RADON AND ALDEHYDE CONCENTRATIONS IN THE INDOOR ENVIRONMENT

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1981-04-01

LBL-12590
UC-41

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

ENERGY & ENVIRONMENT DIVISION

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D.J. Moschandreas and H.E. Rector

April 1981

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

LBL-12590
EEB-Vent 81-12

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for

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Berkeley, CA 94720

April 1981

Prepared under UCLBL PO No. 7052902 for: The Energy Efficient Buildings Program, Energy and Environment Division, Lawrence Berkeley Laboratory. Principal Investigators: Craig Hollowell and Arthur Rosenfeld.

This work was supported by the Assistant Secretary for Conservation and Solar Energy, Office of Buildings and Community Systems, Buildings Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

GEOMET Report Number ES-844

September 19, 1980

RADON AND ALDEHYDE CONCENTRATIONS IN THE INDOOR ENVIRONMENT

Final Report

for

Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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INTRODUCTION

This report summarizes GEOMET Technologies, Incorporated (GTI) findings regarding indoor air contaminants in the energy-efficient residence (EER) in Mt. Airy, Maryland. The objectives of the study were to:

- Collect relevant air quality samples (specifically radon and aldehydes),
- Analyze aldehyde samples with GTI laboratory facilities, and
- Characterize the indoor air quality with respect to radon and aldehydes and develop relationships between air infiltration rates and contaminant levels.

The radon samples were analyzed by the Lawrence Berkeley Laboratory and operational characteristics of the test residence were secured by NAHB.

Section 1.0

FORMALDEHYDE

1.1 Measurement Techniques

Aldehyde sampling employed instrumentation and analytical techniques developed by the Lawrence Berkeley Laboratory (LBL). The sampling apparatus secured indoor/outdoor pairs of 24-hour samples of total aldehydes and formaldehyde. Total aldehydes were sampled using impingers filled with MBTH; formaldehyde was sampled using impingers filled with distilled water. The impingers were kept cool inside a refrigerator that was an integral part of the sampling system. Timing was automatically controlled. The samples were shipped to the GTI analytical facility via overnight express freight in chilled containers. The samples were analyzed under protocol defined and developed by LBL. Formaldehyde concentrations were determined using the pararosaniline technique (Miksch, 1980); total aldehyde concentrations were determined using the MBTH technique as modified by LBL (LBL, 1980a).

1.2 Data Presentation

The data obtained in the course of the study are given in Table 1.1. Clearly, 15 24-hour measurements do not constitute a sufficiently large data base for rigorous statistical analyses, yet a careful review of the data reveals trends establishing cause and effect relationships.

The maximum formaldehyde (HCHO) concentration measured was 0.14 mg/m^3, the average concentration was 0.06 mg/m^3. A comparison with existing indoor standards (see Table 1.2) indicates that 20 per cent of the observed indoor HCHO concentrations are within the range that should cause concern.

Table 1.1 Data for Aldehydes

Run No.	Date	Time (EST)	RCHO (mg/m³)	HCHO (mg/m³)	H/R	Basement RH (%)	Living Room RH (%)	Basement Temp. (^{0}F)	Living Room Temp. (^{0}F)	Nominal Air Changes per Hour
1	8/24	1234	0.18	0.10	0.56	75	63	—	—	0.04
2	8/31	1012	0.22	0.12	0.55	73	58	—	—	0.04
3	9/18	1337	0.21	0.14	0.67	74	60	—	—	0.04
4	10/25	1255	0.08	0.03	0.38	45	50	60	60	0.40*
5	11/5	1457	0.15	0.06	0.40	55	45	60	65	0.30
6	11/9	1533	0.19	0.05	0.26	50	55	60	68	0.30
7	11/16	1427	0.18	0.06	0.33	50	45	60	70	0.13
8	11/21	1639	0.20	0.04	0.20	40	45	65	68	0.60
9	11/26	1527	0.23	0.08	0.35	60	65	65	65	0.30
10	12/3	1620	0.14	0.08	0.57	42	45	65	60	0.12
11	12/10	1507	0.10	0.07	0.70	45	45	65	65	0.12
12	12/17	1603	0.10	0.02	0.20	30	35	55	55	0.41
13	1/2	1550	0.09	0.03	0.33	35	48	65	70	0.33
14	1/8	1540	0.13	0.04	0.31	30	35	65	65	0.29
15	1/16	1148	0.13	0.05	0.38	30	35	65	70	0.30

* Heat exchanger on timing cycle: 0.8 ACH^{-1} ventilation + infiltration for 10 hours
0.13 ACH^{-1} infiltration for 14 hours, giving nominal 0.4 ACH^{-1} per day.

Table 1.2. Ambient, Occupational and Indoor Formaldehyde Standards

Country	Ambient Air Standards			Occupational Standards			Indoor Residential Standards		
	Proposed	Recommended	Promulgated	Proposed	Recommended	Promulgated	Proposed	Recommended	Promulgated
United States		0.1 ppm 0.12 mg/m^3 (ACGIH)			1 ppm - 1.2 mg/m^3 30 minutes sampling period max (NIOSH) 2 ppm - 3 mg/m^3 ceiling (ACGIH) 0.1 ppm 90 to 1000 days 0.2 ppm - continuous 0.8 - alert 5.0 ppm - short	0.5 ppm - 90 days 1.0 ppm 24 hour 3.0 ppm 1 hour emergency Nuclear Submarine 3 ppm - 8 hour TWA 5 ppm - MAC 10 ppm - MAK-K (OSHA) Occupational Standard Space Flight			
Wisconsin, U.S.A.							0.4 ppm - (0.48 mg/m^3)* 0.2 ppm - (0.24 mg/m^3)**		
Bulgaria						1.0 mg/m^3 MPC			
Czechoslovakia						2 mg/m^3 average 5 mg/m^3 MAC			
Finland						2 ppm - 3 mg/m^3 ceiling			
German Democratic Republic						2 mg/m^3 MAK-D 2 mg/m^3 MAK-K			
Federal Republic of Germany			0.03 mg/m^3 MIK		1 ppm 1.2 mg/m^3			0.1 ppm MAC	
Hungary						1 mg/m^3 TWA			
Italy					1.5 mg/m^3 TWA				
Japan					2 ppm - 2.5 mg/m^3 TWA				
Netherlands					2 ppm - 3 mg/m^3 TLV			0.1 ppm MAC	
Poland						2 mg/m^3 ceiling			
Romania					4 mg/m^3 (undetermined)				
Sweden					2 ppm 3 mg/m^3 ceiling			below 0.4 acceptable 0.4 < 0.7 acceptable without irritation > 0.7 unacceptable < 0.1 maximum for new building	
Switzerland						1 ppm - 1.2 mg/m^3 MAC			
USSR		Undetermined 0.1 mg/m^3 - -0.035 mg/m^3				0.5 mg/m^3 MAC			
Denmark						1.0 ppm TLV		0.12 ppm MAC	

NOTES: TWA - time weighted average
MPC - maximum permissible concentration
MAC - maximum allowable concentration
TLV - threshold limit value
MAK D - maximum average concentration, 8 hour 15 min. work period
MIK - German outdoor air standard

OSHA - Occupational Safety and Health Administration
ACGIH - American Conference of Government Industrial Hygienists
NIOSH - National Institute for Occupational Safety and Health

* Strictly for mobile homes
** After May 1, 1981

The history of monitoring for total aldehydes and formaldehyde is illustrated in Figure 1.1.

The study evolved in two phases: Phase A when the heat exchanger was generally not operating, and Phase B when a heat exchanger was operating.

As indicated by the ratio variation (Figure 1.2), the formaldehyde portions of total aldehydes varied considerably over the experimental period. An effort to associate this variation with indoor smoking was not fruitful. A relationship has been identified between HCHO concentrations and the HCHO-to-RCHO ratio (see Figure 1.2). The resulting correlation coefficient of 0.774 explains 60 percent of the variance.

A major source of formaldehyde is 1200 ft.^{2} of plywood, which constitutes the subfloor of the structure. The area of kitchen cabinet chipwood has been estimated at 200 ft.^{2} and should also be considered a major indoor source. The residence was built in 1977, thus the observed elevated formaldehyde concentrations are attributed to a low emission rate from these sources and indoor accumulation due to extremely low air infiltration rates.

Variation of formaldehyde concentrations in room air depends, among other factors, on temperature, relative humidity and air infiltration rates. (Andersen et al, 1975 and others). The experimental design of the present study controlled only the air infiltration rate, it varied from a low value of 0.04 to 0.1 air changes per hour (ACH^{-1}) to as much as 0.8 ACH^{-1} when augmented by mechanical ventilation through air to air heat exchangers. Temperature (living room) averaged 65°F, varying from a low of 55° to a high of 70°F. Relative humidity varied over a much broader range, from 35% to over 65%.

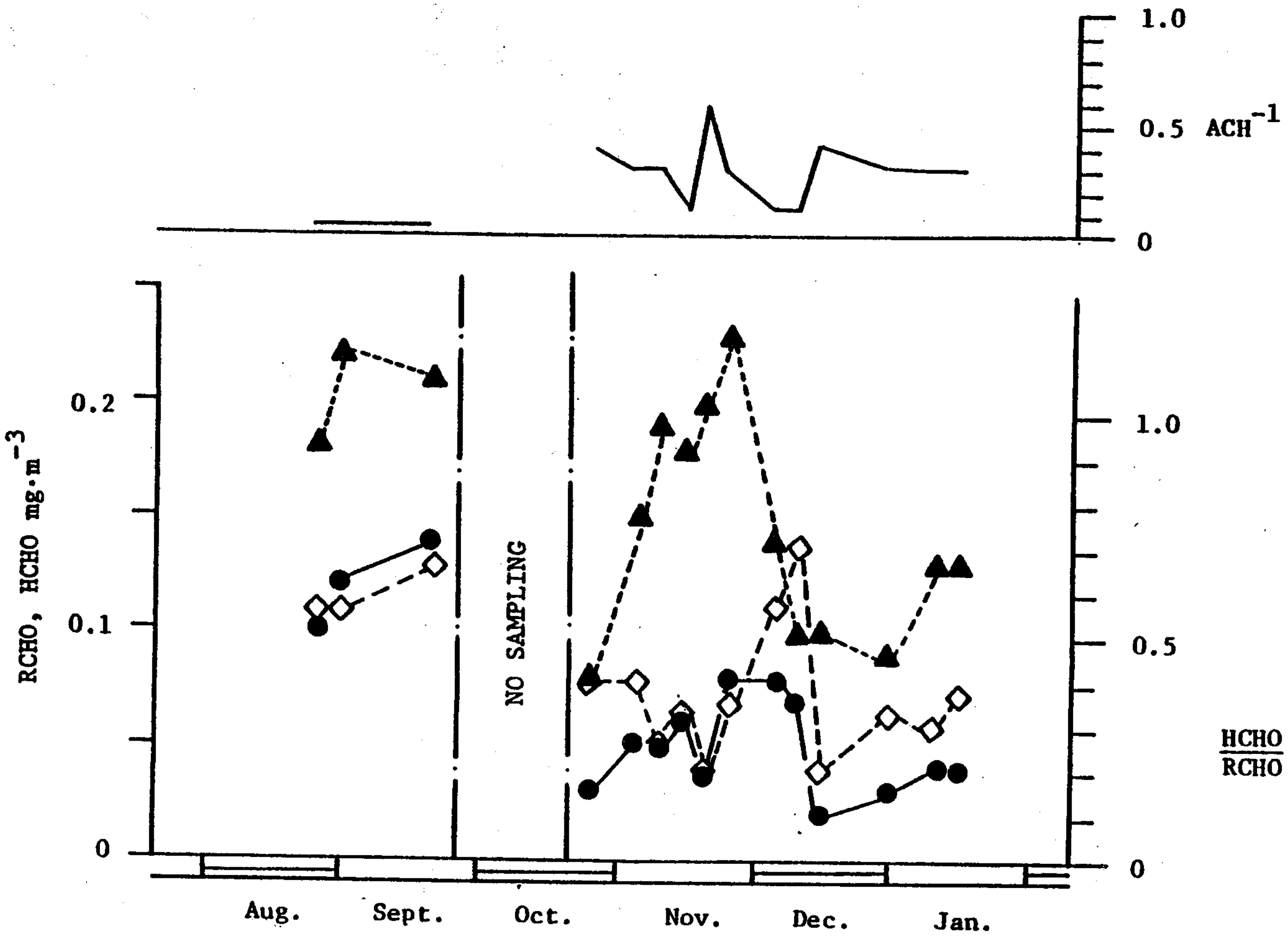


Figure 1.1 Monitoring History of Aldehydes

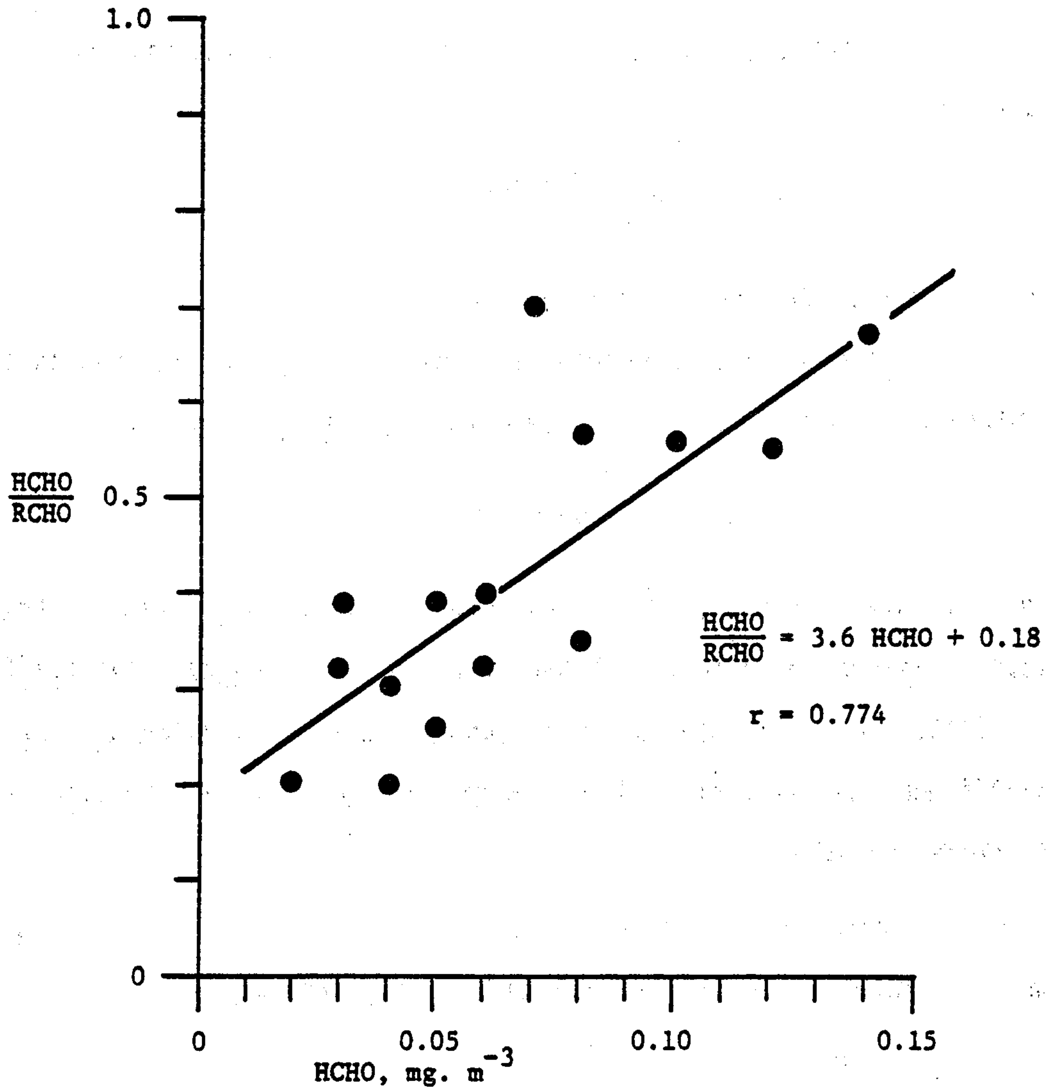


Figure 1.2 Formaldehyde and Total Aldehydes

As shown in Figure 1.3a, formaldehyde concentrations displayed no obvious relationship with temperature. Formaldehyde response to varied relative humidity was more apparent (see Figure 1.3b). The regression equation relating HCHO concentrations (in $mg \cdot m^{-3}$) and relative humidity is given by:

$$HCHO = 0.012\ e^{-0.33\ RH}$$

with a correlation coefficient of 0.750.

The relationship between air infiltration rate and relative humidity is shown in Figure 1.3c. The corresponding regression equation is:

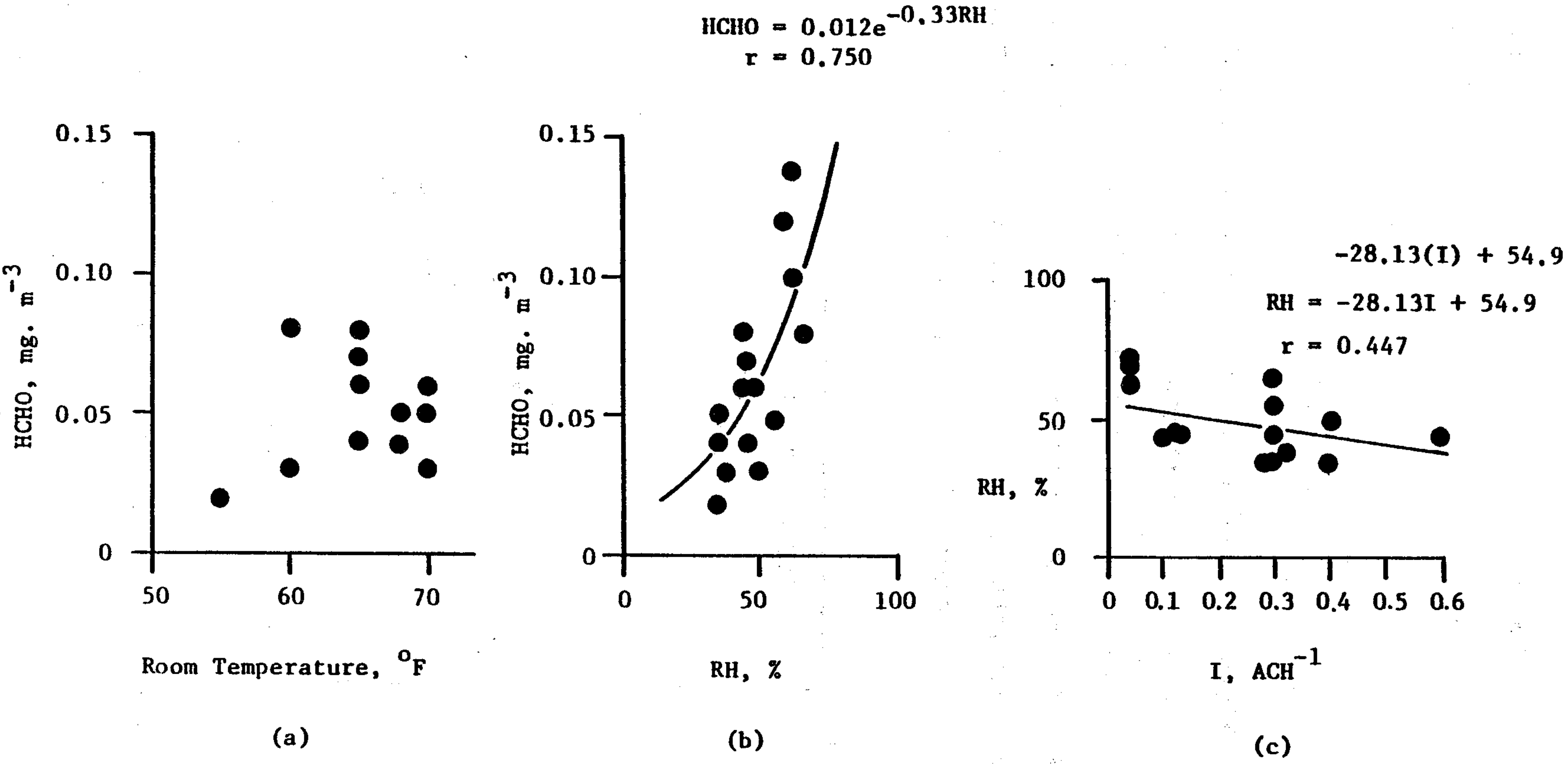
$$RH = -28.13\ I + 54.9$$

where relative humidity, RH, is expressed in percent and air infiltration rate, I, is expressed in air changes per hour. The correlation coefficient is -0.447. While the trends are evident, the relatively low correlation coefficient may be attributed to the small sample size and the clustering of the data points.

In the steady state, the HCHO concentration (C) is equal to the emanation rate (E) divided by the air infiltration rate (I).

$$C = \frac{E}{I}$$

The emanation rate, given in $mgm^{-3}hr^{-1}$, estimates the rate of release necessary to achieve the measured concentration. The emanation rate is dependent upon both relative humidity and air infiltration rate.



(a) (b) (c)

Figure 1.3 Variation of Formaldehyde with Relative Humidity

Figure 1.4 indicates that the relationship between formaldehyde levels and the ventilation rate can be approximated by the linear relationship:

$$\text{Log (HCHO)} = -0.5 \text{ Log I} - 1.62.$$

If the formaldehyde source was constant, the expected slope of this line would be -1. The fact that the observed slope is -0.5 indicates that an increased ventilation rate actually increases the rate of release of formaldehyde. Nevertheless, increased ventilation rates do lead consistently to lower formaldehyde concentrations. Therefore, increased ventilation rates can be an effective tool in reducing indoor formaldehyde levels.

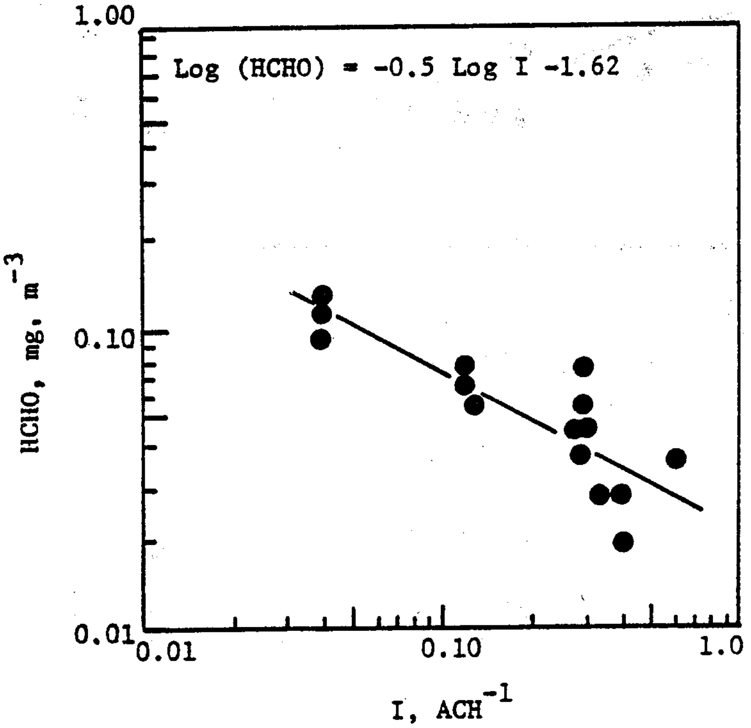


Figure 1.4 Variation of Formaldehyde Concentrations

Section 2.0

RADON

2.1 MEASUREMENT TECHNIQUES

Radon concentrations were monitored over a variety of environmental and operational settings from late August 1979 through April 1980. A coordinated system of measurements addressed the etiology of radon in the indoor environment through integrated, intermittant and continuous measurement of radon levels in key locations indoors and outdoors as explained below.

Week-long integrated values were secured by siting passive environmental radon monitors (PERMs) indoors on the main floor (living room) and in the basement. A third PERM was sited outdoors (carport) to assess ambient conditions. These thermoluminescent chip assemblies were shipped to LBL for analysis at the end of each period of exposure.

The integrated measurements were augmented by intermittant sampling, pumping sample air into Tedlar bags. These samples were then shipped overnight to LBL for analysis. Such grab samples were secured at the two indoor PERM sites as well as from soil gasses (~2 meters depth), the foundation wall cavity, and the basement floor drain. Tap water samples were secured on a monthly basis and shipped to LBL for analysis.

The Continuous Radon Monitor (CRM) developed by LBL was operating throughout much of the study term. This instrument gave radon concentrations every 3 hours for room air. Such sampling was periodically rotated between the basement and main floor.

2.2 DATA PRESENTATION

Analysis of the resulting data product was performed in response to two basic objectives:

- Characterize radon in this indoor environment
- Determine the capabilities of increased ventilation with air to air heat exchangers for reducing high levels of radon.

Each data product (i.e., integrated, intermittant, continuous) was analyzed separately in the light of environmental and ventilation conditions that prevailed. Conclusions were then drawn from each of these analysis products as well as from the integrated ensemble. Radon monitoring history for PERMs and grab samples is displayed in Figure 2.1.

2.2.1 PERMs Data

The data from indoor (basement, main floor) PERM concentrations are presented in Table 2-1. Outdoor readings have not been included because their values were consistently lower than corresponding indoor readings; the average outdoor Rn concentration was 0.4 $nCim^{-3}$. Because of variations in air infiltration rate during many of the 1 to 3 week averaging periods, a nominal air infiltration rate was developed by weighting each applicable air infiltration rate by duration into an average for each sample period. In some cases the air infiltration rate was inferred from knowledge of the operating state of the house and previous infiltration measurements performed under similar circumatances.

Basement Rn concentrations are consistently higher than those secured from the main (above ground) floor, indicating entry through (and

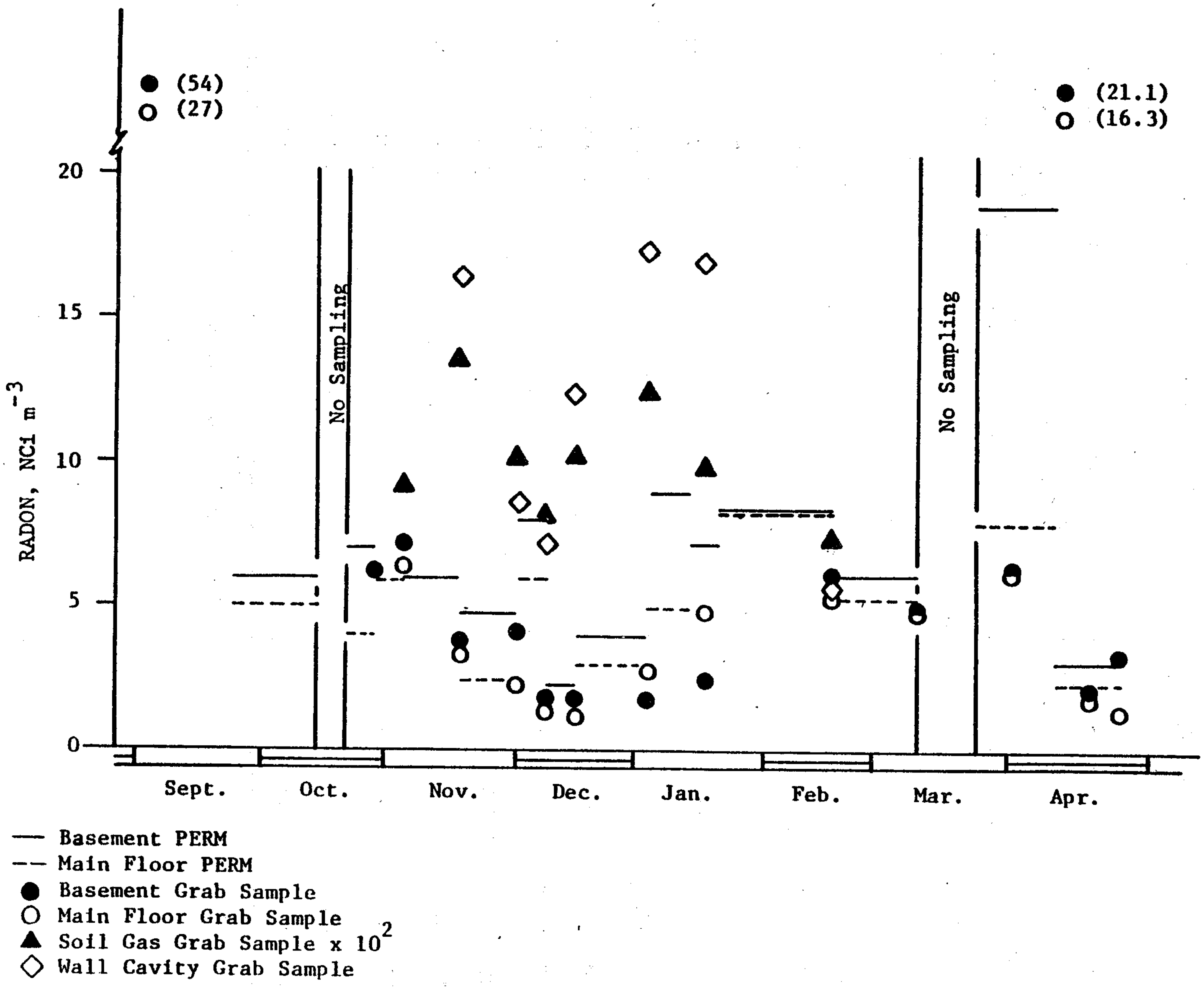


Figure 2.1 Monitoring History of Radon

Table 2.1. Integrated (PERM) Samples of House Air

Sample Period	Average Concentration (nCim^{-3})		Nominal Air Exchange Rate (ACH^{-1})
	Basement	Main Floor	
1) 09/23-10/15	6.0	5.0	0.21
2) 10/22-10/29	7.0	4.0	0.40
3) 10/29-11/05	--	6.0	0.47
4) 11/05-11/19	6.0	--	0.22
5) 11/19-12/03	4.6	2.5	0.45
6) 12/03-12/10	8.0	6.0	0.12
7) 12/10-12/17	2.3	1.6	0.60
8) 12/17-01/02	4.0	3.0	0.40
9) 01/02-01/14	9.0	5.0	0.30
10) 01/14-01/21	7.3	--	0.30
11) 01/21-02/18	8.6	8.5	0.30
12) 02/18-03/10	6.3	5.5	0.10*
13) 03/27-04/10	19.0	8.1	0.10*
14) 04/10-04/24	3.2	2.5	0.60*

* Infiltration rate inferred from operational state of house.

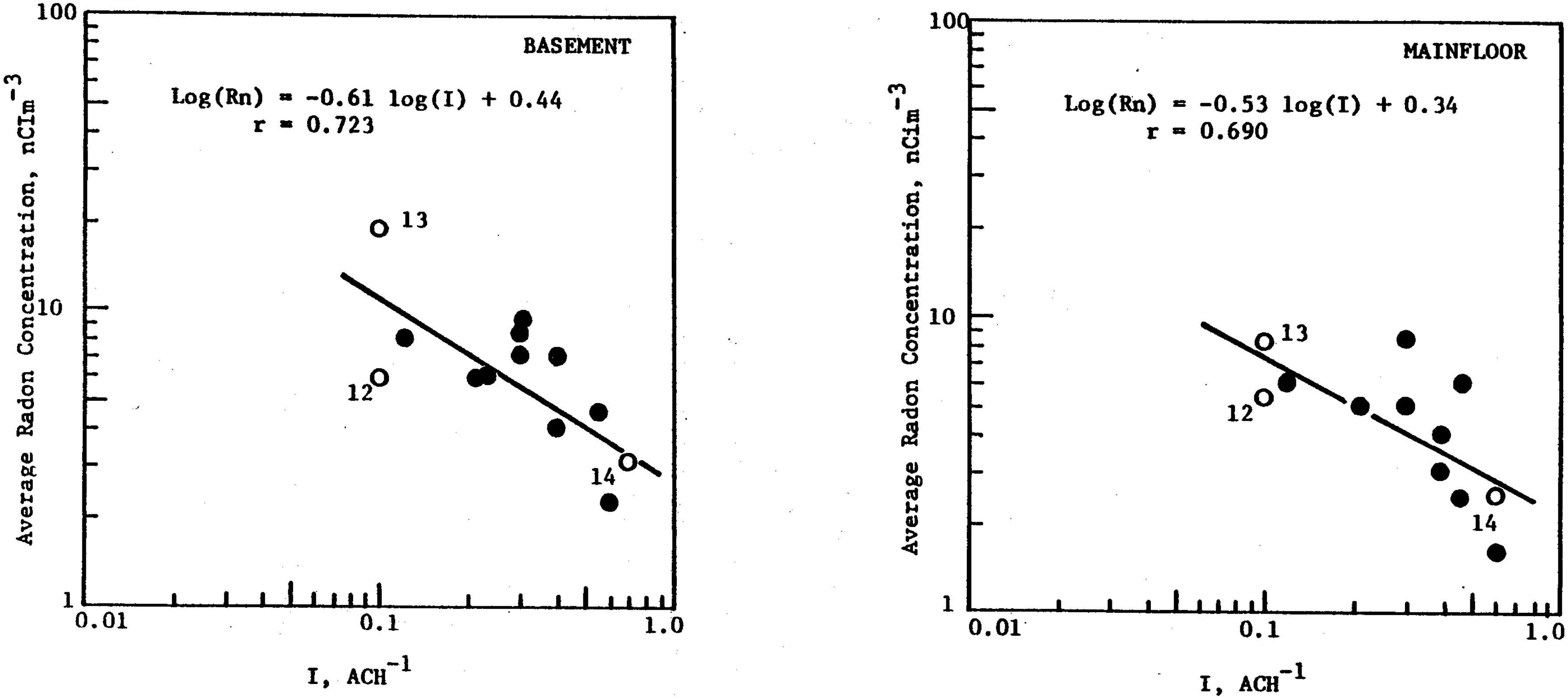
possibly from) the foundations of the house. This upstairs/downstairs contrast appears to be related to the ventilation/recirculation characteristics that prevailed during each sample period. Unfortunately, these operating characteristics were rarely constant within a given sample period. Most readings were obtained under varying combinations of internal recirculation of house air, mechanical ventilation and natural infiltration.

PERM sample set 12 coincided with operation of the internal recirculation fan only. Sample set 13 occurred with the ventilation/recirculation systems idle. Sample set 14 occurred with the heat exchanger processing basement air only. Sample set 7 was secured under conditions like those prevailing in sample set 14.

The value of introducing (outdoor) dilution air to the house system is obvious. The highest levels of radon concentrations--basement and main floor --occurred when the house was ventilated by infiltration only (sample set 13). This case also represented the highest upstairs/downstairs contrast. Recirculating house air with the aid of the interior fan lowered both the contrast and the concentrations substantially (sample set 12). Increasing the dilution air through the air-to-air heat exchanger in the basement further reduced the Rn concentrations (sample set 14 and 7).

The value of introducing fresh air to the house system is displayed in Figure 2-2. Depletion of radon concentration through increased air exchange is consistent for both the main floor and basement air spaces. The basement data yielded the regression equation:

$$\log [Rn] = -0.61 \log(I) + 0.44$$



(a) (b)

Figure 2.2 Radon Concentration and Infiltration where radon, (Rn) is expressed in nCi m^{-3} and air infiltration rate (I) is expressed in air changes per hour. A similar regression equation emerged from the main floor data:

$$\log[Rn] = -0.53 \log(I) + 0.34.$$

Sample sets 12, 13 and 14 have been highlighted on the graph to illustrate the relationships explained above.

2.2.2 Grab Samples of Room Air

The raw data from grab sampling of basement and main floor house air is presented in Table 2.2. The broad range of values registered here may be indicative of the short term variability of radon concentrations in the indoor environment. The air infiltration rates quoted here denote the rate of the 24 hours preceeding each sample set. Five of these sample sets (2,3,4,5,6) were obtained on days when the air exchange rate was modified (by altering the heat exchanger setting) in the morning. This alteration usually preceeded sampling by roughly 5 hours. The five sample sets that sustained changes in air infiltration were excluded from the computations.

As with the PERMs data, reduction of radon concentration was strongly correlated with air infiltration rate. Figure 2.3 illustrates the relationships for the basement as well as the main floor, the correlation coefficients are -0.83 and -0.88 respectively.

The logarithmic regression equations are simplified to a common form,

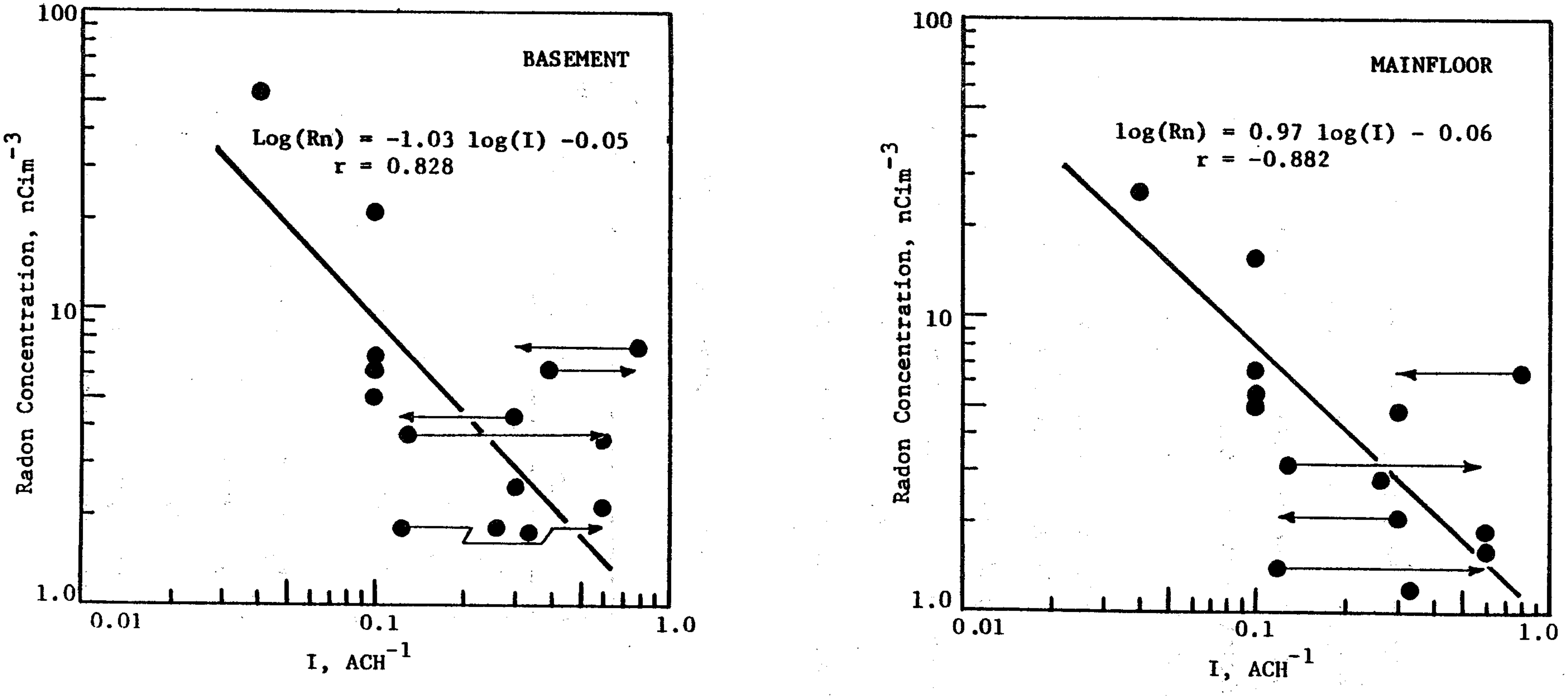
$$Rn = 0.88 \times I^{-1}$$

Table 2.2. Radon Grab Samples of House Air

Sample No.	Date	Time (EST)	Nominal Air Exchange (ACH-1)	Basement nCim^{-3}	Main Floor nCim-3
1	09/04/79	1300	0.04	54.0	27.0
2	10/29/79	1500	0.4,0.8*	6.3	--
3	11/05/79	1430	0.8, 0.3*	7.3	6.5
4	11/19/79	1500	0.13, 0.6*	3.8	3.3
5	12/03/79	1600	0.3, 0.12*	4.2	2.2
6	12/10/79	1430	0.12, 0.6*	1.8	1.4
7	12/17/79	1500	0.34	1.7	1.2
8	01/02/80	1500	0.26	1.8	2.8
9	01/16/80	1100	0.30	2.5	4.8
10	02/18/80	1130	0.10**	6.3	5.4
11	03/10/80	1100	0.10**	5.0	5.0
12	04/02/80	1030	0.10**	6.5	6.6
13	04/10/80	1000	0.10**	21.1	16.3
14	04/19/80	0930	0.60**	2.2	1.9
15	04/24/80	1100	0.60**	3.5	1.6

* Air exchange rate altered @ ~1000 EST that day.

** Air exchange rate inferred from operational state of house.



(a)

(b)

Figure 2.3 Radon Grab Samples and Infiltration
Arrows indicate shift in air infiltration rate 5 hours prior to sampling,

which may be applied to either air space. The difference between the regressions indicated for grab samples and for the PERMs data (Section 2.1.1) is attributable to the different time scales involved with the measurements. The multiday exposure interval for the PERMs allows "smoothing" of the short term (i.e., turbulent) processes that may occur with grab sampling.

2.2.3 Grab Samples of Soil Gases, Foundation Cavities and Tap Water

Data from the grab sampling of soil gases (~2 meters depth) and foundation wall cavity are presented in Table 2.3. Radon concentrations in the soil gases were commonly ten times those in the foundation wall cavity. The wall cavity readings were on the same order of magnitude as the house air samples (see section 2.1.2).

Table 2.3. Radon Grab Samples - Soil, Gas, Wall Cavity

Date	Time (EST)	Soil Gas ($nCim^{-3}$)	Wall Cavity ($nCim^{-3}$)
1) 10/29	1500	918	--
2) 11/19	1500	1340	16.5
3) 12/03	1600	1020	8.4
4) 12/10	1430	800	7.1
5) 12/17	1500	1030	12.4
6) 1/02	1500	1260	17.3
7) 1/16	1100	980	17.0
8) 2/18	1130	760	5.6

As shown in Figure 2.4a, radon levels in the wall cavity were closely related to levels in the soil gases. The regression equation:

$$\text{Log}\,[w] = 1.87 \log [s] - 4.6,$$

where w, the wall cavity radon concentration and s, the soil gas radon concentration are expressed in nCim^{-3} gave a correlation coefficient of 0.85, denoting a 0.72 explained variance.

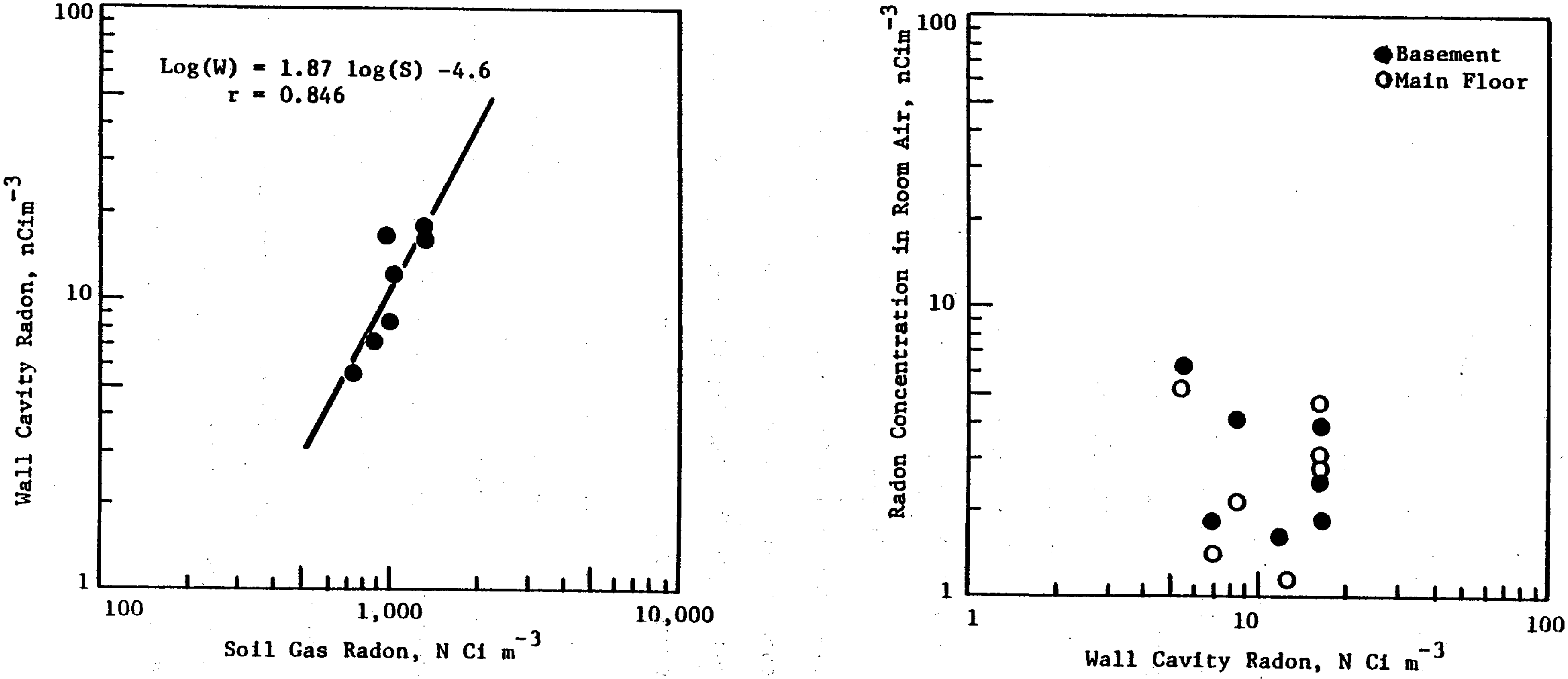
No correlations were found between this system and house air. This is not surprising because the permeation/infiltration of radon through the soil/foundation system should fluctuate over much slower time scales than would house air (see Figure 2.4b).

The grab samples of tap water and air from the floor drain in the basement were too few to support statistical analysis. Concentrations of radon in tap water were commensurate with those found in soil gases ($\sim 10^3$ nCim^{-3}). The two floor drain samples were slightly higher than the corresponding wall cavity samples.

2.2.4 Continuous Monitoring

The Continuous Radon Monitor (CRM), developed by LBL was in service from early December until late April. Sampling was alternated between the basement and main floor on a regular basis. A small portion of this record has been excerpted to illustrate the short term variability of indoor radon alluded to earlier (Section 2.1.2).

Figure 2.5 shows the continuous record of radon levels measured in the basement from 1/2/80 to 1/14/80. This coincided with PERM sample set 9 (section 2.1.1). During this period, the house was idle - the nominal



(a)

(b)

Figure 2.4 Radon in the Soil and the Foundation Wall

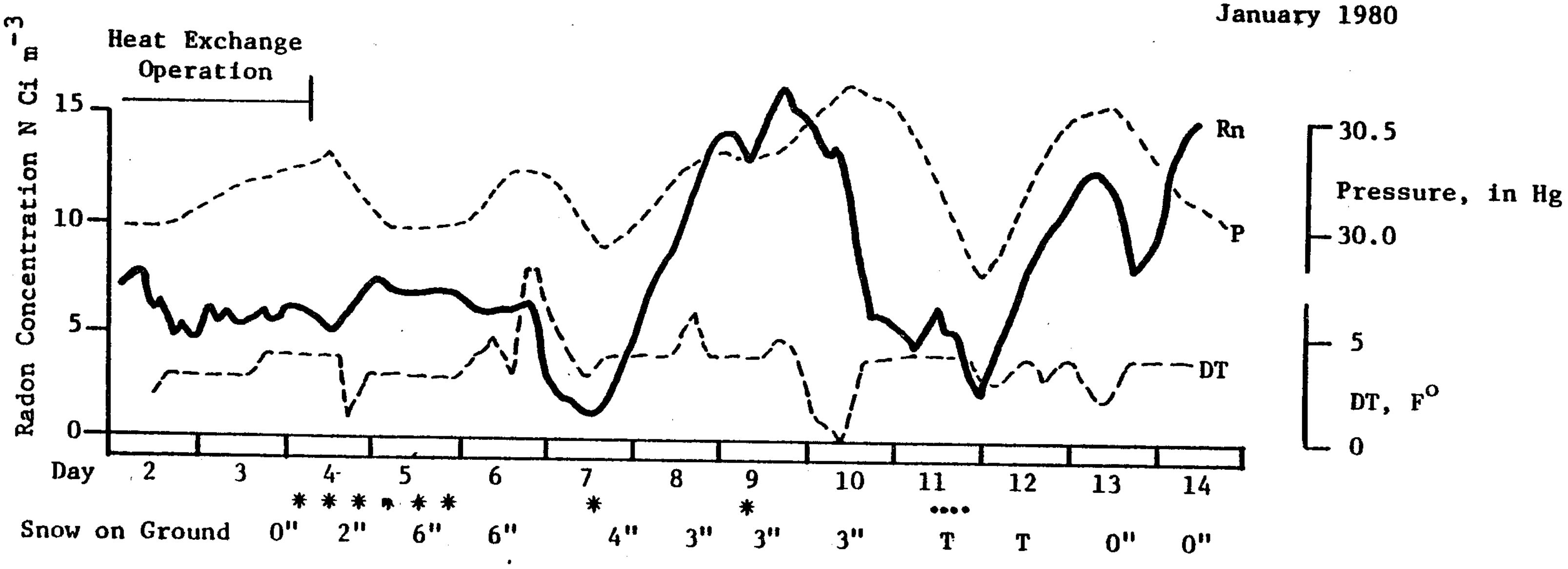


Figure 2.5 Variability of Radon in the Basement

air exchange rate of 0.3 ACH^{-1} was due primarily to natural infiltration. Infiltration was augmented by operation of the heat exchanger on the 2nd, 3rd and 4th of January, boosting the air exchange rate to 0.33 ACH^{-1} on those days. Barometric pressure (inches of mercury, corrected to sea level) and temperature differential (living room minus basement, F°) are also plotted. The occurrence of precipitation in the form of snow (*) and rain (●) is included along with indications of snow depth at 1830 EST. The pressure data was secured from the microbarograph operated by the University of Maryland in College Park (~45 km SE of Mt. Airy). Weather data came from a cooperative observer in Damascus, Maryland (~12 km SW of Mt. Airy) and is courtesy of the state climatologist.

The PERM sample set corresponding to this portion (1/2/80 to 1/14/80) of the CRM record indicated 9 $nCim^{-3}$ average basement concentration, 5 $nCim^{-3}$ average main floor concentration. The CRM indicates excursions between lowest values near 1.0 $nCim^{-3}$ and highest value in excess of 16.0 $nCim^{-3}$. The average of all CRM (3-hour) Rn concentrations during this period was 7.8 $nCim^{-3}$ which is in measurable agreement with the corresponding PERM sample from the basement.

The following points bear mentioning regarding an apparent broad correspondence between Rn concentration barometric pressure, precipitation and indoor temperature differentiation.

- When the heat exchanger and recirculation systems are not operating, radon growth and decline tends to be in phase with barometric pressure trends when there is snow on the ground.

- Rapid growth/decline of radon concentration appear to often coincide with variation in temperature difference between main floor and basement. Should this premise be operative intrastructure air movement due to temperature differentiation is an important factor affecting Rn concentrations.

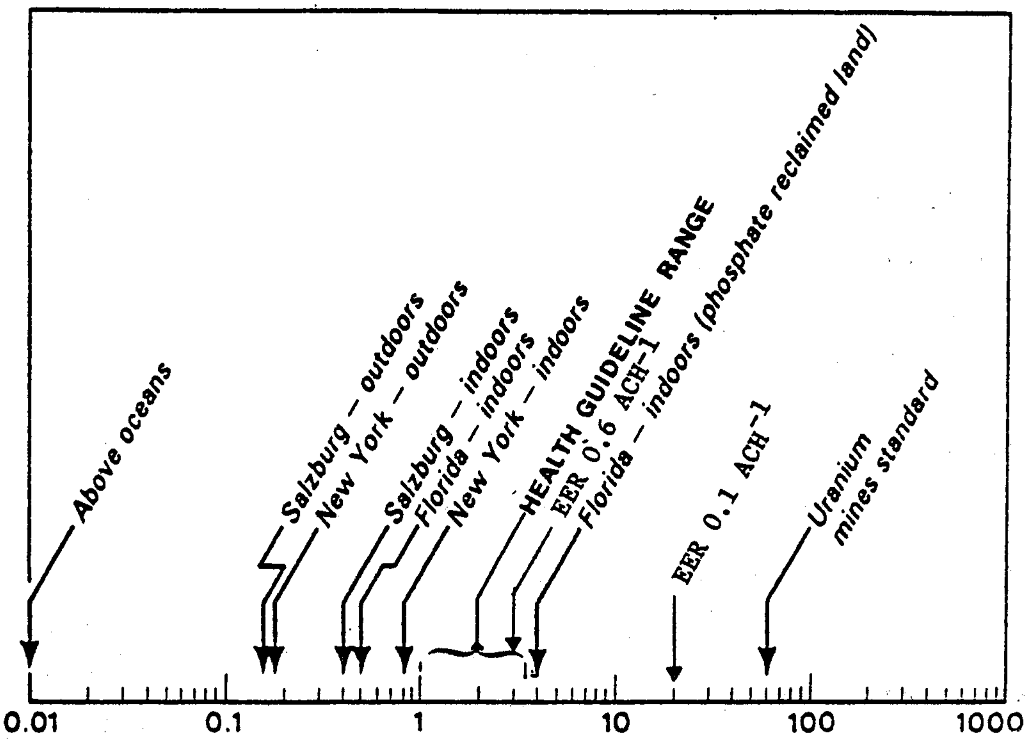
Two mechanisms appear to be at work. Barometric pressure would appear to have the lead role in moderating the potential magnitude of radon infiltration from the soil matrix surrounding the foundation, and transport of air between floors accounts for radon depletion that is too rapid for air exchange dilution. Unfortunately, the CRM was confined to sample in only one indoor location at a time. The expected symetry of growth/decline as air is transfered between floors cannot be verified without paired measurements.

Section 3.0

CONCLUSIONS

One-fifth of the measured formaldehyde concentrations were in the range that may cause health concerns. Although indoor temperature and relative humidity affect indoor HCHO concentration, the elevated formaldehyde concentrations were measured under very low air infiltration rates. The data show that ventilation of the indoor air space is somewhat effective in reducing high HCHO concentrations. The operation of the heat exchanger led to an increase of the air infiltration rate which in turn resulted in substantial reduction of formaldehyde concentrations.

A considerable number of the collected samples of indoor air displayed radon concentrations at levels higher than 1.0 - 4.0 $nCim^{-3}$ (assuming an equilibrium factor of 0.5, these radon levels would correspond to working levels above the health guidelines suggested by the U.S. EPA for homes in Florida built on land reclaimed from phosphate mining). As in the case of indoor formaldehyde concentrations, elevated indoor concentrations are substantially reduced when the infiltration rate is increased. The data base shows that the use of the air to air heat exchanger leads to reduction of indoor radon concentration by increasing the residential ventilation rate. The effect of air infiltration rate and the relative position of the EER observed radon concentrations (obtained by PERM measurements) against measurements taken elsewhere (LBL, 1980b) is illustrated in the figure below.



Radon concentration (nCi/m^3)

(after LBL, 1979b)

It is apparent that at low infiltration levels, the Rn and HCHO concentrations are above health recommendation levels, however, increasing the mechanical ventilation brings those levels within the recommended range.

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