-0.51
M-3B	0.41 ± 0.15	0.47	-0.06
M-3C	0.61 ± 0.13	0.47	0.14
M-4B	0.36 ± 0.17	0.19	0.17
M-4C	0.15 ± 0.09	0.19	-0.04
Average difference (pCi g <sup>-1</sup> )			-0.06
Average absolute difference (pCi g <sup>-1</sup> )			0.16
Relative standard deviation (all detected)			14.6 percent
Relative standard deviation (> 1 pCi g <sup>-1</sup> )			6.0 percent

a. One standard deviation uncertainty based on gamma-ray counting statistics.

The agreement of similar analyses with standard reference material demonstrates the accuracy of the radium concentration measurements. The Isotope Products Laboratory (IPL) reference material was

spiked into several powdered quartz sand aliquots to prepare the standard at  $1014 \pm 16$  pCi g<sup>-1</sup>. The material was sealed in a can similar to those used for the concrete samples and was analyzed regularly at the beginning and end of each batch of samples. Table 6-2 presents the results of these analyses.

**TABLE 6-2. ANALYSES OF STANDARD REFERENCE MATERIAL FOR <sup>226</sup>Ra**

<sup>226</sup> Ra ± S.D.	Difference from Reference	IPL Spike	<sup>226</sup> Ra ± S.D.	Difference from Reference
		(pCi g <sup>-1</sup> )		
1020 ± 1	6		1025 ± 2	11
1020 ± 2	6		1019 ± 1	5
1024 ± 2	10		1024 ± 2	10
1016 ± 2	2		1023 ± 2	9
1018 ± 2	4			
	Average ± Standard Deviation:		1021 ± 3	7 ± 3
	Average Relative Bias:		0.69 percent	

As Table 6-2 indicates, the accuracy of the IPL standard measurement compares favorably with the actual value and uncertainty of the reference material. The positive bias is not statistically significant at the 90 percent confidence level. The positive bias was relatively consistent for all measurements. This is probably the result of a small bias in either the calibration or reference radium concentration. An estimate of the average relative bias for the standard is positive at 0.69 percent. The estimate of the average relative bias is computed as:

$$\text{Bias} = 100[\Sigma(x_m - x_{ref}) / n] / x_{ref} \quad (6-2)$$

where

- Bias = average relative bias (percent units)
- x<sub>m</sub> = measured value (pCi g<sup>-1</sup>)
- x<sub>ref</sub> = reference value (pCi g<sup>-1</sup>)
- n = number of measurements.

Figure 6-2 presents a plot of the IPL standard analyses in a control chart format. The control chart illustrates the 3σ computed confidence interval shifted slightly toward the upper portion of the uncertainty range of the reference value for the standard. None of the individual measurements exceeded this interval, suggesting good control of measurement system operation and variations.

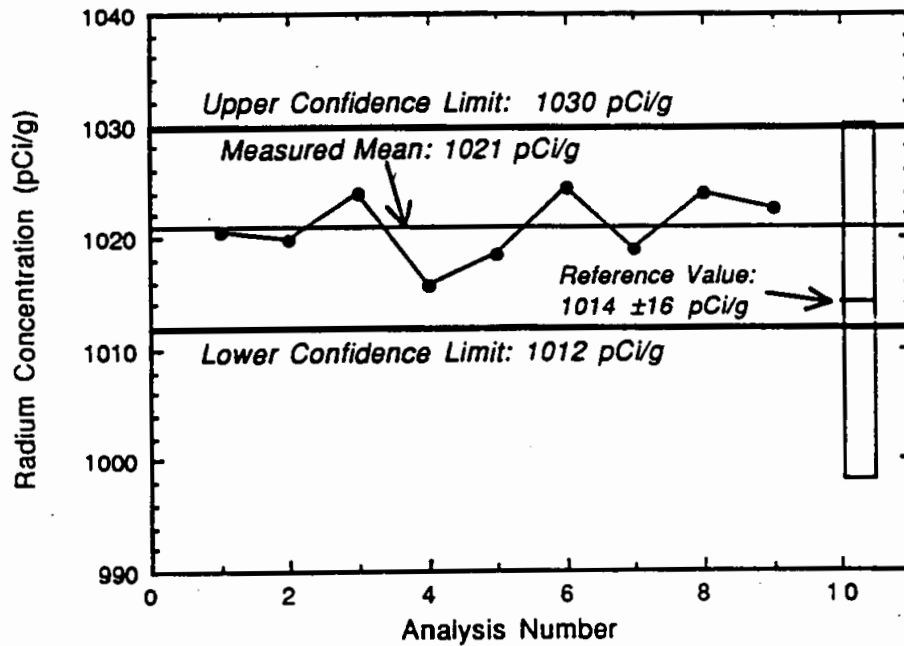


Figure 6-2. Individual radium measurements on the IPL standard in QC chart format.

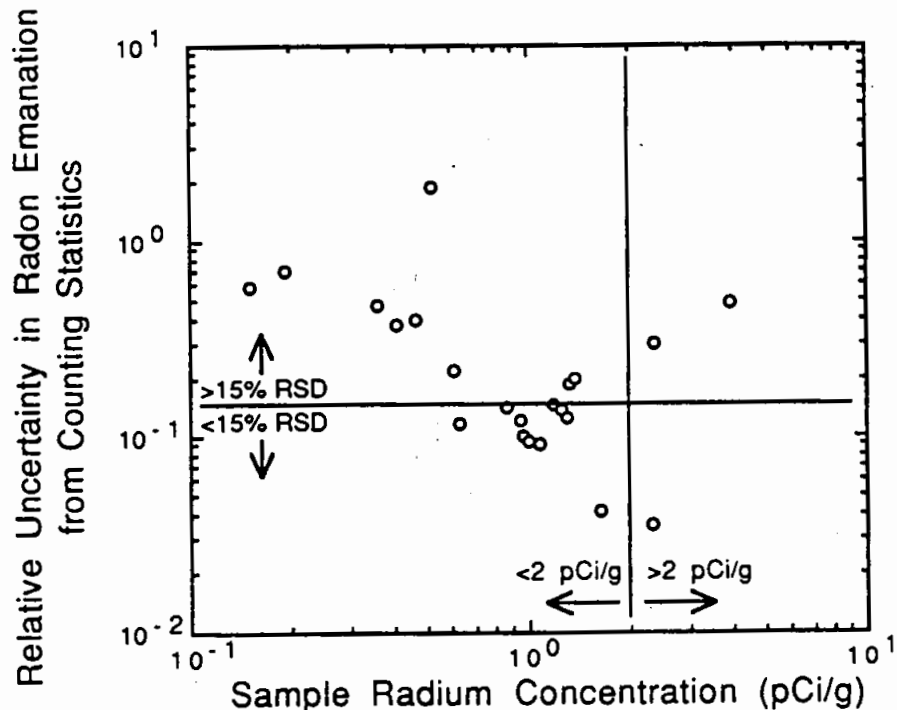
Analyses of blanks utilized a 300 g aliquot of onyx rock that had previously been determined by extended counting to contain negligible quantities of radium or thorium ( $<0.1 \text{ pCi g}^{-1}$ ). The blank sample was sealed in a can similar to those used for the concrete samples and was counted repeatedly during the sample analyses. Table 6-3 presents the results of these counts. The average measured quantity of radium in this sample is  $0.0 \pm 0.1$ , well within the measured standard deviation of  $0.2 \text{ pCi g}^{-1}$ .

TABLE 6-3. REPLICATE ANALYSES OF A BLANK SAMPLE FOR  $^{226}\text{Ra}$

$^{226}\text{Ra} \pm \text{uncertainty}$ ( $\text{pCi g}^{-1}$ )
$-0.1 \pm 0.3$
$0.2 \pm 0.3$
$0.1 \pm 0.3$
$-0.1 \pm 0.1$
$-0.2 \pm 0.1$
$0.0 \pm 0.1$
Average Measured Value: $0.0 \pm 0.1$
Average Uncertainties: 0.2

## 6.2 RADON EMANATION MEASUREMENTS

As with radium concentration measurements, there are no numerical data quality indicators for radon emanation measurements in concrete due to the lack of prior testing. However, comparisons between the data quality indicators for soils (27) and concrete illustrate the precision of this method. Measurements of radon emanation by the radon effluent method were analyzed for analytical precision by plotting their standard deviations computed from counting statistics versus the radium concentration. Standard deviations in the emanation measurements were expressed on a relative basis as  $\Delta E/E$  (uncertainty/mean). These precisions are plotted in Figure 6-3, illustrating the generally-successful achievement of the precision goal (only two data points in the upper right quadrant). Most of the samples that contained radium concentrations less than 2 pCi g<sup>-1</sup> had relative standard deviations greater than 15 percent due to the increased uncertainty in measuring the emanation coefficient. This is illustrated in the figure below by observing the data points that appear in the upper left quadrant.



**Figure 6-3. Relative uncertainties in radon emanation determinations by the radon effluent method, as computed from gamma ray counting statistics.**

Another estimate of the precision of the radon emanation determinations was demonstrated by replicate tests conducted on samples M-1, M-2, M-3, and M-4. The samples were divided into three separate concrete plugs (A, B, and C) and were sealed in cans as described previously. These results are presented in Table 6-4. The standard deviations for all samples, excluding set M-4, do not exceed 4 percent which indicates a high degree of precision between the duplicate measurements. Sample M-4,



however, had a relative standard deviation of 14.8 percent which is due to the extremely low radium content and thus a high associated relative uncertainty as shown previously in Table 6-1.

**TABLE 6-4. COMPARISON OF DUPLICATE RADON EMANATION MEASUREMENTS**

Sample No.	Measured Emanation Coefficient	Relative Standard Deviation (percent)
M-1A	0.087 ± 0.012	
M-1B	0.087 ± 0.011	0.5
M-1C	0.097 ± 0.014	
M-2A	0.081 ± 0.016	
M-2B	0.117 ± 0.014	4.0
M-2C	0.178 ± 0.025	
M-3A	0.151 ± 0.061	
M-3B	0.169 ± 0.063	2.0
M-3C	0.120 ± 0.026	
M-4A	0.407 ± 0.291	
M-4B	0.201 ± 0.094	14.8
M-4C	0.562 ± 0.326	

The accuracy of the radon emanation measurements could not be directly evaluated because there are no emanation standards for concrete materials. The accuracy of this method was found acceptable for soil samples in a previous report (27).

### 6.3 DIFFUSION COEFFICIENT MEASUREMENTS

The precision of the transient radon diffusion measurement technique was directly evaluated by replicate measurements performed on samples F-1, F-2, F-3, and F-4. Sample set F-1 and F-2, as well as set F-3 and F-4, originated from the same concrete core and were cut in half to permit duplicate testing.

Table 6-5 lists measurements with their associated uncertainties. The uncertainties are based on earlier reported values that analyzed numerous soil sample replicates for diffusion coefficients with varying

moisture saturations (35). The samples with moisture saturation levels below 50% were observed to have uncertainties around 12 to 20%. Uncertainties increased to about a factor of two as full saturation was reached. The uncertainties associated with the diffusion measurements range from 20 to 30%.

**TABLE 6-5. COMPARISON OF DUPLICATE RADON DIFFUSION COEFFICIENTS**

Sample No.	Measured D ( $\times 10^3 \text{ cm}^2 \text{ s}^{-1}$ )
F-1	$1.0 \pm 0.3$
F-2	$2.9 \pm 0.9$
F-3	$1.3 \pm 0.4$
F-4	$1.2 \pm 0.4$

The results from the duplicate samples, F-3 and F-4, are well within estimated experimental error. Comparing samples F-1 and F-2, however, reveals that the diffusion coefficient for the latter is almost a factor of three higher. This difference may be influenced by settling of aggregate in the cement core from which the specimen was taken.

In order to verify the accuracy of the diffusion coefficient measurements, two diffusion tests were conducted using dry sand and one using uniform glass beads. These diffusion measurements allow comparisons with theoretically-derived diffusion coefficients. As Table 6-6 indicates, the percent difference between these standard measurements does not exceed 7%.

**TABLE 6-6. DIFFUSION MEASUREMENTS ON STANDARD REFERENCE MATERIALS**

Sample No.	Measured D ( $\text{cm}^2 \text{ s}^{-1}$ )	Standard D ( $\text{cm}^2 \text{ s}^{-1}$ ) <sup>a</sup>	Percent Difference
Dry Sand	0.059	0.063	6.3
Dry Sand	0.064	0.063	1.6
Glass Beads	0.070	0.071	1.4

a. Reference 35.

#### 6.4 PERMEABILITY COEFFICIENT MEASUREMENTS

The methods for determining air permeability coefficients are similar to those developed by Snoddy et al. (24). Since these procedures are rather new, limits of precision and accuracy are still considered experimental. In order to verify the precision of data, duplicate analyses were conducted on samples CC013 and CC033. Table 6-7 summarizes these results.

**TABLE 6-7. COMPARISON OF DUPLICATE AIR PERMEABILITY COEFFICIENTS**

Sample No.	Measured K (cm <sup>2</sup> )	Percent Difference
CC013	1.7x10 <sup>-12</sup>	5.6
CC013	1.8x10 <sup>-12</sup>	
CC033	8.8x10 <sup>-13</sup>	3.4
CC033	8.5x10 <sup>-13</sup>	

The duplicate permeability coefficients demonstrate the precision of this technique with respect to the reproducibility of its measurements. The differences in these values do not exceed 6% which signifies a strong agreement between the duplicate tests.

The accuracy of the permeability data cannot be directly measured due to the lack of test standards for this new procedure. The simple equation used to determine the permeability coefficient assumes that the log of the pressure drop throughout the concrete sample is linear with respect to time. This is not necessarily valid for some initial pressure ranges. However, using this assumption, deviations from log-linearity increase the uncertainty associated with the reported K value. A correlation coefficient, r, was calculated for each sample by comparing the pressure drop to a log-linear best fit curve as shown in Figure 2-2. The values for r ranged from about 0.94 to 1.00, demonstrating the excellent correlation between the data points for each measurement. The linearity of the pressure drop on a semi-log scale gave an indication of any leakage in the measurement system that would suggest the need for retesting.

## Section 7

### SUMMARY AND CONCLUSIONS

The Florida concretes tested generally have radium concentrations less than 3 pCi/g, and emanation coefficients less than 0.08. These values are consistent with literature values for a wide variety of concretes. When the radium concentration exceeds 1 pCi/g it may either be due to the radium in the cement or the aggregate. However, the aggregate has very low E values, rendering its radium less important than radium in the cement component.

The measured diffusion coefficients for the Florida concretes ranged from about  $2 \times 10^{-4}$  to  $5 \times 10^{-3}$   $\text{cm}^2\text{s}^{-1}$ . These values are consistent with previous values in the literature, but extend the upper limit of the range by about a factor of five. The measured air permeability coefficients ranged from about  $9 \times 10^{-14}$  to  $7 \times 10^{-12}$   $\text{cm}^2$ . The D and K values measured by Snoddy in a cooperative effort are consistent with the present measured values.

The correlations that are presented for W/C, D, K, and  $Q_c$  are useful and provide sufficient accuracy for general scoping studies. Example calculations of radon entry into a dwelling indicate that concretes with radium content less than about 2 pCi/g contribute less than 10 percent of the total radon entry.

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